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## Wednesday Morning

## Symposium PL

## **Plenary Lectures**

## Wednesday 9:00 Conference Auditorium / Plenary Lectures From individual trajectories to collective motion in suspensions of E-coli bacteria

#### Anke Lindner

Laboratoire de Physique et Mécanique des Milieux Hétérogènes, CNRS, ESPCI PSL, Sorbonne Université, Université Paris Cité, Paris, Île-de-France 75005, France

Active fluids consist of self-propelled particles (as bacteria or artificial microswimmers) and display properties that differ strongly from their passive counterparts. Unique physical phenomena, as enhanced Brownian diffusivity, viscosity reduction, active transport and mixing or the extraction of work from chaotic motion, result from the activity of the particles, locally injecting energy into the system. The presence of living and cooperative species may also induce collective motion and organization at the mesoscopic or macroscopic level impacting the constitutive relationships in the semi-dilute or dense regimes. Individual bacteria transported in viscous flows, show complex interactions with flows and bounding surfaces resulting from their complex shape as well as their activity. Understanding these transport dynamics is crucial, as they impact soil contamination, transport in biological conducts or catheters, and constitute thus a serious health thread. Here we investigate the trajectories of individual E-coli bacteria in confined and complex geometries with and without flow, using microfluidic model systems in combination with a novel Lagrangian 3D tracking method. Combining experimental observations and modelling we elucidate the origin of upstream swimming, lateral drift, persistent transport along edges as well as bacteria accumulation in specific geometries. Increasing bacteria concentrations collective motion emerges, where typical lengthscales can be identified in the velocity correlations. Using PIV measurements, we characterize the emerging vortex like structures as a function of confinement and we discuss how the corresponding lengthscale can be controlled though bounding walls or flows. The understanding gained can be for example used to control bacteria transport in complex geometries or shed light on the role of emergent mescoscopic structures on the macroscopic properties of active suspensions.

## Symposium MR

## **Microrheology and Microfluidics**

#### Organizers: Sepideh Khodaparast, Anke Lindner and Monica Oliveira

Wednesday 10:30 LT 1 (Level 7) / Track 1

## Margination of blood cells

Christian Wagner

Experimental Physics, Saarland University, Saarbruecken, Saarland 66123, Germany

The term margination refers to the movement of white blood cells from the centre of blood vessels to the wall of the vessels. There is still a debate on how differences in the size and stiffness of cells affect this movement. We present experiments and simulations in a microfluidic device where we quantify the amount of margination as a function of cell stiffness. Numerical simulations and experiments are in good agreement and we find a surprising stable positions for the stiff cell in the four corners of the channel as well as in the middle of flow, at least at larger haematocrit and flow rate.

Wednesday 10:50 LT 1 (Level 7) / Track 1

**Rigid and flexible micro-disks under viscous flows.** <u>Andrea de la Sen</u>, Anke Lindner, and Olivia du Roure *PMMH. ESPCI. Paris. Île-de-France 75005. France* 

The dynamics of rigid and flexible micro-disks under viscous flows are of fundamental interest because of the analogy that can be established with that of Red Blood Cells (RBCs), and also of practical interest for applications such as drug delivery. With an experimental technique well established in our lab [1] we are able to produce micro-disks by the cross-linking of a solution of Polyethylene Glycol Diacrylate (PEGDA) and a photoinitiator (2-hydroxy-2-methylpropriophenone, PI) exposed to UV radiation. We select the size of the particles, which normally have diameters of the order of a hundred microns, and a thickness of about 20 microns. We can also design their morphology, and we are able to create shapes like rings, triangular grids or hexagonal grids, to name a few, which will have smaller bending rigidities as compared to that of a full disk,

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1

being therefore more flexible. Using a micromanipulator, we are able to grab the micro-disks and bring them into micro-channels with welldefined initial conditions. We study their dynamics under elongational and shear flows, and compare them to the well-known Jeffery orbits of rigid disks [2]. We expect these orbits to be modified in the presence of deformability or non-Newtonian carrier fluids. Ultimately we hope our simple model system will be able to shed light on some of the dynamics observed for RBCs in complex flows.

[1] D. Dendukuri et al., Nature Materials, 5, 365 (2006). [2] G. B. Jeffery, Proceedings of the Royal Society of London, 102, 161 (1922).

## Wednesday 11:10 LT 1 (Level 7) / Track 1

# Brain rheology: Exploring extensional characteristics of brain-mimicking ultrasoft gels in two-phase microfluidic flow

## Ashhar Hussain and Heon Sang Lee

Department of Chemical Engineering, Dong-A University, Busan 49315, Republic of Korea

Deformation of biological cells and soft tissues is often considered linearly elastic; however, the assumption is only valid in a very limited stress range and often leads to significant errors in mechanical evaluation. We demonstrated the hyper-elastic behavior of ultra-soft poly(N-isopropyl acrylamide) (PNIPAm) microgels (USPNMs) in a converging channel flow, as a representation for brain tissues and similar biological samples. While larger particles are prone to gravitational sagging, employing micro-sized particles in microchannels provide accurate rheological evaluation by mitigating this challenge. The hyper-elasticity of USPNMs in response to a broad range of deformation was characterized at the centerline of the converging flow, where the elongational stress increases with change in velocity due to Bernoulli's principle. We introduced a carrier fluid consisting of baby hydrogels (avg. diameter, 10 µm) and oil that carried the hydrophilic USPNM sample (avg. diameter, 100 µm) on the centerline of oil background fluid. By "baby hydrogel," we mean small PNIPAm particles obtained during USPNM synthesis, using which, enabled settling-free flow, prevented wall contact, and enhanced carrier fluid viscosity for increased stresses at lower flowrates. Furthermore, drastic reduction of interfacial tension was observed in the converging area due to contact of baby gels with USPNM particles in the carrier fluid. The shear and elongational stresses were balanced with the elastic stress and interfacial Laplace pressure. As a result, we obtained a stress-strain curve from the microscopic images during flow. The non-linear stress-strain curve was characterized by conventional hyper-elastic models. The elastic modulus of the synthesized USPNM was 24 Pa, which is as low as animal brain tissue. This method holds great potential for evaluating rheology of brain and other soft tissues, blood rheology, drug delivery mechanics as well as in disease diagnoses and forensic.

## Wednesday 11:30 LT 1 (Level 7) / Track 1

Microfluidic rheology of vesicle prototissues

Majid Layachi, Remi Merindol, and Laura Casanellas

Laboratoire Charles Coulomb, Montpellier, France

The aim of this work is to provide a rheological description of biomimetic tissues in microfluidic confinement, as a simplified model for the flow of cellular tissues observed in complex physiological problems such as in embryogenesis or tumour metastasis. We designed a microfluidic chip that allowed us to tune the assembly of GUVs and to synthesize prototissues with controlled shape and size. GUV-GUV adhesion was mediated by the inclusion of complementary DNA strands enabling the control of prototissue cohesivity and the quantification of GUV-GUV adhesion by fluorescence microscopy. Rheological properties of prototissues were probed in a "pipette-aspiration" inspired microfluidic device, under controlled pressure conditions (creep test). We reported a viscoelastic response, which was well captured by a generalized Kelvin-Voigt fluid model. The prototissue rheology was complemented with a velocimetry analysis which allowed us to identify spatial reorganizations taking place within the prototissue, and with the quantification of the deformation field at the scale of individual vesicles.

Wednesday 11:50 LT 1 (Level 7) / Track 1 Keynote

## Microfluidic model of micro-haemodynamics

Qi Chen<sup>1</sup>, Eleanor Doman<sup>2</sup>, Oliver Jensen<sup>2</sup>, Igor Chernyavsky<sup>2</sup>, and <u>Anne Juel<sup>1</sup></u>

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The human placenta relies on well-orchestrated haemodynamics to deliver its multiple functions. Its geometrical complexity and lack of appropriate animal models mean that laboratory models offer a powerful tool to investigate haemodynamics and haemorheology in the human placenta and other complex biological tissues. We develop a model of red blood cells (RBCs) with polydimethylsiloxane capsules of adjustable diameter and membrane thickness, which are microfabricated using a 3D nested glass capillary device. The elastic modulus of the membrane can be varied systematically by an order of magnitude by adjusting the chemistry, and the capsules are further deflated by osmosis to match the relative surface-area-to-volume ratio of real RBCs. We start by testing the capacity of our capsules to mimic the motion and large deformations of RBCs in simple conduits. We then explore capsule suspension flows in porous media of increasing complexity. We aim to relate the global resistance of our porous medium to the local distribution of particles and their velocity as a function of the disorder and porosity of the medium, volume fraction and capillary number Ca, the ratio of viscous to elastic forces. We find that in a uniformly random array, the interaction between suspension and geometry results in a non-linear variation of the relative flow resistance as a function of both volume fraction and Ca, which is associated with the formation of networks of preferential paths.

MR4

MR5

MR3

## Symposium PF

## **Polymeric Fluids**

Organizers: Francesco Del Giudice, Richard Graham and Evelyne van Ruymbeke

### Wednesday 10:30 LT 17 (Level 7) / Track 2 Monitoring isothermal crystallization of poly(butylene succinate) (PBS) using rheo-dielectric measurements Hyeong Yong Song and Manfred Wilhelm

Institute for Technical Chemistry and Polymer Chemistry, Karlsruhe Institute of Technology, Karlsruhe 76131, Germany

Recently, aliphatic polyesters have received much attention regarding their biodegradability, particularly from an ecological point of view. Poly(butylene succinate) (PBS) is one of the interesting biodegradable polymers. It has been utilized in packaging film, bags, flushable hygienic products, and mulching film. PBS exhibits excellent characteristics including susceptibility to biodegradation, thermoplastic behavior, reasonably balanced thermo-mechanical properties, and processibility with conventional equipment for plastics. Mechanical and processing properties of PBS are affected by crystallinity and crystallization rate as well as amorphous chain characteristics. For instance, high crystallinity of PBS slows down degradation rate and deteriorates barrier properties. Thus, it is important to understand crystallization kinetics of PBS for processing aspect. To this end, this study utilizes a rheo-dielectric set-up for in-situ measurements of isothermal crystallization. By doing so, rheological modulus developments can be recorded simultaneously with dielectric permittivity changes. We present how to obtain consistent crystallinity results from dielectric properties. Finally, by comparing rheological data with dielectric spectroscopic data, we investigate the kinetics of early-stage crystallization process of PBS.

#### Wednesday 10:50 LT 17 (Level 7) / Track 2

Unveiling the restricted mobility of carbon nanotubes inside a long chain branched polymer matrix via measuring the rheological behavior and conductivity under shear flows

Jixiang Li<sup>1</sup>, Abderrahim Maazouz<sup>2</sup>, and Khalid Lamnawar<sup>3</sup>

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The present work aimed to gain deep understanding about the effects of long-chain branching (LCB) on the rheological and electrical behaviors of nanocomposites. Accordingly, measurement consists of fast shear steps (transient start up shear rheology) coupled with small amplitude oscillatory sweep (SAOS) and dielectric test was designed. Linear polypropylene (PPC) and branched polypropylene (PPH) were chosen as the polymer matrix and carbon nanotubes (CNTs) as the nanofillers. The percolation threshold concentration of LCB PPH nanocomposites was higher than linear PPC, which was due to the high viscosity and elasticity of LCB PPH. The contribution of the filler network to the apparent rheological properties became less significant. A transient shear with different shear rates was imposed on the composites after which SAOS and electrical conductivity measurements were conducted. The liquid-solid transitions of the nanocomposites were different and were found to be dependent on the shear flow conditions (shear rate). For the linear PPC, higher shear rates caused the filler network to break down while lower shear rates helped the nanofillers to agglomerate. Interestingly, for LCB PPH, both higher and lower pre-shear rates resulted in the breakup of the filler networks, which possibly due to the restricted mobility of the CNTs by the LCBs. The confinement of the polymer chains to the CNTs and their aggregates made it difficult for the fillers to move and align thus causing the formed network to be easily destroyed even under slow and slight shears. Similarly, the same trend was found after shear flows reflected by the increase and decrease of electrical conductivities. The restricted mobility of CNTs inside a LCB matrix was also confirmed by the inapparent increase of conductivity during thermal annealing compared with inside a linear matrix and the discrepancy of conductivity change during the melt to solid cooling process.

#### Wednesday 11:10 LT 17 (Level 7) / Track 2

PF3

## Distribution of Mechanical Properties in Poly(ethylene oxide) / silica Nanocomposites under Deformation: From the Glassy towards the Melt Regime

HILAL REDA1 and Vangelis Harmandaris2

<sup>1</sup>The Cyprus institute, Nicosia 2121, Cyprus; <sup>2</sup>The Cyprus institute, Nicosia 2121, Cyprus

Polymer nanocomposites exhibit a heterogeneous mechanical behavior that depends strongly on the interaction between the polymer matrix and the nanofiller. Here we provide a detailed investigation of the mechanical response of model polymer nanocomposites under deformation, across a range of temperatures from the glassy towards the melt regime, via atomistic molecular dynamics simulations. We study poly(ethylene oxide) matrix with silica nanoparticles (PEO/SiO2) as a model polymer nanocomposite system with attractive polymer/nanofiller interactions. We focus on factors governing the mechanical response of the PEO/SiO2 systems by investigating the distribution of the (local) mechanical properties, focusing in the interphase and matrix regions, across a wide range of temperatures below and above the glass transition temperature. We also shed light on characteristic structural and dynamic attributes during deformation. As expected upon heating the system, a decrease in the Young's modulus is observed, accompanied by an increase in the Poisson's ratio. The observed differences in rigidity between the interphase and the matrix region decrease as temperature rises; at temperatures well above the glass transition temperature, the rigidity of the interphase approaches the one

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of the matrix behavior. To describe nonlinear viscoelastic behavior of the polymer chains, the elastic modulus of the PEO/SiO2 systems is further calculated as as function of the strain for the entire nanocomposite s well as the polymer/nanoparticle interphase and matrix regions. The elastic modulus drops dramatically with the increase of strain for PNC, bulk and local regions, especially at small deformations to reach a plateau. The Payne effect for local regions(interphase and matrix) is less dominate.

## Wednesday 11:30 LT 17 (Level 7) / Track 2

PF4

## Controlling the flow behavior of ABA triblock copolymer-based networks by microphase separation

Clément Coutouly, Evelyne van Ruymbeke, and Charles-André Fustin

IMCN, BSMA, Université catholique de Louvain, Louvain-la-Neuve, Belgium

ABA triblock copolymers are interesting materials because of their ability to form microphase separated structures where the B blocks form bridges between A domains, leading to reversible networks. Due to this property, triblock copolymers have been considered for a variety of applications, especially in the bulk state, such as pressure sensitive adhesives or thermoplastic elastomers. However, a major drawback of these systems is their rapid loss of mechanical properties at high temperatures, limiting their range of applications. A potential way to circumvent this limitation would be to design ABA triblock copolymers that keep, or even reinforce, their microphase separation at high temperatures. We report here on two such systems based on ABA triblock copolymers having a low Tg Poly(n-butyl acrylate) central block and either rigid or soft A blocks. These materials have been characterized by linear shear rheology, DMA and SAXS to establish structure-properties relationships. The first system uses Polystyrene as rigid outer blocks and exhibits, if the composition is properly adjusted, a lower disorder-to-order temperature behavior increasing the microphase separation upon a temperature increase. Contrary to triblock copolymers with a classical upper order-to-disorder temperature behavior, the flow regime is not reached, even at temperatures well above the glass transition of the PS. Instead, a low frequency plateau is observed in rheology, showing the increased lifetime of the microphase separated PS domains. The second system is based on the low Tg poly(heptafluorobutyl acrylate) (PHFBA) as soft outer blocks. These triblocks show strong microphase separation, even for very short PHFBA blocks, from room to high temperature, conferring high structural and thermal stability to the material. As for the PS-based triblocks, rheology revealed the appearance of a second plateau in the low-frequency region, observable up to different temperatures that depend on the PHFBA block length.

## Wednesday 11:50 LT 17 (Level 7) / Track 2

PF5

# Phase separation in Styrene Butadiene block copolymer solutions: effects of solvent quality and polymer polydispersity

<u>Gaia Paradiso</u><sup>1</sup>, Marta Gamberoni<sup>1</sup>, Salvatore Coppola<sup>1</sup>, Irene Perna<sup>2</sup>, Rosalia Ferraro<sup>2</sup>, and Sergio Caserta<sup>2</sup> <sup>1</sup>Versalis SpA, Ravenna, Ravenna 48123, Italy; <sup>2</sup>Department of Chemical Engineering, University of Naples Federico II, Naples,

## Naples 80125, Italy

Almost all applications of block copolymers involve mixtures, such as blends with a parent homopolymer, low molecular weight "tackifying" resins and plasticizers, or solvent. The behavior of block copolymers in solvent continues to be an active area of research [1]. A significant theoretical and experimental effort has been directed at the characterization and identification of the factors that determine the phase separation in polymeric solutions. In this work, we focus our attention on two different types of Styrene-Butadiene block copolymers, one possessing nearly monodisperse molar mass distribution and the other displaying polydisperse molar mass distribution and heterogenous chain composition. We investigated the interactions of these polymers with solvents of different quality and polarity. To this end, we subjected the polymer solutions to temperature sweep analysis to assess the relevant rheological changes related to phase separations. In parallel, we also conducted an experimental campaign based on microscopy analysis to highlight optical evidence of phase separation of aggregate structures with slower dynamics, which determines a greater viscosity of the solution. By macro-phase separation we mean the formation of adgregate structures with slower dynamics, which determines a greater viscosity of the solution. By macro-phase separation at different polymer concentrated solution. Typically, macro-phase separation resulted into a sort of emulsification, thus producing a decrease in viscosity. Our rheological results show how the copolymer type, and the solvent quality may influence the phase separation at different polymer concentrations. References: [1] Hanley, Kenneth J.; Lodge, Timothy P.; Huang, Ching-I. Phase behavior of a block copolymer in solvents of varying selectivity. Macromolecules, 2000, 33.16: 5918-5931.

## Wednesday 12:10 LT 17 (Level 7) / Track 2

PF6

# The linear and nonlinear rheological properties of Polylactic acid/Low-density polyethylene blends with different hydrophobicity inorganic fillers

Minchan Kim and Kyu Hyun

## School of Chemical Engineering, Pusan National University, Pusan, Republic of Korea

Among the biodegradable polymers, polylactic acid (PLA) is considered a promising material to solve the environmental pollution caused by plastic waste. However, PLA has weak mechanical properties such as brittleness and poor processability. For this reason, some researchers have tried polymer blend with materials that can compensate for the weaknesses of PLA1. In this study, low density polyethylene (LDPE) with strain hardening behavior in elongational viscosity was used as the dispersed phase to overcome the disadvantage of PLA. To improve compatibility between PLA and LDPE, we added inorganic fillers, hydrophilic fumed silica, hydrophobic fumed silica, hydrophilic organoclay, and hydrophobic organoclay as compatibilizers. To evaluate the compatibilizer effect of various fillers, linear viscoelastic properties from small amplitude oscillatory shear (SAOS) test and nonlinear viscoelastic properties from elongational test were measured and anlyzed. Rheological measurements were used by using a strain-controlled rheometer ARES-G2 (TA Istruments Inc). In case of frequency sweep, 25mm parallel plate geometry was

used at 180°C under the Linear viscoelastic region (LVER). For the extension viscosity anaylsis, we measured used extension viscosity fixture (EVF) geometry with ARES-G2 at 180°C. Elongational viscosities were analyzed by molecular stress function (MSF) model. In addition, we investigated morphology obtained by transmission electron microscopy (TEM) and scanning electron microscope (SEM) to check the location of filler and droplet size of dispersed phase. As a result, we confirmed the relation between rheological properties and morphology in PLA/LDPE blend systems.

## Symposium CG

## **Colloids and Glasses**

Organizers: Michel Cloitre, Johan Mattsson and George Petekidis

Wednesday 10:30 LT 6 (Level 8) / Track 3 Keynote Analysis of Thixotropic Timescale

## Yogesh M. Joshi

Department of Chemical Engineering, Indian Institute of Technology Kanpur, Kanpur Nagar, Uttar Pradesh 208016, India

Thixotropy is characterized by an increase in viscosity when a material is subjected to no flow (quiescent) or weak flow conditions and a decrease in viscosity when it is subjected to strong flow conditions. The characteristic timescale associated with the thixotropic phenomenon, particularly how the viscosity increases with time, has been termed the thixotropic timescale. In the literature, several approaches have been suggested for estimating the thixotropic timescale. The most prominent approach, however, infers it from a specific form of a kinetic expression for structure parameter evolution. In this paper, we study the various kinds of structural kinetic models, and by carrying out a careful analysis of the same, we propose a universal candidate parameter for the thixotropic timescale that is associated with the most generic form of the kinetic expression for structure parameter evolution. We observe that only when we consider the viscosity of the structural kinetic model undergoes continuous increase with time and eventually diverges under quiescent conditions, we can relate change in the thixotropic timescale to change in the strength of the thixotropic phenomenon. For this case, which we believe is the most practical scenario, our analysis suggests that increasing the thixotropic timescale based on the time derivative of the viscosity. The proposed definition allows a straightforward and unique way to determine thixotropic timescale through experiments and agrees well with the conventional notion of thixotropy.

## Wednesday 11:10 LT 6 (Level 8) / Track 3

Innovative conductive hydrogels for biomedical applications

Nicola Antonio Di Spirito, Nino Grizzuti, and Rossana Pasquino

University of Naples Federico II, Naples, Naples 80125, Italy

Electrically conductive hydrogels combine the electrical properties of conductive materials with the unique features of hydrogels. They are attractive for various biomedical applications due to their intelligent response to electrical fields, and can be used in tissue engineering, biosensors, drug delivery systems, biomedical devices, electro-assisted stimulation of mammalian cells and to power wearable or implantable systems. Owing to their distinctive properties, such as biocompatibility, thermosensitivity and self-assembling behavior, Pluronics can be adopted for the generation of innovative hydrogels for biomedical applications. In this work, the rheological, morphological, and structural properties of Pluronic F68 in the presence of poly(3,4-ethylenedioxythiophene):poly-(styrenesulfonate) (PEDOT:PSS) are studied to optimize the synthesis of novel biocompatible and highly conductive hydrogels. Aqueous solutions with 45 wt% Pluronic F68 and different PEDOT:PSS contents are investigated by means of experimental rheology and Small Angle X-Ray Scattering (SAXS), to unveil the influence of PEDOT:PSS on the Pluronic-based systems. Furthermore, Ag flakes are added to the aqueous solutions of Pluronic F68/PEDOT:PSS in order to further enhance the system conductivity. The presence of PEDOT:PSS and Ag flakes endows Pluronic solutions with high conductive properties, while preserving the characteristic self-assembly and gelation properties and microstructural evolution of Pluronic F68 in water. Here, innovative electrically conductive hydrogels for biomedical applications are developed, providing a full characterization of their macroscopic and microscopic properties thanks to the adoption of different and complementary experimental techniques.

## Wednesday 11:30 LT 6 (Level 8) / Track 3

## The central role of colloids to explain the crystallization dynamics of halide perovskites

Davide Amoroso<sup>1</sup>, Giuseppe Nasti<sup>2</sup>, Massimiliano M. Villone<sup>1</sup>, Tim Kodalle<sup>3</sup>, Carolin M. Sutter-Fella<sup>3</sup>, Pier Luca Maffettone<sup>1</sup>, and Antonio Abate<sup>1</sup>

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Perovskite materials have emerged as highly promising candidates for several applications due to their distinctive electronic, optical, and structural properties. In the solar cells sector, they are very attractive because of their low costs and excellent performance. Lead-based perovskites have been extensively researched for their superior electron mobility and carrier properties, even though lead causes environmental and health risks. Substituting lead with tin, a viable alternative from the same periodic table group, holds promise. Nevertheless, the elevated crystallization rates

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of tin-based perovskites make the control of the material's morphology challenging. In this study, we employ in situ characterization techniques to elucidate the intricate relationship between the properties of perovskite colloidal suspensions and the mechanisms governing the crystallization of tin perovskite films. Specifically, we investigate the solution engineering approach, wherein additives and solvents are employed to tune the properties of perovskite precursor suspensions. Their impact on the dynamics of film crystallization is rationalized by using principles derived from the DLVO theory with the objective of establishing a comprehensive understanding of how the solid formation process can be strategically controlled to obtain high-efficiency perovskite materials. In addition, Brownian Dynamics simulations are employed as a predictive tool to deepen the investigation of the intricate process of perovskite formation.

#### Wednesday 11:50 LT 6 (Level 8) / Track 3

Compression of capillary suspension networks with and without nanoparticles

Lingyue L. Liu<sup>1</sup>, Jens Allard<sup>2</sup>, and Erin Koos<sup>2</sup>

<sup>1</sup>Department of Chemical Engineering, KU Leuven, Leuven, Belgium; <sup>2</sup>Department of Chemical Engineering, KU Leuven, Leuven, Belgium

Capillary suspensions, suspensions with sample-spanning particle networks, are typically created by adding a small amount of immiscible secondary fluid to a suspension [1]. Using nanoparticles integrated in the bridging fluid, porous sintered ceramics with superior porosity and compressive strength can be produced [2]. These nanoparticles also have a striking effect on the rheological properties and structure of the paste, particularly in response to compression.

For the sample without nanoparticles, the volume fraction remains constant with compression and the bottom slides flex. This rigid network shows little change, increasing the coordination number from  $5.02 \pm 0.13$  to  $5.26 \pm 0.11$ . The rheological response is also strongly dependent on the normal force during gap setting on the rheometer with the storage modulus increasing by a factor of two with only 50 mN of force. For the sample with nanoparticles, the network is much more flexible and did not displace the lower plate of the shear cell. Instead, the volume fraction increased from 25 to 35 vol% and the coordination number increased from  $5.45 \pm 0.06$  to  $6.14 \pm 0.15$ . This network is also less sensitive to the normal force during gap setting and shows little influence of the particle hydrophobicity on the results. We hypothesize that the nanoparticles are able to induce the formation of thin liquid films on microparticle surfaces. These films are able to eliminate the contact line pinning effect of bridges and promote internal liquid exchange, thus, lubricating the movement of the microparticles and narrowing the bridge size distribution. The nanoparticles at the contact region between microparticles further reduce the Hertzian contact giving rise to the ability to rearrange.

[1] E. Koos and N. Willenbacher, Science. 331, 6019 (2011).

[2] M. Weiß, et al., Journal of the European Ceramic Society. 40, 8 (2020).

Wednesday 12:10 LT 6 (Level 8) / Track 3

### **Spreading of Glass-Forming Fluids**

Thomas Voigtmann and Linnea Heitmeier

Institute of Materials Physics in Space, German Aerospace Center DLR, Cologne 51147, Germany

Glass-forming fluids are highly viscoelastic, demonstrate shear thinning, and a re characterized by a dynamical yield stress. Hence their spreading is an intricate balance between hydrostatic pressure, surface tension, yield stress, and the effect of shear thinning. In this talk I discuss simulations and theoretical results for droplet spreading of glass-forming fluids. For the simulations we combine the mode-coupling theory of the glass transition, as a microscopic theory, with the finite-element method to perform macroscopic simulation with a microscopically justified constitutive equation. The model describes the slow evolution of internal stresses as the droplet spreads, and allows for the emergence of frozen-in residual stresses that persist after spreading has come to rest. We investigate the dependence of such residual stresses on the previous flow history.

## Symposium SG

## **Suspensions and Granular Materials**

Organizers: Erin Koos, Wilson Poon and Olivier Pouliquen

Wednesday 10:30 LT 18 (Level 8) / Track 4

Onsager-symmetric constitutive laws for 3D granular flow in the inertial regime

Thomas Barker<sup>1</sup>, Yuhao Hu<sup>2</sup>, and David Schaeffer<sup>3</sup>

<sup>1</sup>School of Mathematics, Cardiff University, Cardiff, Wales CF24 4AX, United Kingdom; <sup>2</sup>School of Mathematical Sciences, Shanghai Jiao Tong University, Shanghai, China; <sup>3</sup>Mathematics Department, Duke University, Durham, NC, United States

Many constitutive relations for inertial granular flow assume local proportionality of the shear stress and shear strain-rate tensors. However, even in simple shear, this approximation is inaccurate because normal stress differences exist. Here we derive new three-dimensional visco-plastic constitutive laws which address this discrepancy by imposing Onsager symmetry in combination with a truly 3D yield condition and inertialnumber-dependent planar rheology. In addition to reproducing experimental data of normal stress differences, the new relations also predict

CG4

CG5

nontrivial deviations, away from the predictions of 2D theories, for general triaxial deformations. Perhaps illustrating the consequences of imposing Onsager symmetry, which arises from thermodynamic considerations, is as important as the actual constitutive laws themselves.

#### Wednesday 10:50 LT 18 (Level 8) / Track 4

### Unsteady rheology of dense inertial granular flows

Wenwen Wang<sup>1</sup>, Jin Sun<sup>2</sup>, and Eric C. Breard<sup>3</sup>

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Dense granular flows are commonly observed in nature and industrial processes, such as debris and powder flows. Their solid-like to liquid-like behaviour can be distinguished by the inertial number *I*, defined as the ratio of the microscopic rearrangement time scale to that for macroscopic deformation. The steady-state rheology of liquid-like (inertial) flows is well described by the  $\mu(I)$  rheology, which defines the shear-stress-topressure ratio ( $\mu$ ) and the solid volume fraction as functions of inertial number (*I*). Applying  $\mu(I)$  rheology to unsteady flows would show that stresses respond instantaneously to changes in *I* and vice versa - the history effect is not considered. The question thus arises regarding whether the history effect is important for describing unsteady inertial flows. To answer the question, we study unsteady flows by performing discrete element simulations of simple shear of monodisperse spheres with a varying shear rate (step changes in either magnitude or direction) at a constant volume fraction. We found that  $\mu$  and *I* evolve over a significant strain of about 0.2 or 1 following the rate change in magnitude or direction, respectively, contrasting the constant values predicted by the  $\mu(I)$  rheology due to the constant volume fraction constraint. We attribute the mechanism of these history effects to the microstructure evolution, which is revealed and quantified by the coordination number and the fabric tensor defined using the topology of the nearest neighbours whose centres are located at 1.01 diameter from a particle centre. The microstructure evolves similarly to  $\mu$  and *I*, corroborating the microstructural mechanism. Our results therefore clearly demonstrate the need to take into account history effect for unsteady inertial flows in general. However, whether including the microstructural variables suffices to model such flows remains to be confirmed.

Wednesday 11:10 LT 18 (Level 8) / Track 4

#### The behaviour of rotating intruders in dense granular beds Robin Burton

#### School of Physics and Astronomy, University of Edinburgh, Edinburgh EH9 3FD, United Kingdom

We investigate, via simulation and experiment, the forces felt by a cylindrical intruder pushed vertically into a dense granular bed. When the intruder rotates about its axis perpendicular to gravity it experiences a reduction in the vertical force resisting penetration. This reduction is positively correlated with the rotation speed. To understand how this arises we study the relationship between the competing forces of drag, lift, gravity and torque on the intruder. Unlike movement through liquids or air our understanding of locomotion in granular media is still quite limited, and as well as being of scientific interest, it is hoped that an increased understanding in this area will help our industrial partner Crover to enhance the performance of their granular drone.

#### Wednesday 11:30 LT 18 (Level 8) / Track 4

#### Flow of a granular material through a forest of obstacles Baptiste Darbois Texier

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The inclined plane configuration is ideal for studying the rheology of granular materials under imposed-pressure conditions. We investigate this situation when a forest of obstacles is present in the path of the grains and thus affects the global flow properties of the material. Our experimental set-up consists of a plane which includes a forest of cylindrical pillars planted perpendicular to the plane and positioned at regular intervals. We quantified the influence of the inter-pillar distance on the stationary flow rate of the grains for different layer thicknesses and different angles of inclination. We observe that when the inter-pillar distance is reduced, the flow rate of the grains decreases and reaches a new regime where the flow rate is practically independent of the thickness of the grain layer. In order to rationalize these observations, we develop a model that is based on a depth-averaged approximation associated with the  $\mu$ (I)-rheology and that considers the average force exerted by the pillars on the granular layer. This approach yields an effective rheology of the system composed by grains and obstacles and that depends on the ratio between the distance between the pillars and their diameter.

#### Wednesday 11:50 LT 18 (Level 8) / Track 4

#### Effect of nanocomposites on crystallization of waxy crude oils

Nezia Rosso, Roberta Gimenes, Julio A. Abdala, G Muhlstedt, and <u>Cezar Negrao</u> Academic Department of Mechanics, Federal University of Technology - Paraná - UTFPR, Curitiba, Parana 81280-340, Brazil

Oil gelation is a significant problem in producing and transporting waxy crude oils as wax crystallization occurs at low temperatures. High pressures required to restart the oil flow in subsea pipelines can be much larger than the usual operational pressure, as the temperature in such an environment can be as low as 4 °C. The literature has shown that adding nanoparticles can reduce WAT (Wax Appearance Temperature) and, consequently, oil gelation. The current work investigates the effect of adding graphene, graphene oxide, and reduced graphene oxide on the WAT, crystal size and morphology, and rheological properties of a model waxy crude oil. The oil was formulated by adding wax paraffin (Sigma Aldrich 327212 CAS-No: 8002-74-2) in mineral oil (Sigma Aldrich 330779 CAS-No: 8042-47-5). We have used Optical Polarized Microscopy, Scanning

SG2

SG3

SG4

Electron Microscopy, Differential Scanning Calorimetry, and Rheometric tests for the investigation. We anticipate that not only the structure of wax crystals but also the WAT and rheological properties of the gelled waxy crude oil are significantly affected by the addition of graphene nanocomposites.

## Wednesday 12:10 LT 18 (Level 8) / Track 4 **Rheological modelling of elongated particles**

Nathan R. Berry<sup>1</sup>, Sina Haeri<sup>1</sup>, and Jin Sun<sup>2</sup>

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Flows of elongated particulate granular systems are encountered in many industrial and natural settings. However, in comparison to spheres, the rheological properties of such systems are much less understood. Notably, behaviours not encountered with spherical systems have previously been reported, including non-monotonic jamming and macroscopic friction responses with respect to particle aspect ratio. Moreover, important microscopic effects including the tendency of elongated particles to align preferentially in the flow direction are expected to be crucial for capturing the rheology of such systems. In this work, using the multi-sphere discrete element method, the steady-state rheology of monodisperse elongated particle systems is investigated by performing volume controlled simple shear simulations using the Lees-Edwards boundary condition. Moderately dense to jammed systems of elongated particles with four different aspect ratios and different inter-particle friction coefficients are studied while fixing other primary particle properties. The stress shows different functional dependencies on the shear rate, depending on the solid volume fraction and the shear rate, similar to what is observed for spherical particles. A rheological framework developed for spherical particles, which employs the volume fraction distance to jamming as a key scaling factor, has been extended to accurately predict elongated particle flow behaviour. The key extensions include an aspect-ratio-dependent jamming volume fraction and a lengthscale accounting for particle alignment in the flow direction. This lengthscale can be expressed analytically as a function of simple geometric properties of the particles. Crucially, the new model allows the use of identical parameter values as those used for spherical systems with the same interparticle friction, providing a powerful and convenient tool to predict the flow behaviour of elongated particles based on the calibration to spherical particles.

## Symposium BL

## **Bio-rheology, Living and Active Matter**

Organizers: Laura Casanellas, Alexander Morozov and Christian Wagner

## Wednesday 10:30 LT 23 (Level 8) / Track 5

Structural and rheological evaluation of collagen hydrogels for tissue regeneration

Konstantina Lyroni<sup>1</sup>, Konstantina Lyroni<sup>2</sup>, Dimitris Vlassopoulos<sup>1</sup>, Dimitris Vlassopoulos<sup>2</sup>, Dimitrios Tzeranis<sup>3</sup>, and Michel Cloitre<sup>4</sup>

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Collagen type I, the most abundant protein in mammals, due to the unique mechanochemical properties that exhibits, it is widely used in the production of porous scaffolds via lyophilization for biomedical applications. To optimize its usage, a deeper understanding of the structure and rheological properties of collagen suspensions is necessary. In this work, we explored the morphology of fibrillar collagen suspensions via phase contrast microscopy, and further investigated the characteristics of the networks such as the mesh size and the fiber diameter utilizing confocal microscopy. For the determination of its rheological response, we employed a specially designed cone-partitioned plate geometry that separates the measured bulk properties from the interfacial contributions due to the tendency of proteins to move towards the outer sample edge and form membranes. The viscoelastic behavior of the suspensions is attributed to their network nature. In addition, preliminary data from Particle Image Velocimetry (PIV) contributed to an enhanced understanding of the complementary structural phenomena that occur when the material is subjected to shear.

Wednesday 10:50 LT 23 (Level 8) / Track 5

## Bridging the gap between bulk rheology and local deformations in cell-embedded collagen via rheo-confocal microscopy

<u>Margherita Tavasso</u><sup>1</sup>, Nagle Irène<sup>2</sup>, Iain Muntz<sup>2</sup>, Gijsje H. Koenderink<sup>2</sup>, and Pouyan E. Boukany<sup>1</sup> <sup>1</sup>Department of Chemical Engineering, Delft University of Technology, Delft, The Netherlands; <sup>2</sup>Department of Bionanoscience,

Kavli Institute of Nanoscience, Delft University of Technology, Delft, The Netherlands

During tumour progression and invasion, cancer cells alter the rheological properties of their surrounding extracellular matrix (ECM) in order to promote their dissemination. Invading cancer cells often remodel the ECM to develop a migration path and exert pulling forces to create aligned matrix fibers that facilitate persistent cell motion. It is still unclear how the presence of biochemical modifications and local mechanical

BL1

BL2

deformations of the ECM originated by the cells affect the overall global mechanics of the tissue. Here we addressed this question by creating model tissues composed of ECM networks made of fibrillar collagen I with embedded MDA-MB-231 cells, a highly invasive breast cancer cell line, at varying volume fractions. We find by bulk rheology that the cells strongly affect the mechanics of the collagen networks above a specific volume fraction. To connect this finding to the underlying local changes in structure, we are using rheo-confocal microscopy for simultaneous rheology measurements and in situ visualisation of the interactions between collagen fibers and cells. Our findings can pave the way for a better understanding of how local cell-ECM interactions at microscale affect tissue mechanics at the macroscopic scale.

## Wednesday 11:10 LT 23 (Level 8) / Track 5

BL3

**Stress-ramp during gelation of collagen triggers a tenfold surge in strength and alignment beyond prior limits** <u>Lens M. Dedroog<sup>1</sup></u>, Minne P. Lettinga<sup>2</sup>, Erin Koos<sup>3</sup>, Anja Vananroye<sup>4</sup>, Olivier Deschaume<sup>1</sup>, Carmen Bartic<sup>1</sup>, Yovan de Coene<sup>5</sup>, Wim Thielemans<sup>6</sup>, and Mehdi Bouzid<sup>7</sup>

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Collagen is one of the main building blocks of the mammalian extracellular matrix, due to its ability to form tough structures with a wide variety of non-linear mechanical properties allowing it to support multiple tissue types. The mechanical properties of collagen gels have been extensively studied under static conditions, however, in nature gelation will mostly take place in the presence of flow. Here we show how the elastic modulus of collagen hydrogels can be increased up to one order of magnitude by applying a stress-ramp at a well-defined moment during gelation, following up on our earlier study [1]. Where the first stress block induces most of the final strain and alignment, however sequential increases in stress cause a dramatic increase of the modulus. This high modulus is preserved by keeping the high stress until the gel is fully matured. Coarse grained simulations of a model gel model system suggest that the microscopic mechanism of inducing high stiffness could be very generic. Thus, we not only show that the true non-linear capabilities of biomaterials are tenfold higher than previously assessed, but also provide insight into in vivo structure formation of collagen and potential other (bio-)polymers.

## Wednesday 11:30 LT 23 (Level 8) / Track 5

BL4

## Rheological and mechanical characterization of hydrogel beads as artificial erythrocytes for multiphase blood flow measurements

<u>Gesine Hentschel</u><sup>1</sup>, Christina M. Winkler<sup>1</sup>, Florian Rummel<sup>2</sup>, Katharina Nikutta<sup>3</sup>, Marc Müller<sup>1</sup>, and Birgit Glasmacher<sup>1</sup> <sup>1</sup>Institute for Multiphase Processes, Leibniz University Hannover, Garbsen, Lower Saxony 30823, Germany; <sup>2</sup>NETZSCH-Gerätebau GmbH, Selb, Bavaria 95100, Germany; <sup>3</sup>Department of Prosthetic Dentistry and Biomedical Materials, Hannover Medical School, Hannover, Lower Saxony 30625, Germany

Objectives: Cardiovascular hemodynamic flow models often rely on Particle Image Velocimetry (PIV) experiments, traditionally using a singlephase glycerin/water substitute for visualizing blood flow. This study introduces a novel approach with a multiphase blood substitute utilizing hydrogel microparticles (beads) from poly-sodiumarcylate-co-arcylamide P(SA-Am), P(SA-Am)/Alginat-Co-Hydrogels. Beads, suspended in glycerol/water mixtures to simulate blood plasma, undergo rheological analysis at varying shear rates and are compared to blood samples. Mechanical stability is also assessed. Methods: Microfluidic systems (MFS) were utilised to produce beads with mineral oil as the continuous phase. The beads were dispersed in glycerin/water (5-40vol.%) and their rheology was studied using a plate-plate rheometer at shear rates ranging from 5-20001/s. The viscosity influence of glycerin/water solutions (5-70vol.%) was also tested. The rheological data was then compared to that of bovine and porcine blood samples. Finally, the elastic modulus of the beads was determined using an AFM with a 50 nN load force. Results: We successfully produced P(SA-Am) beads with an average size of 350µm and P(SA-Am)/Alginate-beads with an average size of 20µm. Rheometry confirmed that the blood substitute fluid exhibits shear-thinning behavior at moderate shear rates. The viscosity and sedimentation time of the fluid are affected by the concentration of glycerol. The elastic modulus of the beads is comparable to that of blood samples. Conclusions: The rheology of the blood substitute fluid is characterised, allowing precise control of viscosity and sedimentation time by adjusting the glycerol and bead concentrations. The modulus of elasticity of the beads mimics the mechanical response of erythrocytes. Ongoing PIV testing evaluates the detectability of artificial erythrocytes.

## Wednesday 11:50 LT 23 (Level 8) / Track 5

BL5

Magnetic microwire rheometer reveals differences in hydrogel degradation via disulfide reducing agents

Margaret Braunreuther<sup>1</sup>, Justin Arenhoevel<sup>2</sup>, Raju Bej<sup>3</sup>, Rainer Haag<sup>4</sup>, and <u>Gerald Fuller<sup>5</sup></u>

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Mucus viscoelasticity is determined by the underlying microstructure formed by the mucin polymer network, which is regulated by several factors including polymer concentration, density of covalent and physical bonds, and hydration. In airway diseases such as asthma and cystic fibrosis, mucus hypersecretion and dehydration results in thick, solid-like mucus that impairs transport, leading to mucus accumulation and blockage of

the airway. Mucolytic drugs have been developed to reduce the viscoelasticity of the mucus gel. Reducing agents break down the disulfide bonds between mucins via thiol exchange chemistry. The most commonly used reducing agent drug is N-acetylcysteine; however, its clinical efficacy has been found to be limited due to low activity and a side-effect of bronchoconstriction. Novel reducing agents that are well-tolerated by human patients and effective at breaking down mucin crosslinks are in the process of being developed. Improved understanding of reducing agents effect on mucus material properties is needed to aid in the design of such drugs. Mucus-inspired hydrogels that capture aspects of the dynamic mucin network can help to elucidate the mechanisms of mucus rheology regulation in health and disease. We have demonstrated a magnetic microwire rheometer (MMWR) to measure the changes in material properties of dynamic hydrogels in response to agents that modulate disulfidecrosslinking. In this work, we use the MMWR to compare multiple reducing agents and characterize the degradation of disulfide crosslinked hydrogels as these agents diffuse through the network. While these reducing agents have been investigated via macrorheology methods, we show that the diffusion of these reagents, rather than mixing, into the hydrogel reveals key differences in degradation mechanisms that could have major implications for drug-delivery and efficacy in vivo.

Wednesday 12:10 LT 23 (Level 8) / Track 5

BL6

SV1

## Theoretical and experimental determination of hydrogel mesh size distribution combining rheology and low field NMR

Stefano Mezzasalma<sup>1</sup>, Michela Abrami<sup>2</sup>, Gabriele Grassi<sup>3</sup>, and Mario Grassi<sup>2</sup>

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As the process of polymeric chains crosslinking can be considered a random process, Flory's assumption of a Gaussian distribution of the end-toend distance between consecutive crosslinks is absolutely reasonable. However, the occurrence of network defects, polymer chain stiffness and excluded volume related questions (e.g. non-zero polymer chains diameter) leads to suppose that Gaussian distribution is not always the best choice. Accordingly, our implementation of Flory's theory relys on the idea that the shear modulus (G) of a polymeric network depends on the crosslink density and on a corrective term accounting for the non-gaussian distribution of the end-to-end distance between consecutive crosslinks. The experimental G knowledge allows the determination of the crosslink density that, in turn, according to the equivalent network theory [1], permits the determination of the average mesh size. Finally, relying on the average mesh size, the hydrogel Low-field NMR characterization allows to get the complete mesh size distribution of the polymeric network pervading the hydrogel. Thus, based on our previous work [2], the aim of this contribution is to find a general end-to-end distribution able to properly describe every polymeric network. In order to confirm the general character of our approach, different hydrogels of practical and scientific interest will be considered. In particular, classical agar, alginate and scleroglucan hydrogels will be studied jointly with the sputum of patients affected by cystic fibrosis, the most frequent life-limiting genetic disease in Caucasian populations. This disease, responsible for the anomalous chloride and sodium ion exchange across epithelial membranes, promotes the formation of a thick and viscous mucoid secretion in multiple organs with particular strength in airways, making breathing very difficult.

[1] Schurz, J. (1991). Progress in Polymer Science, 16 (1991), 1-53. [2] Mezzasalma et al., Int. J. Eng. 176 (2022) 103676.

## Symposium SV

## Soft Solids and Viscoplastic Fluids

Organizers: Maria Charalambides, Maziyar Jalaal and Ian Wilson

Wednesday 10:30 LT 11 (Level 10) / Track 6

Shear banding as a cause of non-monotonic stress relaxation

Vanessa K. Ward and Suzanne M. Fielding

Department of Physics, Durham University, Durham DH1 3LE, United Kingdom

Soft glassy materials arise widely in nature and technology. These elastoplastic materials respond as soft solids at rest, but flow as liquids under external shear, displaying a shear-induced solid-to-liquid transition. When flow subsequently stops, they recover their solid-like properties. However, due to the liquid-to-solid transition, there is often only a partial relaxation of the internal stresses, which typically follow a monotonic decay towards a residual stress. Residual stresses affect both linear and nonlinear material responses, and are significant for the mechanical properties of a broad range of materials. Soft glassy materials can demonstrate a memory effect, where the residual stresses are a signature of the plastic deformation history of the sample.

Several recent experiments on different systems have shown a remarkable non-monotonic stress relaxation towards a residual stress, following the switch-off of a steady imposed shear flow. In this work, we show that the Soft Glassy Rheology (SGR) model - a mesoscopic approach widely used to interpret experiments on soft matter - can capture this experimentally observed non-monotonic stress relaxation. We offer a mechanistic understanding, showing that non-monotonic stress relaxation can arise under conditions where both shear banding and a significant degree of frustration of locally stresses are present. Accordingly, we argue that this effect may be generic across many different materials.

Wednesday 10:50 LT 11 (Level 10) / Track 6

Using surface charge to manipulate slip of yield-stress fluids at solid surfaces

Elliott M. Sutton<sup>1</sup>, Athanasios A. Papaderakis<sup>2</sup>, Sri Ganesh Subramanian<sup>3</sup>, Robert A. Dryfe<sup>4</sup>, Finn Box<sup>5</sup>, Claudio P. Fonte<sup>6</sup>, and Anne Juel<sup>7</sup>

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Many yield-stress fluids exhibit slip on solid surfaces. This significantly impacts their flow and can dramatically decrease pressure losses by reducing wall friction. Hence, promoting slip can be beneficial to reduce the energy consumption of industrial processes. It can also help to control the structure of certain products, improving their texture and taste. However, the physical origin of slip in yield-stress fluids remains poorly understood. In this talk, we demonstrate that the slip of a Carbopol microgel on a nanometrically smooth gold surface can be controlled by surface charge imposed electrochemically. When a charge is imposed on the gold surface, an electrical double layer is established, leading to interactions between the negatively charged Carbopol molecules, positively charged co-ions, and the surface. Enhancement of slip is observed for both positive and negative surface charges, with a decreased yield threshold,  $\tau_s$ , to overcome the adhesion of fluid particles to the wall and an increase of the slip coefficient,  $\alpha$ . These parameters determine the increased velocity at the wall due to slip,  $u_s=\alpha(\tau_w-t_s)$ , where  $\tau_w$  is the wall shear stress. We determine the thickness of the nanometric lubrication layer from these measurements and find that they are consistent with the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory predictions. Furthermore, we relate this charge-induced variation in slip to that observed for triboelectric coatings such as Teflon and PDMS.

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Wednesday	11:10	LT 11 (Level 10) / Track 6	SV3
Stress and bo	oundary	sensitivity in hydrogel particle packing flow behavior	

Joshua A. Dijksman<sup>1</sup> and Tom Mullin<sup>2</sup>

<sup>1</sup>Van der Waals - Zeeman Institute, University of Amsterdam, Amsterdam, The Netherlands; <sup>2</sup>Department of Applied Mathematics, Oxford University, Oxford, United Kingdom

Hydrogel particle packings have extraordinary mechanical properties. Notably, they can show creep behavior and other non-Newtonian flow resistance, depending on their composition and the conditions at the boundaries of the packing. We probe the mechanical properties using a classical sinking ball viscometer. This constant stress rheology test which allows us to vary the particle packing fraction, surface compression and relative density of the penetrating sphere. We find that creep behavior is both intruder and boundary stress dependent. Some of the spheres used in the experiments are heavy and sink in the hydrogel sample while others are buoyant and rise from the bottom. The rising sphere dynamics is consistent with the sinking sphere, despite the different boundary conditions applied. Particle image velocimetry measurements provide insight in the extent of the flow field around the sphere. Our results indicate the relevance of both boundary conditions and imposed stresses for the bulk rheology of dense, soft, frictionless particle packings.

Wednesday 11:30 LT 11 (Level 10) / Track 6

#### Delayed material failure after straining in elastic networks

Samuel Walker and Suzanne M. Fielding

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After the rapid imposition of a "step" shear strain to an amorphous material, one might expect to observe either a gradual, smooth stress relaxation as the material settles into its new configuration or a rapid abrupt material failure if the yield strain is exceeded [1, 2]. Through detailed simulations of a simple network model of an amorphous gel-like or fibrous material with thermally activated strand failure, we demonstrate that sudden catastrophic material failure can be delayed to ultra-long timescales after strain imposition. We characterise this failure in terms of rapid system-spanning fractures and a rapid drop in stress. We furthermore, demonstrate a steep increase in failure time with decreasing imposed strain amplitude, decreasing temperature, and network connectivities around the strain stiffening "critical point" [2]. This provides the first direct evidence, obtained via network simulation, of a phenomenon predicted theoretically in the form of highly delayed shear banding instabilities in mesoscopic and continuum constitutive models of amorphous materials [3]. We suggest that the failure mechanism is linked to bonds that are critically strained to just below their breaking threshold, which then breaks on delayed timescales via thermal activation, inducing follow-on avalanche-like fracturing of surrounding bonds and material failure. We hope that both these simulations and analyses will stimulate experimental efforts to demonstrate the phenomenon we observe.

[1] S. Dussi, J. Tauber, and J. Van Der Gucht. "Athermal Fracture of Elastic Networks: How Rigidity Challenges the Unavoidable Size-Induced Brittleness". In: Physical Review Letters (2020).

[2] J. L. Shivers, A. Sharma, and F. C. MacKintosh. "Strain-Controlled Critical Slowing Down in the Rheology of Disordered Networks". In:

#### SV2

SV4

#### Physical Review Letters (2023).

[3] H. A. Lockwood, E. S. Carrington, and S. M. Fielding. "Ultra-delayed material failure via shear banding after straining an amorphous material". In: arXiv (2023).

#### Wednesday 11:50 LT 11 (Level 10) / Track 6

# Direct Evidence of Electric Double Layer (EDL) Repulsive Force Controlling the Time-dependent Behaviour of Clay Gels in the Structural Rejuvenation

#### Yee-Kwong Leong

Chemical Engineering, University of Western Australia, Perth, Western Australia 6009, Australia

In 2018, we postulated that the electric double layer (EDL) repulsive force controls the structural development process during ageing and was therefore responsible for the time-dependent behaviour of charge anisotropic clay gels. The same explanation goes with structural rejuvenation process during the stepdown in the shear rate. Now we have direct evidence that this is indeed the case. A strong EDL repulsive force was needed to accentuate the time-dependent behaviour of charge and shape anisotropic clay gels at the stepdown shear rate. This force was strengthened by pyrophosphate anion adsorption increasing the negative charge density of the clay particles. At the stepdown shear rate of 10 s-1 it is strong enough to disrupt the flow aligned structure attained at 1000 s-1 and orient the particles to form more bonds. The resultant outcome is a stepdown shear stress increasing with time until these structure disruption and bond forming processes reached an equilibrium state. The number of lower energy approach configurations (-ve face - +ve edge) for bonding is reduced by the strengthened EDL repulsive force slowing down the bonding process. The time to reach the equilibrium stepdown shear stress value increased initially and then decreased and became zero at high negative charge density where charge anisotropy of the particles no longer exists. The need of a sufficiently strong EDL repulsive force for the display of time dependent behaviour is true for all clay gels; Laponite, hectorite, NaMnt, sepiolite and kaolin gels. The untreated NaMnt gel displayed time-dependent behaviour as the EDL repulsive force is already strong enough. The same EDL-control time dependent behaviour is similar to the reaction mechanism proposed by collision theory.

## Wednesday 12:10 LT 11 (Level 10) / Track 6

SV6

EF1

SV5

Rheo-optics of giant micelles: SALS patterns of CTAT solutions in presence of sodium bromide

<u>J. Esteban Lopez-Aguilar</u><sup>1</sup>, Moises Romero-Urena<sup>1</sup>, Luis Medina-Torres<sup>1</sup>, and Octavio Manero<sup>2</sup> <sup>1</sup>Faculty of Chemistry ChemEng Dept, National Autonomous University of Mexico, Mexico City, Mexico City 04510, Mexico; <sup>2</sup>Materials Research Institute, National Autonomous University of Mexico, Mexico City, Mexico City 04510, Mexico

In this work, we present the linear and non-linear rheological response of semidiluted gel-like solutions of cetyltrimethylammonium tosylate (CTAT) in the presence of sodium bromide (NaBr) at different concentrations. Small-angle light scattering (SALS) patterns under simple shear flow are presented. We found that the viscosity at vanishing shear-rates, and the relaxation time of the material, decrease with increasing NaBr concentration. In the case of the elastic modulus, the variation was small, which suggests the transition from an entangled to a multiconnected network. We found that the micelles formed by CTAT-NaBr present flow-induced orientation, which cause increased concentration fluctuations. These concentration fluctuations were observed through "butterfly"-like scattering patterns. The analysis of the scattering patterns showed that, with increasing NaBr concentration and shear-rate, the light scattering increased. However, the analysis of the structure factor showed that the intensity of the scattering patterns decreases with increasing counter-ion concentration. This finding suggests that the "butterfly" patterns begin to evolve into "tulip" patterns. The data of the shear viscosity and the magnitude of the complex viscosity showed that the CTAT-NaBr solutions comply with the Cox-Merz rule when the salt-to-surfactant molar ratio is larger than 1.5. These results are consistent with those reported in other investigations and reveal the influence of the type of counter-ion used on the mechanical and optical response of the material.

## Symposium EF

## **Emulsions, Foams and Interfacial Rheology**

Organizers: Simon Cox, Thibaut Divoux and Nick Jaensson

Wednesday 10:30 LT 19 (Level 10) / Track 7 **Role of interfacial viscoelasticity on shear-induced deformation of MXene-covered droplets** <u>Benedetta Attaianese</u> and Ruth Cardinaels *Soft Matter Rheology and Technology, KU Leuven, Heverlee 3001, Belgium* 

Interfacial assembly of a particle network is an interesting way to shape multifunctional materials into 3D architectures. Among many others, leveraging the interfacial properties of conductive 2D particles, such as MXenes, for fabrication of conductive nanocomposites has received heightened interest. Therefore, studying the viscoelastic properties of MXenes at liquid-liquid interfaces and relating these to the dynamics of the dispersed phase under flow is essential to explore the full potential of these materials. In this study, we combine a counter-rotating rheo-optical setup and interfacial rheometry to probe the properties of MXenes at the interface and their effects on the dynamics of MXene-laden droplets under flow. Ti3C2TX nanosheets were synthesized by the MILD method. The MXenes assembly at the water-PDMS interface was promoted by

adding mono- and bi-amine terminated PDMS to the oil phase; due to their negative charges, the amines act as a ligand interacting electrostatically with the nanosheets at the interface and, therefore, creating a mechanically stable interfacial network. The viscoelastic properties of the MXenes interfacial network were investigated both under shear and dilatational deformation: the interfacial shear rheological properties were determined with a double wall-ring geometry (DWR) attached to a stress-controlled rheometer, while the response of the particle network to dilatation of the interface was investigated with pendant drop experiments. Our findings suggest that a strong gel-like network is formed between the two phases and the interfacial viscoelastic response depends on the number of amine groups on the ligand. Subsequently, in-situ visualizations of single droplet dynamics were performed. The MXene-covered droplets act as microcapsules, starting to deform only after a yield point. Moreover, they respond differently under flow and after flow cessation depending on the viscoelastic response of the MXene-covered interface.

## Wednesday 10:50 LT 19 (Level 10) / Track 7

Interfacial shear rheology of lipid films on water contaminated by amyloid protein

<u>Jorge Peixinho</u><sup>1</sup>, Henrique Uchôa<sup>2</sup>, Priscilla R. Varges<sup>2</sup>, Eliana P. Marín Castaño<sup>2</sup>, Nicolás A. Rey<sup>3</sup>, Mônica F. Naccache<sup>2</sup>, and Paulo R. de Souza Mendes<sup>2</sup>

<sup>1</sup>Laboratoire PIMM, CNRS, Arts et Metiers, Paris, France; <sup>2</sup>Mechanical Engineering, PUC-Rio, Rio de Janeiro, Brazil; <sup>3</sup>Chemistry, PUC-Rio, Rio de Janeiro, Brazil

Amyloid  $\beta$  protein (A $\beta$ ) plays a central role in Alzheimer's disease. However, the interaction between A $\beta$  and lipid membranes in not fully understood in static and in dynamic conditions. Here, we show that the surface pressure of DPPC (dipalmitoylphosphatidylcholine) lipids at airwater interface is modified by the addition of small amounts of A $\beta$  fractions. Using Langmuir through experiments combined with a double wall ring, we measure the linear viscoelastic moduli of the lipid layer containing A $\beta$  as both surface pressure and frequency are varied. At relatively large surface pressure, the surface viscosity increases exponentially as expected from a condensed liquid monolayer with no significant increase due to the addition of A $\beta$ . Visualizations using a cryo-SEM (scanning electron microscope) indicate the A $\beta$  forms fibrils and no binding with the lipid membrane has been observed.

#### Wednesday 11:10 LT 19 (Level 10) / Track 7

## 3D shape of sheared droplets in low viscosity emulsions: the effect of inertia

<u>Valentina Preziosi</u><sup>1</sup>, Anik Tarafder<sup>2</sup>, Giovanna Tomaiuolo<sup>1</sup>, Giovanna Tomaiuolo<sup>3</sup>, Kausik Sarkar<sup>4</sup>, Stefano Guido<sup>1</sup>, and Stefano Guido<sup>3</sup>

<sup>1</sup>Department of Chemical, Materials and Production Engineering, University of Naples Federico II, Naples, Italy; <sup>2</sup>Department of Mechanical and Aerospace Engineering, The George Washington University, Washington, WA, United States; <sup>3</sup>CEINGE Advanced Biotechnologies, Naples, Italy; <sup>4</sup>Department of Mechanical and Aerospace Engineering, The George Washington University, Washington, WA, United States

The dynamics of droplet emulsions under flow is a topic of great interest in the field of soft matter and in many industrial applications including food, petroleum, and pharmaceutical sectors. The deformation of a droplet immersed in a continuous immiscible fluid is characterized by a single value of the interfacial tension ? [1, 2] and, in laminar flow regime, depends mainly on: i) the viscosity ratio ??= ?c/?d, where ?c and ?d are the continuous and dispersed phase viscosities, respectively; ii) and on the capillary number Ca = (? ?R?c)/? where ?? is the shear rate, R is the undeformed radius of the droplet [1]. Extensive research, both theoretical and experimental, has been conducted on sheared emulsion droplets, particularly focusing on model flow fields like simple shear, which can provide important insights into more complex flow conditions. However, most of the papers have focused on slow flows, where inertia can be neglected and when the role of inertia has restricted in the continuous phase[3]. In this work, we have used high-speed video microscopy in microcapillaries and numerical simulations with a front-tracking finite-difference method to study the effect of inertia inside the droplet flowing in Stokes flow. Our findings have exploited the three-dimensional shape of droplets under shear flow and indicate that internal inertia increases droplet deformation and alignment with the flow direction, contrasting with external fluid inertia effects. We also examine the rotation of the droplet surface and internal circulation in relation to the capillary number under conditions of inner droplet inertia. This study offers new perspectives on droplet behavior in flow regimes that have been largely unexamined, despite their relevance in widespread applications like in microfluidic devices.

## Wednesday 11:30 LT 19 (Level 10) / Track 7

## Simultaneous Interfacial Rheology and Neutron Reflectometry studies of interfacial films

Pablo Sanchez-Puga<sup>1</sup>, Javier Carrascosa-Tejedor<sup>1</sup>, Mariana Rodríguez-Hakim<sup>2</sup>, Javier Tajuelo<sup>3</sup>, Armando Maestro<sup>4</sup>, Philipp Gutfreund<sup>1</sup>, and Miguel A. Rubio<sup>2</sup>

## <sup>1</sup>Large Scale Structures, Institut Laue-Langevin, Grenoble, France; <sup>2</sup>Departamento de Fisica Fundamental, UNED, Madrid, Spain; <sup>3</sup>Departamento de Fisica Interdisciplinar, UNED, Madrid, Spain; <sup>4</sup>Materials Physics Center, Donosti, Spain

Adsorbed substances fluid interfaces are present in many systems in nature and industrial processes. They can have a complex structural configuration which usually provides a non-linear response to strains. Consequently, they have been the object of study in recent decades both from a structural and rheological point of view. To date, there have been limited examples of simultaneous measurements of interfacial rheology and the structure of fluid interfaces. This aspect holds particular significance, considering the challenge of comparing independently conducted experiments where reproducing identical experimental conditions, such as temperature, is difficult. Specifically, there is great interest in the study of Langmuir monolayers of fatty acids and phospholipids which play a role in many biophysical processes. This work focuses on the development, building, and exploitation of a measurement system based on DWR geometry coupled to a rotational rheometer that allows for simultaneous

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measurements of neutron reflectometry and interfacial rheology on the neutron horizontal reflectometer FIGARO at Institut Laue-Langevin (ILL). In particular, a DWR probe made in titanium by 3D printing has been commissioned for the Anton Paar MCR702e Space rheometer. Additionally, a suitable shear channel with annular geometry (machined in PTFE) has been designed and built to be used on the Langmuir trough. Notably, an improved Flow Field-Based data analysis program has been developed to properly subtract bulk phases contribution and consider non-linear velocity profiles, by implementing a more precise calculation of the strain close to the border of the ring. The studies carried out attempt to shed light on the mechanism of loss of molecules observed in condensed phases at high interfacial pressures in monolayers of fatty acids as well as to understand from a dynamic and structural point of view the role of cholesterol in phospholipid membranes.

#### Wednesday 11:50 LT 19 (Level 10) / Track 7

Interfacial rheological properties of pepsin-hydrolyzed lentil protein isolate at oil-water interfaces

Chaya Chutinara, Leonard Sagis, and Jasper Landman

Wageningen University & Research, Wageningen, The Netherlands

Lentil protein isolate (LPI) was investigated for its potential as a plant protein-based emulsifier. LPI consists mainly of globulins with high molecular weights which are relatively slow to adsorb at an oil-water interface. Smaller proteins, such as whey protein, can adsorb much faster at the interface, quickly forming a viscoelastic film that slows down the rate of droplet recoalescence, leading to smaller droplets. To increase the rate of adsorption, LPI was enzymatically hydrolyzed by pepsin at 1.5% and 4.5% degree of hydrolysis (DH), to obtain small peptides. We studied the behavior of these peptides at the oil-water interfaces in comparison to whey protein isolate (WPI) and native lentil protein isolate (LPI). These proteins were investigated in large amplitude oscillatory shear (LAOS) and large amplitude oscillatory dilatation (LAOD) to investigate the interfacial behavior and relate this to the protein characteristics and emulsion formation. The 1.5% and 4.5% DH LPI showed high surface hydrophobicity, and consisted mainly of low molecular weight peptides, which contributed to faster adsorption kinetics and consequently covered the interface faster than LPI. For the interfacial properties, both hydrolyzed LPI formed brittle and less stretchable interfaces compared to LPI, and in-plane interactions between adjacent protein molecules were clearly weaker than in WPI-stabilized interfaces. Nonetheless, the emulsion prepared with both hydrolyzed LPI samples was not stable due to the droplet flocculation during emulsion preparation. Upon the addition of SDS to break flocs, the droplet size formed by both hydrolyzed LPI was indeed smaller than LPI stabilized droplets, and close to the droplet size formed by WPI.

Wednesday 12:10 LT 19 (Level 10) / Track 7

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## rheoFoam: An open-source package for interfacial rheology simulations implemented in OpenFOAM

Adolfo Esteban<sup>1</sup>, Julio Hernández<sup>1</sup>, Javier Tajuelo<sup>2</sup>, and Miguel A. Rubio<sup>3</sup>

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The experimental characterization of the rheology of complex interfaces is based, in numerous techniques, on measuring the drag on a probe placed in contact with the interface. The analytical solution of the drag force is known in very few geometrical and flow field conditions due to the intricate coupling between momentum transport from the bulk phases, transport at the interface, surface tension, and the interface rheological response. Indeed, analytical solutions are usually found by solving the governing equations in asymptotic limits, which reduces their applicability for experimental data analysis. In this work we present an open-source package for the numerical simulation of the rheological behavior of planar complex interfaces based on the Boussinesq-Scriven constitutive model. The main application goal of the package is the numerical simulation of the flow around objects, such as probes, immersed at a complex interface formed by compounds insoluble in the bulk phases and in isothermal conditions. Two solvers have been developed for simulating both steady and time-dependent problems, while the coupling between the interface coverage and velocity is treated implicitly at each time step. Three validation tests are presented, showing that the straightforward implementation of new interface models and solvers makes this open-source package a versatile tool in the field of computational rheology. Results on the flow and drag forces on a disc at an incompressible interface, a spherical particle moving on a compressible interface, and the Cambridge Interfacial Tensiometer are compared with analytical, numerical, and experimental results reported in the literature.

## Symposium NF

## Non-Newtonian Fluid Mechanics and Flow Instabilities

Organizers: Marco Ellero, Stylianos Varchanis and Helen Wilson

Wednesday 10:30 LT 24 (Level 10) / Track 8

## **Global nonlinear stability of Oldroyd-B fluids in plane Couette flow** Andrew Wynn

#### Department of Aeronautics, Imperial College London, London, United Kingdom

This talk will give a rigorous proof of global nonlinear stability for Oldroyd-B fluids in plane Couette flow. Specifically, we derive conditions under which perturbations of arbitrary magnitude must decay asymptotically, which naturally extend classical results for Newtonian fluids to the non-Newtonian setting. The challenge of performing nonlinear stability analysis for viscoeasltic flows is in finding an appropriate quantification

of both perturbations from a base-state. To this end, we introduce a new measure, the perturbation entropy, which generalises the notion of relative entropy and gives a probabilistic measure of the deviation of the polymer configuration tensor from its steady distribution. We will show that at any Reynolds number lower than the Newtonian energy stability limit, there exists a range of polymer densities and Weissenberg numbers for which the Oldroyd-B viscoelastic model is also nonlinearly stable.

#### Wednesday 10:50 LT 24 (Level 10) / Track 8

## The role of elastic instability on the self-assembly of particle chains in simple shear flow

Matthew G. Smith<sup>1</sup>, Graham M. Gibson<sup>2</sup>, Andreas Link<sup>1</sup>, Anand Raghavan<sup>3</sup>, Andrew Clarke<sup>3</sup>, Thomas Franke<sup>1</sup>, and <u>Manlio Tassieri<sup>1</sup></u>

## <sup>1</sup>Biomedical Engineering, University of Glasgow, Glasgow G12 8LT, United Kingdom; <sup>2</sup>School of Physics, University of Glasgow, Glasgow, Glasgow G12 8LT, United Kingdom; <sup>3</sup>Schlumberger Cambridge Research, Cambridge CB3 0EL, United Kingdom

Flow-Induced Self-Assembly (FISA) is the phenomena of particle chaining in viscoelastic fluids while experiencing shear flow. FISA has a large number of applications across many fields including material science, food processing and biomedical engineering. Nonetheless, this phenomena is currently not fully understood and little has been done in literature so far to investigate the possible effects of the shear-induced elastic instability. In this work, a bespoke cone and plate shear cell is used to provide new insights on the FISA dynamics. In particular, we have fine tuned the applied shear rates to investigate the chaining phenomenon of micron-sized spherical particles suspended into a viscoelastic fluid characterised by a distinct onset of elastic instability. This has allowed us to reveal three phenomena never reported in literature before, i.e.: (I) the onset of the elastic instability is strongly correlated with an enhancement of FISA; (II) particle chains break apart when a constant shear is applied for 'sufficiently' long-time (i.e. much longer than the fluids' longest relaxation time). This latter point correlates well with the outcomes of parallel superposition shear measurements, which (III) reveal a fading of the elastic component of the suspending fluid during continuous shear flows.

## Wednesday 11:10 LT 24 (Level 10) / Track 8

#### Aerosolization mechanisms of saliva during speech

<u>Tristan Xabada</u><sup>1</sup>, Rodolfo Brandão<sup>2</sup>, Howard A. Stone<sup>2</sup>, Christian Ligoure<sup>1</sup>, and Manouk Abkarian<sup>3</sup> <sup>1</sup>Soft Matter, Laboratoire Charles Coulomb, Montpellier, Occitanie 34000, France; <sup>2</sup>Department of Mechanical and Aerospace Engineering, Princeton, NJ 08544, United States; <sup>3</sup>Centre de Biologie Structurale, CNRS, Montpellier, Occitanie 34000, France

Phonation produces aerosols, and speech has proven to be a pernicious, invisible, yet potent viral transmission route among asymptomatic individuals in the COVID-19 pandemic. To develop comprehensive mitigation strategies and mechanisms to control transmission, it is crucial to understand the atomization processes during speech.

Here we present a preliminary study on the destabilization of saliva filaments produced artificially by the fast and controlled displacement of two surfaces in a few milliseconds mimicking the opening of the lips observed during bilabial stop consonant phonation.

We describe the process by which these filaments are deformed and destabilize under rapid air flow conditions of tens of meters per seconds analogous to the conditions representative of consonant plosion. Then, we observe and analyse saliva filaments created on the lips in vivo, within an uncontrolled environment, to better understand real-life atomization.

We finally propose an analytical model to describe the shape of the filaments and discuss their destabilization into droplets in relationship to the rheology of the solution.

#### Wednesday 11:30 LT 24 (Level 10) / Track 8

#### Transition sequences of 1D, 2D nanomaterials and their hybrids in Taylor-Couette flow

Kesavan Sekar<sup>1</sup>, Viney Ghai<sup>1</sup>, Ann E. Terry<sup>2</sup>, Kim Nygård<sup>2</sup>, and Roland Kádár<sup>1</sup> <sup>1</sup>Chalmers University of Technology, Göteborg, Sweden; <sup>2</sup>MAX IV Laboratory, Lund University, Lund, Sweden

We examine non-Newtonian effects in supercritical flow between independently rotating concentric cylinders of nanostructured self-assembling systems. Instabilities in the flow between rotating concentric cylinders have been one of the benchmarks for stability analysis for over a century. For non-Newtonian fluids, a significant amount of research has been dedicated to identifying the roles of elasticity and shear-thinning using polymer solutions. However, much less is known about the flow stability of self-assembling, lyotropic liquid crystalline systems. Here, we compare the laminar-turbulent transition of aqueous suspensions of one-dimensional (1D; cellulose nanocrystals) and two-dimensional (2D; graphene oxide) suspensions and hybrid compositions thereof. The experiments are performed in ramped outer cylinder rotation and counter-rotation modes using a custom-built Taylor-Couette (TC) cell based on an Anton Paar MCR702e Space rheometer with twin drives. While visualization aids are required in conventional TC experiments, we use a special TC cross-polarized light setup, which we have previously developed. Thus, we map the instability modes through the intrinsic shear-induced birefringence of the suspensions in the form of colourful flow patterns. The transition sequences are thereafter identified using 2D Fourier transform pattern spectral analysis. Interestingly, considering only the case in which the inner cylinder is rotating, and the outer cylinder is at rest, CNC suspensions showed Newtonian-like instability modes modified by shear-thinning, as evidenced by lower characteristic wavelengths compared to the Newtonian reference case. However, GO suspensions revealed the onset of axisymmetric toroidal vortices (Taylor vortex flow) having higher wavelength than the Newtonian reference, as well as distinct high Re turbulent flow patterns.

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## Wednesday 11:50 LT 24 (Level 10) / Track 8

# Three-dimensional elastic instabilities in the flow of viscoelastic and elastoviscoplastic fluids around a confined circular cylinder

Kazi Tassawar Iqbal<sup>1</sup>, Saeed Parvar<sup>2</sup>, and Outi Tammisola<sup>3</sup>

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The flow of complex fluids around a confined cylinder is ubiquitous in many industrial processes in polymer processing and is a canonical flow case of great theoretical importance in experimental and computational rheology. Fluids exhibiting elasticity can display flow instabilities at conditions of negligible inertia which are not be observed in Newtonian fluids. McKinley et al. [1] reported the formation of a three-dimensional cellular structure in the wake a viscoelastic flow past a confined cylinder, which motivates the foundation of the present work. We perform direct numerical simulations to study the three-dimensional flow structures which form in the flow of viscoelastic (VE) and elastoviscoplastic (EVP) fluids around a confined circular cylinder at low Reynolds number. We use an in-house research code based on the finite-difference method to solve the Navier-Stokes equation, and use the Oldroyd-B and Saramito models, respectively, to describe the VE and EVP fluids. The immersed boundary method is used to embed the cylinder geometry into the computational domain and enforce the no-slip boundary condition on its surface. We perform parametric studies of the Deborah (De) and Bingham (Bn) numbers to investigate the effects of elasticity and plasticity on the flow instabilities. We successfully observe that steady, three dimensional cellular structures form on the velocity and stress fields along the spanwise direction in VE fluids, as well as transition to unsteady perturbations with increasing De, as reported in [1]. Furthermore, our parametric studies of Bn show not only that the instability persists in the presence of a yield stress, but becomes intensified by it; increasing Bn at a constant De results in an increase in the amplitude of both the instantaneous perturbations in the velocity profile and that of the unsteady fluctuations.

[1] G. McKinley, R. C. Armstrong, and R. Brown. "The wake instability in viscoelastic flow past confined circular cylinders." Phys. Sci. Eng. 344.1671 (1993)

#### Wednesday 12:10 LT 24 (Level 10) / Track 8

NF6

NF5

Linear stability analysis of the viscoelastic flow around a confined cylinder

Alexandros Spyridakis, Pantelis Moschopoulos, Yannis Dimakopoulos, and John Tsamopoulos

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Increasing elasticity in a viscoelastic fluid, turns the 2D symmetric flow around a confined cylinder into either an asymmetric 2D flow <sup>[1]</sup> or a 3D flow at the wake of the cylinder with periodic velocity fluctuations along the axis of the cylinder <sup>[2,3]</sup>. We investigate both elastic instabilities by means of linear stability analysis. We solve the steady 2D problem with our new stabilized finite element method <sup>[4]</sup>. Then, we impose a small perturbation on all flow variables. We solve the resulting eigenvalue problem using the SLEPc toolkit <sup>[5]</sup>. We reproduce the 2D asymmetric flow instability, which is present in shear-thinning viscoelastic fluids. Our predictions agree with previous simulations and experiments <sup>[1]</sup>. Moreover, we capture the 3D periodic instability in Boger fluids, which has previously been observed only experimentally <sup>[2,3]</sup>. We investigate the effect of the material properties and the geometry on the onset of the instability. Additionally, we perform an energy analysis, and we propose the physical mechanism of the instability. Finally, we assess the complications of introducing shear-thinning.

Research supported by the Hellenic Foundation for Research and Innovation (H.F.R.I. FM17-2309) and HORIZON 2020: YieldGAP Project (955605).

<sup>[1]</sup> S. Varchanis, C. Hopkins, A. Shen, J. Tsamopoulos, S. Haward, Phys. fluids. 32, 053103 (2020).

<sup>[2]</sup> G. McKinley, R. Armstrong, R. Brown, Phil. Trans., Phys. Sci. Eng. 344, 1671 (1993).

<sup>[3]</sup> J. A. Byars, PhD dissertation thesis (1996).

- <sup>[4]</sup> S. Varchanis, A. Syrakos, Y. Dimakopoulos, J. Tsamopoulos, JNNFM. 267 (2019).
- <sup>[5]</sup> V. Hernandez, J. Roman, V. Vidal, ACM Tran. Math. Softw. 31, 3 (2005).

## Wednesday Afternoon

## Symposium MR

## **Microrheology and Microfluidics**

## Organizers: Sepideh Khodaparast, Anke Lindner and Monica Oliveira

Wednesday 1:50 LT 1 (Level 7) / Track 1

## Dynamics of non-spherical particles in viscoelastic fluids flowing in a microchannel

Andrea Langella<sup>1</sup>, Giorgia Franzino<sup>2</sup>, Pier Luca Maffettone<sup>1</sup>, Domenico Larobina<sup>2</sup>, and Gaetano D'Avino<sup>1</sup>

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Particle manipulation through viscoelastic liquids in microfluidics is a well-established technique to achieve focusing, separation, and ordering in geometrically simple channels. Since the first application of the viscoelasticity-driven lateral displacement of particles in microfluidics, several works have been carried out clarifying the effect of fluid rheology, flow conditions, channel geometry, particle properties on the efficiency of focusing, i.e., the capability to obtain a single plane or line of particles. Most of the available works deal with spherical particle suspensions. However, in real situations, the shape of the suspended particles may be different from the sphere, especially in biological applications. In this work, the migration and orientation dynamics of prolate spheroidal particles suspended in a viscoelastic liquid flowing in a square microchannel is experimentally investigated. The suspending fluid is an aqueous solution of PolyEthylene Oxyde at relatively high concentration characterized by a high level of elasticity and shear-thinning. Fluid viscoelasticity drives the spheroids towards the channel central region at relatively low flow rates when the particles explore the constant viscosity region of the fluid, without showing a preferential orientation. As the flow rate increases and the fluid enters in the shear-thinning region, a smaller fraction of particles migrates at the central channel region, reducing the focusing efficiency. The focused spheroids rotate sufficiently fast to attain a stable orientation with major axis aligned along the flow direction. The present results are rationalized through a comparison with previous experiments for spheres and numerical predictions for spheroids, clarifying the complex interplay between fluid rheology, particle shape, and confinement on the migration mechanism due to viscoelasticity.

## Wednesday 2:10 LT 1 (Level 7) / Track 1

## Enrichment of soft capsule suspensions under shear flow in confined vessels leads to clogging

Valeria Ciccone<sup>1</sup>, Igor Chernyavsky<sup>2</sup>, and Anne Juel<sup>1</sup>

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Our bodies function on the premise that trillions of soft particles suspended in our blood do not clog our vessels. However, this premise is not met in extreme cases, such as in sickle cell disease where red blood cells (RBCs) under certain conditions change properties and can clog distal blood vessels. In this work, we explore how mechanical clogging can occur in confined suspensions of soft objects, like RBCs in the microcirculation. There remains a paucity of experiments on the collective dynamics of soft objects on the mesoscale despite a wealth of applications ranging from biotechnology, e.g., targeted drug delivery and tissue engineering, to geophysical engineering such as carbon capture.

To study the flow of RBCs, we use a biomimetic model which consists of elastic capsules - drops encapsulated by an elastic membrane - which have been shown to accurately capture the fluid mechanics of RBCs in confined tubes on the order of the size of a RBC (Chen, 2023). We vary the stiffness of the capsules, and we match the surface area-to-volume ratio of RBCs through partial deflation by osmosis. Experiments were performed by flowing the suspension of capsules with a constant flow rate in long microfluidic channels with a cross-section which accommodates approximately ten capsules. The capsule suspension was at a low enough volume fraction to allow for a fluid-like behaviour. We characterise the enrichment process which enhances spatial non-uniformity of the capsule volume fraction and can locally cause jamming when it leads to a sufficiently large volume fraction. We show that the resulting travelling plugs of jammed particles behave like an elastic solid.

Chen, Q., Singh, N., Schirrmann, K., Zhou, Q., Chernyavsky, I. and Juel, A. (2023). "Robust fabrication of ultra-soft tunable PDMS microcapsules as a biomimetic model for red blood cells". In: *Soft Matter* 19, p. 5249.

MR7

MR6

## Wednesday 2:30 LT 1 (Level 7) / Track 1

## Continuous Manufacturing of Microfluidic Fibers Embedded with Ordered Microparticles Combining Viscoelastic Ordering and Ionic Gelation

Francesco Del Giudice<sup>1</sup>, Dave J. Adams<sup>2</sup>, Antonio Maisto<sup>3</sup>, and Daniel McDowall<sup>4</sup>

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Fibers loaded with either particles or cells are widely employed across a variety of fields, including material science, tissue engineering, and pharmaceutical research. However, the concentration of such objects along the fiber length remains stochastic, thus resulting in fibers having heterogeneous properties along their length. We here introduce a new class of material featuring fibers loaded with "equally spaced" microparticles. The fibers were obtained thanks to the combination between the recently discovered viscoelastic particle ordering phenomenon and the well-established process of fiber synthesis via ex situ ionic gelation. We employed a simple experimental apparatus made of a syringe pump connected to a 100 µm tube ending in a calcium chloride bath. The liquid forming the fiber was an aqueous solution of hyaluronic acid and sodium alginate. We studied the effect of volumetric flow rate, sodium alginate concentration, and spinning speed on the fiber diameter and the particle-spacing in the fiber. We also discussed the advantages of this type of fiber over the existing ones and suggested potential applications across several fields

## Wednesday 2:50 LT 1 (Level 7) / Track 1

MR9

## Clogging of interlocking particles in a 2D Hopper

Jules Tampier<sup>1</sup>, Anke Lindner<sup>2</sup>, and Philippe Bourrianne<sup>3</sup>

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Flows of particle suspensions through channels are present in a broad range of situations, ranging from filtration or 3D printing to biopharmaceutical manufacturing and drug delivery. At large concentrations, particle suspensions are known for their ability to block and resist flows. In particular, when particles are forced through a narrow constriction, clogging can occur, leading to an intermittent or permanent drop of discharge. Numerous studies have already demonstrated the key role played by the orifice-to-particle size ratio and interparticle adhesion forces using cylindrical or spherical model particles. The role played by complex particle shapes, of particular interest in practical situations, remains to be understood. We here investigate experimentally the clogging of a suspension of non-convex particles in a 2D microfluidic Hopper flow. We use in-situ micro-fabrication techniques to produce undeformable micrometric particles of tunable shape. We focus our interest on interlocking particles able to jam under flow. Our results show that interlocking strongly increases the probability of clogging by enhancing the formation of stable structures close to the constriction.

## Wednesday 3:40 LT 1 (Level 7) / Track 1

## Enhanced colloidal particle trapping in microgrooved channels via diffusiophoresis

<u>Naval Singh</u><sup>1</sup>, Goran Vladisavljevic<sup>2</sup>, Francois Nadal<sup>2</sup>, Cecile Cottin-Bizonne<sup>3</sup>, Christophe Pirat<sup>3</sup>, and Guido Bolognesi<sup>4</sup> <sup>1</sup>Chemical Engineering, Lancaster University, Lancaster, Lancashire LA14YW, United Kingdom; <sup>2</sup>Loughborough University, Loughborough, United Kingdom; <sup>3</sup>Institut Lumière Matière, CNRS, Lyon, France; <sup>4</sup>Chemistry, University College London, London, United Kingdom

The controlled transport of sub-micron colloidal particles within a confined environment, such as a porous medium or a dead end channel, is a key feature in several technological applications (e.g., drug delivery, diagnostics) as well as in living systems (e.g., mass transport in tissues and capillaries). Recently, we demonstrated how solute gradients in steady-state continuous flows past a microgrooved surface can be exploited to induce the controlled and reversible trapping of sub-micron particles within the dead-end grooves. The trapping mechanism is governed by diffusiophoresis, which drives particle motion along a solute gradient. In this study, we investigate how the particle trapping is affected by the groove geometry, channel surface chemistry and solute gradient intensity, thereby determining the conditions for enhancing the particle trapping performance. The microfluidic devices, featuring a 3-inlet junction for generating the salt gradients, are made of an optical glue (NOA-81) layer, laid on a silicon microgrooved substrate. The proposed approach for particle transport in lab-on-a-chip devices has potential applications in point-of-care, drug delivery and biosensing industry.

## Wednesday 4:00 LT 1 (Level 7) / Track 1

## Colloidal Ratchet Effect in a Viscoelastic Fluid

Guillermo Camacho<sup>1</sup>, Pietro Tierno<sup>2</sup>, Sofia Kantorovich<sup>3</sup>, Pedro A. Sánchez<sup>3</sup>, and Juan de Vicente<sup>1</sup>

<sup>1</sup>Applied Physics, University of Granada, Granada, Andalucía 18071, Spain; <sup>2</sup>Departament de Física de la Matèria Condensada, University of Barcelona, Barcelona, Spain; <sup>3</sup>Dipolar Soft Matter, University of Vienna, Vienna, Austria

Ratchet mechanisms leverage spatial or temporal asymmetries to convert external fluctuations into directed forces, yielding a direct current or flux upon the application of a periodic drive. At a practical level, the ratchet effect enables the extraction of useful work from thermodynamic systems, manifesting in various physical and biological processes.

Microscopic colloidal systems serve as accessible models for investigating ratchet transport, employing a diverse range of mechanisms [Annu. Rev. Condens. Matter Phys. 2017. 8:51-75]. However, the majority of these models utilize Newtonian fluids as carrier media.

**MR10** 

MR11

This study proposes a departure from the convention by advocating for the use of non-Newtonian fluids to exploit their rheological properties and generate the required asymmetries. Our experimental setup involves micron-sized magnetic particles suspended in various non-Newtonian fluids, subjected to a square wave magnetophoretic force alternating in opposite directions while adhering to the condition  $|F_1 \cdot T_1| = |F_2 \cdot T_2|$ . This modulation condition ensures a zero average force, and no asymmetric dissipation in obtained in the case of Newtonian fluids. In contrast, viscoelastic carrier fluids can asymmetrically respond to opposite forces, inducing directed motion on colloids.

To elucidate the impact of viscoelasticity and non-Newtonian viscosity on the ratchet mechanism, we employ a numerical model to solve the Generalized Langevin Equation, exploring diverse constitutive models and their associated drift dynamics. Additionally, we present an experimental realization of this phenomenon using dilute biopolymer solutions or micellar solutions as carrier media. This effect demonstrates promising applications in "self-viscophoresis" [Langmuir 2020, 36, 25, 7148-7154] and offers insights into inferring rheological properties by analyzing particle dynamics in the medium.

Wednesday 4:20 LT 1 (Level 7) / Track 1

**MR12** 

#### Fluid dynamics of coffee extraction

<u>Marco Ellero</u><sup>1</sup>, Chaojie Mo<sup>2</sup>, Luciano Navarini<sup>3</sup>, Furio Suggi Liverani<sup>3</sup>, and Richard Johnston<sup>4</sup> <sup>1</sup>Basque Center for Applied Mathematics - BCAM, BILBAO, BIZKAIA 48009, Spain; <sup>2</sup>Aircraft and Propulsion Laboratory,, Ningbo Institute of Technology, Beihang University, Ningbo, Beihang, China; <sup>3</sup>Illycaffe', Trieste, Italy; <sup>4</sup>Swansea University, Swansea, United Kingdom

Espresso extraction is a very complex process involving flows through a porous medium undergoing geometric changes, solubilisation of hydrophilic substances, emulsification of insoluble coffee oils and suspension of coffee cell-wall fragments ("fines"). In this talk we discuss the effects of changing porous bed micro-morphology on the extraction kinetics. We devise mesoscopic flow models in the framework of smoothed particle hydrodynamics (SPH) [1] to model fines migration, particle erosion, and particle swelling. It is found that, fines migration leads to a transient permeability for the porous matrix, and results in an increase of compound content in the cup [1]. Particle erosion induced by the mechanical force of flow leads to channelisation phenomenon, and if the erosion if pressure-dependent, heterogeneity in the filtration direction will also be induced. As a consequence, the flow is heterogeneous and the extraction is unbalanced in the matrix [2]. Particle swelling affects both the intra-granular and inter-granular transport of the soluble compounds. A very small degree of coffee particle swelling will significantly enhance the compound content in the cup, potentially altering taste and aroma of the espresso [3]. Finally, the possibility to target real systems using X-ray microCT data of coffee capsules will be also discussed [4].

References:

[1] Ellero, M., Navarini, L. "Mesoscopic modelling and simulation of espresso coffee extraction" J Food Engng, 263, 181-194 (2019).

[2] Mo, C et al. "Modeling the effect of flow-induced mechanical erosion during coffee filtration" Physics of Fluids, 33(9) (2021).

[3] Mo, C. et al. "Modeling swelling effects during coffee extraction with smoothed particle hydrodynamics". Physics of Fluids, 34(4), 043104 (2022).

[4] Mo, C et al. "Exploring the link between coffee matrix microstructure and flow properties using combined X-ray microtomography and smoothed particle hydrodynamics simulations", Sci. Rep. 13, 16374 (2023).

#### Wednesday 4:40 LT 1 (Level 7) / Track 1

**MR13** 

## Elongational properties of low concentration Carbopol/PEO solutions by a flow focusing microfluidic technique

#### Andrea I. Cirillo, <u>Fabiana Gallo</u>, Emanuela Longo, Raffaele Graziano, Giovanna Tomaiuolo, and Stefano Guido Department of Chemical, Materials and Production Engineering, University of Naples Federico II, Napoli, Na 80125, Italy

The elongational behavior of fluids turns out to be of great interest for scientific research being numerous application fields in which it is present, especially for the industry (e.g., Enhanced Oil Recovery, food and polymer processing, adhesives production). However, the main problem lies in the difficulty of obtaining experimentally measurements of elongational viscosity for low concentrated solutions, which cannot be studied with the classical analysis devices used for particularly viscous fluids. One possible solution lies in microfluidics, a technology that involves the manipulation of extremely small volumes of fluids in micrometer-sized channels. The use of microfluidic technologies in recent years has also managed to make its way into the analysis of the elongational properties of Newtonian and complex fluids, including low-concentration solutions characterized by low viscosity. In this work, we characterize the elongational properties of low concentration Carbopol, Polyethylene oxide (PEO), and Carbopol - PEO aqueous solutions by using a purely microfluidic approach. In detail, a cross microfluidic chip was used to collimate these polymer solutions by means of two lateral streams of an immiscible phase (i.e., silicone oil), exploiting the principle of flow focusing. Given the presence of two non-miscible fluids, it was necessary to evaluate the interfacial tension, a fundamental parameter for understanding a two-phase system. As for extensional viscosity evaluation, a purely microfluidic approach was used also for interfacial tension measurements.

## Symposium PF

## **Polymeric Fluids**

Organizers: Francesco Del Giudice, Richard Graham and Evelyne van Ruymbeke

## Wednesday 1:50 LT 17 (Level 7) / Track 2

## Interaction between polymer grafted surfaces in a melt

Christos Psevdos<sup>1</sup>, Giovanni Ianniruberto<sup>1</sup>, Giuseppe Marrucci<sup>1</sup>, Salvatore Coppola<sup>2</sup>, and Francesco Della Penna<sup>2</sup>

<sup>1</sup>Department of Chemical, Materials & Production Engineering, University of Naples Federico II, Naples, Italy; <sup>2</sup>Versalis SpA, Ravenna, Italy

Filler dispersion in rubber compounds may be facilitated by grafting polymer chains on the surface of the filler particles. The main idea is that the grafted chain conformations will be significantly restricted in the limited space between approaching particle surfaces, giving rise to repulsive forces. If these forces are of sufficient magnitude, particle aggregation/agglomeration may be prevented (1). Such physical system may be represented - in the simplest case - by polymer brushes placed on opposing flat planes. Configuration properties of grafted chains in this setup have been studied from a theoretical, computational, and experimental standpoint, both in quiescent and dynamic (steady shear) conditions (2). Another quantity of particular interest is the net force developing between the two grafted walls as a function of their separation and other system properties (e.g., chain length, nature of wall/polymer interactions, flow regime). However, net surface interaction has so far been studied just for the case where the brush is immersed in a low molecular weight (good) solvent (3). Obviously, this does not account for polymer composites, where the grafted polymer brush is surrounded by a melt consisting of free chains. In this work we perform coarse-grained molecular dynamics simulations of polymer brushes immersed in a melt, in order to explore how the force between the grafted surfaces evolves with separation in different scenarios.

1. S. K. Kumar, N. Jouault, B. Benicewicz, T. Neely, Macromolecules. 46, 3199-3214 (2013).

2. R. A. L. Jones, R. W. Richards, Polymers at Surfaces and Interfaces (Cambridge University Press, ed. 1, 1999;

https://www.cambridge.org/core/product/identifier/9780511623196/type/book).

3. G. S. Grest, in Polymers in Confined Environments, S. Granick et al., Eds. (Springer Berlin Heidelberg, Berlin, Heidelberg, 1999;

http://link.springer.com/10.1007/3-540-69711-X\_4), vol. 138 of Advances in Polymer Science, pp. 149-183.

## Wednesday 2:10 LT 17 (Level 7) / Track 2

## Synthesis and Characterization of Di- and Tri-block copolymers serving as adaptative matrix in magnetoresponsive nanocomposites

Simon Fritz<sup>1</sup>, Florent Dalmas<sup>1</sup>, Julien Bernard<sup>2</sup>, and Guilhem P. Baeza<sup>1</sup>

<sup>1</sup>Matéis UMR5510, INSA Lyon, Villeurbanne 69621, France; <sup>2</sup>Imp UMR5223, Insa Lyon, Villeurbanne 69100, France

Model di- and tri-block copolymers based on "hard" Polystyrene (PS) or Poly(2-vinylpyridine) (P2VP) and "soft" Poly(n-butylacrylate) (PnBua) segments were synthetized through RAFT polymerization [1,2]. The choice of the repeating units was guided by our will to selectively adsorb the hard segments onto the surface of magneto-responsive nanoparticles, making possible the tailoring of the (biphasic) matrix structure from an electromagnetic stimulus. We will first present structure-properties oriented results on the neat polymer, combining AFM and Small Angle X-ray Scattering on one side, and linear and non-linear rheological tests on the other side. We will then show how the incorporation of responsive nanoparticles modify the matrix self-assembly and can be used to tailor the so-formed nanocomposites in a promising way.

[1] Keddie DJ. A guide to the synthesis of block copolymers using reversible-addition fragmentation chain transfer (RAFT) polymerization. Chem Soc Rev 2014;43:496-505. https://doi.org/10.1039/C3CS60290G.

[2] Coutouly C et al. Exploiting the lower disorder-to-order temperature in polystyrene- b -poly(n- butyl acrylate)- b -polystyrene triblock copolymers to increase their flow resistance at high temperature. J Rheol 2022;66:1305-18. https://doi.org/10.1122/8.0000506.

## Wednesday 2:30 LT 17 (Level 7) / Track 2

## Assessing the effect of functionalized polymers on filler dispersion in elastomer compounds

Salvatore Coppola<sup>1</sup>, Federico Sebastiano Grasso<sup>1</sup>, Massimo Demaio<sup>2</sup>, Francesco Della Penna<sup>1</sup>, Luigi Franchini<sup>1</sup>, Marco Valerio Morbidelli<sup>1</sup>, and Fabio Bacchelli<sup>1</sup>

## <sup>1</sup>Versalis SpA, Ravenna, Italy; <sup>2</sup>Versalis SpA, Ferrara, Italy

Reducing tire rolling resistance is a very important goal for tire producers (see e.g. Regulation (EU) 2020/740). The crucial factor impacting tire rolling resistance is energy dissipation in the tire tread elastomeric compound. Energy dissipation is mainly influenced by polymer network defects, as well as by irreversibility related to filler network deformation and breakage caused by the cyclic strains occurring during service. A possible way of reducing negative effects related to filler network is to use functionalized polymers. The latter, in fact, can create a link between polymer matrix and fillers already during mixing. In such a way, they improve filler dispersion, and preserve the attained dispersion by reducing filler "flocculation". In this work, we applied two distinct experimental methods to assess filler dispersion in carbon black filled compounds. The first method is derived from rejuvenation protocols, often used with colloidal systems; using a combination of small strain and large strain rheological tests on the uncured compound, we evaluated its sensitivity to large strains (filler network breakage) and the subsequent kinetics of filler

PF7

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**PF10** 

flocculation. We found satisfactory agreement with the ranking derived from classical procedures on vulcanized samples (Payne effect). The second method is based on Large Amplitude Oscillatory Shear (LAOS). This has been applied to a set of two carbon black filled compounds, based on two distinct SBR polymers, having same molar mass and architecture, one with, and the other without chain-end functionalization. Experimental results were analyzed with two different protocols, i.e. Fourier Transform (FT) and Chebyshev decomposition. We show the effect of the functionalization through the analysis of a peak of the third-to-first harmonic ratio, occurring at strains in the range 5% to 10% ca. We checked that this peak is artifact-free and corresponds to a similar local maximum of the strain hardening parameter obtained by Chebyshev decomposition.

#### Wednesday 2:50 LT 17 (Level 7) / Track 2

## Time-Temperature Superposition for Mechanical Properties of Thermoplastics during Aging and Liquid-Solid **Phase Transitions**

#### Dietmar W. Auhl, Oliver Löschke, Felipe Porcher, and Peng Wang

Department of Polymer Materials and Technologies, Technische Universität Berlin, Berlin, Berlin 10587, Germany

Plastics in commercial applications often undergo liquid-solid phase changes during processing and long-lasting loads during life time. To properly design and manufacture for example load-bearing plastic products, it is necessary to know the long-term material behaviour under realistic conditions. However, long-time measurements are costly and therefore, accelerated testing at elevated temperatures is often used. Timetemperature superposition (TTS) can be used in order to both extend the accessible time range, but also speed up testing due to faster kinetics. The procedure aims ultimately to create a master curve for a certain reference temperature. Yet, there arise several issue of correctly obtaining data from experiments due to material structures changing with time and thus, creating such master curves remain a challenge. In this study, different thermoplastics with partial melting, slow crystallisation and aging effects have been investigated, e.g. polylactide (PLA), polypropylene (PP) with 30 wt.-% glass fiber and talcum as well as thermoplastic elastomers (TPE). Oscillatory, stress relaxation, and creep tests were conducted over larger periods of measuring times under controlled stresses and temperatures. The test data could typically be shifted using Arrhenius-type equations with individual activation energies, but the results sensitively indicate early onsets of molecular and morphological changes, e.g. degradation, partial melting and crystallisation during cooling or heating, respectively. Thus, the limits of extended time ranges and measurement windows become detectable and avoidable.

## Symposium IR

## Industrial Rheology, Sustainability and Additive Manufacturing

Organizers: Esther Garcia Tunon, Claire McIlroy and Sylvie Vervoort

3:40 LT 17 (Level 7) / Track 2 Wednesday

Fresh cement as a frictional non-Brownian suspension

James A. Richards<sup>1</sup>, Hao Li<sup>2</sup>, Rory O'Neill<sup>1</sup>, Fraser Laidlaw<sup>2</sup>, and John Royer<sup>2</sup>

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Cement is an essential construction material due to its ability to flow before later setting, however the rheological properties must be tightly controlled. Despite this, much understanding remains empirical. Using a combination of continuous and oscillatory shear flow, we compare fresh Portland cement suspensions to previous measurements on model non-Brownian suspensions to gain a micro-physical understanding. Comparing the yield stresses obtained under steady and small-amplitude oscillatory shear, we reveal two distinct jamming concentrations,  $\varphi_{u}$  and  $\varphi_{rcp}$ , where the respective yield stresses diverge. As in prior work on model suspensions, the steady-shear jamming point is notably below the oscillatory jamming point,  $\phi_{\mu}$ , suggesting that it is tied to frictional particle contacts. These results indicate that recently established models for the rheology of frictional, adhesive non-Brownian suspensions can be applied to fresh cement pastes, offering a new framework to understand the role of cement additives and fillers. Such micro-physical understanding can guide cement formulation changes to improve performance and reduce environmental impact.

4:00 LT 17 (Level 7) / Track 2 Wednesday

## The potential of gellan gum as an alternative polymer binder in toothpastes

Glen A. Redpath<sup>1</sup>, Tim W. Overton<sup>1</sup>, Eddie Pelan<sup>1</sup>, Lisa Manus<sup>2</sup>, and Andrei Potanin<sup>2</sup>

<sup>1</sup>School of Chemical Engineering, University of Birmingham, Birmingham, Birmingham B15 2TT, United Kingdom; <sup>2</sup>Colgate-Palmolive, Piscataway, NJ 08854, United States

A typical toothpaste formulation includes solids (abrasives, rheology modifiers), aqueous polyol(s), hydrocolloid binder(s), active ingredients, flavourings and preservatives. Products have specific rheological properties to meet consumer requirements such as high yield stresses for shape retention, shear-thinning and thus extrudable, and low levels of 'stringiness' so that paste application is not wasteful or messy. These consumer metrics have been correlated with rheological/tribological properties in the past [1], [2].

IR1

IR2

Gellan gum does not appear widely in commercial toothpaste formulations but is w/w% highly effective as a viscosity modifier. The motivation of this work is to study and quantify the potential use of high acyl gellan gum as an alternative or (synergetic) additive polymer binder in toothpaste formulations. Rotational rheology and mechanical texture analysis were employed to characterise the behaviour of the model pastes. A bespoke geometry and testing/processing protocols have been developed to characterise 'stringiness' based on previous methods [1].

Binders used traditionally in toothpastes such as xanthan gum and sodium CMC exhibited stand-up and stringiness consistent with previous work [1]. Formulations containing gellan gum had excellent stand-up properties for very low w/w%. Formulations containing cellulose derivatives were significantly more "stringy' than those without.

Based on the findings of this work, there could be opportunities to consider the use of more non-traditional binders like gellan gum in toothpaste formulations, thereby improving future supply chain resilience for manufacturers.

[1] A. Ahuja and A. Potanin, "Rheological and sensory properties of toothpastes," Rheol. Acta, vol. 57, no. 6-7, pp. 459-471, 2018, doi: 10.1007/s00397-018-1090-z.

[2] H. Cai, Y. Li, and J. Chen, "Biotribology Rheology and Tribology Study of the Sensory Perception of Oral Care Products," Biotribology, vol. 10, no. March, pp. 17-25, 2017, doi: 10.1016/j.biotri.2017.03.001.

#### Wednesday 4:20 LT 17 (Level 7) / Track 2

IR3

**PVDF/Organocarbonate thermoreversible gels: a thermal, rheological, and conformational study** Davide Nocita<sup>1</sup>, Glen Thompson<sup>2</sup>, Tim D. Gough<sup>3</sup>, Adrian L. Kelly<sup>4</sup>, and Phil D. Coates<sup>5</sup>

<sup>1</sup>Polymer IRC - Faculty of Engineering & Digital Technologies, University Of Bradford, Bradford, West Yorkshire BD7 1AZ, United Kingdom; <sup>2</sup>Polymer IRC - Faculty of Engineering & Digital Technologies, University Of Bradford, Bradford, West Yorkshire BD7 1AZ, United Kingdom; <sup>3</sup>Polymer IRC - Faculty of Engineering & Digital Technologies, University Of Bradford, Bradford, West Yorkshire BD7 1AZ, United Kingdom; <sup>4</sup>Polymer IRC - Faculty of Engineering & Digital Technologies, University Of Bradford, Bradford, West Yorkshire BD7 1AZ, United Kingdom; <sup>5</sup>Polymer IRC - Faculty of Engineering & Digital Technologies, University Of Bradford, Bradford, West Yorkshire BD7 1AZ, United Kingdom; <sup>5</sup>Polymer IRC - Faculty of Engineering & Digital

Polymer gels have a wide range of industrial applications and somewhat complex rheological behaviour, which can lead to significant challenges for processing. Sols and gels of Poly(vinylidene fluoride) (PVdF) and related copolymers, combined with organocarbonate solvents, have been characterised to better understand melt flow behaviour. Thermo-reversible gels were analysed by means of differential scanning calorimetry, parallel plate rheology, capillary rheometry, and vibrational spectroscopy; revealing an underpinning structural complexity and some peculiar visco-elastic properties. The rheological behaviour of the compounds was modelled with an 8 or 9 relaxation modes Giesekus or Phan-Thien Tanner differential visco-elastic constitutive models. The melting point depression and the chain configuration transitions of polymorphic PVdF homopolymer and copolymer structures were found to be composition dependent. The solvent mixture acted as a good plasticizer and gelling agent (?Flory ? 0.5). It promoted the formation of TTT (all trans,  $\beta$ -PVdF) and T3GT3G' (Trans x 3 - Gauche, ?-PVdF) electroactive poly-morphs during gelation. Oscillatory rheology tests performed over a wide temperature range (20 - 160 °C) revealed some instabilities occurring during the first gel-sol transition and a hysteretic thermal-mechanical behaviour. A pronounced die swell and, mostly, melt instability were found for mild to high shear rates (25 -2000 1/s) during capillary tests, which also showed a noticeable extent of wall slip for formulations and shear rates of interest for industrial production. These results, unitedly to the peculiar frequency response of the sols, give new inputs for modelling the viscoelastic properties of complex polymer fluids by means of differential constitutive models and therefore enabling CFD simulations with improved predictive power.

## Wednesday 4:40 LT 17 (Level 7) / Track 2

IR4

## Bottom-up approach for biodegradable polymers used in additive manufacturing: building computational tools to bridge the gaps

Petra Bacova<sup>1</sup>, Eleftherios Christofi<sup>2</sup>, Vagelis Harmandaris<sup>3</sup>, and Sergio I. Molina<sup>1</sup>

<sup>1</sup>Department of Materials Science, Universidad de Cádiz, Puerto Real, Cádiz 11510, Spain; <sup>2</sup>Computation-based Science and Technology Research Center, The Cyprus Institute, Nicosia 2121, Cyprus; <sup>3</sup>Department of Mathematics and Applied Mathematics, University of Crete, Heraklion, Crete 71409, Greece

In the family of biodegradable polymers, poly(lactic acid) (PLA) occupies a special place among those most popular, mainly due to its versatile usage in additive manufacturing. Despite being commercially available at low price, its usage as a full replacement of the synthetic polymers is hindered by its poor mechanical properties and thermal stability.

In order to be able to tackle fundamental problems related to the structure-properties-performance relationship, we present a systematic simulation study of PLA of a wide range of molecular weights and stereochemistry. More specifically, we analyze the basic structural and dynamical properties at atomistic level and build a chemistry-specific coarse-grained (CG) model to extend the time and length scales to those relevant in the experimental studies. In addition, to close the loop, we implement a machine-learning based methodology to reinsert the atomistic details into CG models of different stereochemistry [1].

Since the computational techniques are considered to be a more sustainable alternative to the experimental characterization, we aim to extend the simulation practices commonly used for synthetic polymers to more complex bio-based polymers. By combining different computational techniques, we provide a consistent set of open-access tools [2,3] with the ultimate goal to facilitate the usage of multiscale computational analysis in the fast-growing field of biodegradable materials and additive manufacturing.

A Physics-informed Deep Learning Approach for Re-introducing Atomic Detail in Coarse-Grained Configurations of Multiple Poly(lactic Acid) Stereoisomers, E. Christofi, P. Bacova, V. Harmandaris, Journal of Chemical Information and Modeling, under revision
https://github.com/SimEA-ERA/PLA-BackMap-CG

[3] https://github.com/pbacova/PLA\_analysis\_tools.git

## Symposium CG

## **Colloids and Glasses**

#### Organizers: Michel Cloitre, Johan Mattsson and George Petekidis

#### Wednesday 1:50 LT 6 (Level 8) / Track 3

Predicting glass transition temperatures from chemical structure in poly(aryl ether ketones)

<u>Sebastian B. Croft</u><sup>1</sup>, Peter D. Olmsted<sup>2</sup>, Peter Hine<sup>1</sup>, Richard Mandle<sup>1</sup>, John Grasmeder<sup>3</sup>, Adam Chaplin<sup>3</sup>, and Johan Mattsson<sup>1</sup> <sup>1</sup>School of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, United Kingdom; <sup>2</sup>Department of Physics & Institute for Soft Matter Synthesis, Georgetown University, Washington, DC 20057, United States; <sup>3</sup>Hillhouse International, Victrex PLC, Thornton Cleveleys, Lancashire FY5 4 QD, United Kingdom

The family of Poly(aryl ether ketones) (PAEK) is an important class of polymers characterised by an alternate arrangement of rigid aryl groups and flexible linkers (such as ethers or ketones). PAEK polymers are used in a broad range of applications including smart-phone speakers, electrical insulation, automotive gears, medical implants and aircraft components. For PAEK, as for all industrially-relevant classes of polymers, it is essential to derive predictive structure-property relationships for design and screening. One key material parameter, which links directly to both the relaxation dynamics and rheology of polymers, is the glass transition temperature  $T_g$ . It is important to identify how to effectively predict  $T_g$ directly from the molecular structure.

In this work, we present a model that successfully predicts  $T_g$  directly from the monomer structure for PAEK polymers. By partitioning the monomer structure into sub-units, and combining optimised molecular geometries of these sub-units with linear statistical models, the model can predict  $T_g$  with a root-mean-squared error of 7-10 degrees *K*. Moreover, we demonstrate that the geometry of the monomer sub-units strongly controls the glass transition. The model both provides a successful method for predicting  $T_g$  based on molecular structure in PAEK polymers, and also provides general physical insight into the structural parameters setting the glass transition properties of polymers. We also use dynamic mechanical analysis (DMA) and broadband dielectric spectroscopy (BDS) experiments on a series of PAEK polymers of systematically varying structure to investigate the links between  $T_g$  and the wider relaxation dynamics observed in these polymers.

#### Wednesday 2:10 LT 6 (Level 8) / Track 3

## Universal aspects of Fickian non-Gaussian diffusion in glass-forming liquids

## Francesco Rusciano, Raffaele Pastore, and Francesco Greco

Department of Chemical, Materials and Production Engineering, University of Naples Federico II, Napoli, Italy 80125, Italy

This talk is at the crossroads between two major issues in soft matter, namely glass transition, a long-standing open issue, and the recently discovered Fickian yet non-Gaussian Diffusion (FnGD), and unveils strong connections between them. In 2009, ground-breaking experiments in complex fluids revealed the existence of a novel type of diffusion, that is distinct from both standard and anomalous diffusion, characterized by a linear time-dependent mean square displacement and a non-Gaussian displacement distribution. In the past few years, many examples of such a FnGD have been reported in literature, and it is now generically associated to some dynamical and/or structural heterogeneity of the environment. Motivated by this feature, we investigate the possible occurrence of FnGD in glass-formers, the epitome of dynamical heterogeneity, drawing on experiments on hard-sphere colloidal suspensions and extensive numerical simulations of simple models of molecular liquids both in 2D and 3D. First, we will demonstrate that FnGD is indeed present in all investigated glass-formers (regardless of both interparticle interactions and dimensionality) and "strengthens" on approaching the glass transition, by identifying distinct timescales for Fickianity, &tau\_F, and for restoring of Gaussianity, &tau\_G (and their their associated length-scales, &xi\_F and &xi\_G). We find &tau\_G ? &tau\_F^&gamma with &gamma  $\neq 1$ , for all investigated systems. We then show that in the deep FnGD regime, displacement distributions display exponential tails with a time-dependent decay length l(t) collapsing onto a power-law master-curve l(t)/&xiG ? (t/&tauG)^0.33 for all systems. Thus, a number of universal scaling laws seem to emerge and characterize the Fickian non-Gaussian regime of glass-forming liquids.

F. Rusciano, R. Pastore and F. Greco (2022). Phys. Rev. Lett., 128(16) F. Rusciano, R. Pastore and F. Greco (2023). IJMS, 24(9) Rusciano et al. (2023). Phys. Rev. Lett., 131(11) F. Rusciano, W. Kob, R. Pastore, F. Greco, in prep.

CG6

CG7

## Wednesday 2:30 LT 6 (Level 8) / Track 3

#### Glass transition and yielding of ultrasoft charged spherical micelles

<u>Roshan Akdar Mohamed Yunus</u><sup>1</sup>, Utku Y. Gurel<sup>2</sup>, Domenico Truzzolillo<sup>3</sup>, Aleksander Guzik<sup>1</sup>, Marc Stuart<sup>4</sup>, Patrizio Raffa<sup>1</sup>, Andrea Giuntoli<sup>2</sup>, and Daniele Parisi<sup>1</sup>

<sup>1</sup>Engineering and Technology Institute Groningen, University of Groningen, Groningen 9747 AG, The Netherlands; <sup>2</sup>Zernike Institute for Advanced Materials, University of Groningen, Groningen 9747 AG, The Netherlands; <sup>3</sup>Laboratoire Charles Coulomb, University of Montpellier, Montpellier, France; <sup>4</sup>Groningen Biomolecular Sciences and Biotechnology Institute, University of Groningen, Groningen 9747 AG, The Netherlands

The glass transition in colloidal systems has long intrigued researchers. While the effect of shape, softness, and particle dispersity on the glass transition has been largely addressed, much less is known about the effect of charges. A great deal of research has been dedicated towards charged Laponite suspensions and nanoparticles, and only a dearth of studies has been conducted on charged soft colloids. In this work, we report the structure and dynamics of multiarm charged colloidal particles. The core of the particle comprises of poly (styrene) (PS) with the corona constituting negatively charged poly (methacrylic acid) (PMAA) arms. In the absence of salt, the latter assumes a significantly stretched configuration. Shear rheology experiments, spanning linear (LVE) and nonlinear (NLVE) viscoelastic domains, were conducted to probe the dynamic phase diagrams. Molecular dynamics simulations (MDS) and X-ray scattering complemented the rheological investigations. In contrast, to the traditional repulsive soft colloidal glasses, our particles exhibit a strong frequency dependence in the LVE spectrum. We attributed this unique behavior to the charged PMAA corona. Severe corona stretching induces strong particle-particle correlation at very low concentrations, triggering the topological glass transition at 0.25 wt.%. The repulsive corona prevents arm interpenetration across the glass transition, and it only engages well within the vitrified state. Moreover, X-ray scattering depicts the lack of crystalline phases with uniform dispersion of the particles. Finally, the yield stress dependence on colloidal packing indicates remarkable softness compared to traditional systems. The distinct properties of our charged hairy colloids enhance our comprehension of glass transitions in colloidal systems, offering avenues to finely tune the dynamics of soft colloids at the molecular level.

#### Wednesday 2:50 LT 6 (Level 8) / Track 3

## Soft and Responsive: Rheological Insights into PNIPAM based Microgels Roberta Angelini

## CNR-ISC, Physics Department, Sapienza University, Rome 00185, Italy

Microgels are soft particles made by cross-linked polymer networks with a hybrid nature between that of polymers and colloids. They are widely used as a colloidal model system because of their swelling properties and their responsivity to external control parameters such temperature or pH. The phase behaviour of microgels has attracted great attention thanks to the large variety of new phenomenologies emerging from their ability to pack at very high volume fractions. Combining rheology [1,2], x-ray photon correlation spectroscopy [3] and small angle x-ray scattering, we perform an extensive experimental study of a thermo- and pH-sensitive microgel composed of Interpenetrated Polymer Network (IPN) of poly(N-isopropylacrylamide) (PNIPAM) and poly(acrylic acid) (PAAc) at fixed PAAc content as a function of weight concentration with the ultimate goal of understanding its complex phase behavior. We distinguish three different rheological regimes, characteristic of three different states: liquid, glass and jammed, the possible molecular mechanisms driving the formation of these states is discussed [1] and a preliminary T-C<sub>w</sub> phase diagram is drawn [4].

[1] S. Franco, E. Buratti, V. Nigro, E. Zaccarelli, B. Ruzicka, R. Angelini, Int. J. Mol. Sci. 22 (8), 4032 (2021)

[2] S Franco, E Buratti, B Ruzicka, V Nigro, N Zoratto, P Matricardi, E Zaccarelli, R Angelini, J. Phys. Condens. Matter 33 (17), 174004 (2021)

[3] V. Nigro, B. Ruzicka, B. Ruta, F. Zontone, M. Bertoldo, E. Buratti and R. Angelini, *Macromolecules* 53, 1596-1603 (2020)

[4] S Franco, E Buratti, V Nigro, M Bertoldo, B Ruzicka, R Angelini, Polymers 14 (1), 115 (2022)

## Wednesday 3:40 LT 6 (Level 8) / Track 3

## Nonlinear rheological behavior of hairy nanoparticles in their dense state

## Jiachen Wan and Xiaorong Wang

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The rheological properties of core-shell hairy nanoparticles in the dense state and their relationship with the core-shell composition and hair density of the particles were investigated. These nanoparticles are synthesized using anionic polymerization via a self-assembly process of macromolecules in solutions. The shell of these nanoparticles is made of brush-like polyisoprene, while the core is made of crosslinked polystyrene. The nonlinear rheological behavior of these hairy nanoparticles in their dense state has two unique characteristics. Firstly, the stress response shows an approximate sinusoidal character accompanied by strong strain softening, as depicted by the stress peak. This indicates that the dynamic response of the material to forced oscillatory shear contains the "linear-nonlinear dichotomy" characteristic. Secondly, the Lissajous loop, or the stress-strain curve, evolves over time, where the peak shear stress drops with time. This suggests that the equilibrium state has yielded to the oscillatory shear and is developing toward a new and softer microstructure. Nonetheless, the material responds sinusoidally to the applied dynamic perturbations throughout the evolution. Compared with small strains, the drop in peak shear stress over time is greater under large strains. Remarkably, when shear deformation stops, the initial equilibrium state can automatically resume its structure and initial rheological responses. In this presentation, we will propose theoretical approaches to describe the unusual nonlinear behavior and discuss the challenges encountered during this endeavor.

CG10

CG9

#### Wednesday 4:00 LT 6 (Level 8) / Track 3

#### Numerical modelling of agglomerate formation under shear flow

Amin Rahmat, Alessio Alexiadis, and Mostafa Barigou

School of Chemical Engineering, University of Birmingham, Birmingham, United Kingdom

Particle agglomeration is a common occurrence in various colloidal systems, exhibiting both favourable and undesirable effects. This research introduces a particle-based numerical model to investigate particle agglomeration during the dewatering process. Our model focuses on the interaction dynamics among dispersed particles, utilising an inter-particle potential. The study systematically assesses the impact of key system parameters on the formation of agglomerates by introducing quantitative measures to characterise their structure and behaviour. Our findings reveal that the interplay between the Reynolds number and pair potential results in the formation of four distinct agglomerate structures. This paper contributes valuable insights into optimising the dewatering process by comprehensively understanding the influential factors and their relationships.

### Wednesday 4:20 LT 6 (Level 8) / Track 3

### CG12

## Three-layer orthotropic organization of cellulose nanocrystals achieved through the combined action of frontal ultrafiltration and ultrasound revealed by in situ SAXS

<u>Frédéric Pignon<sup>1</sup></u>, Emilie Guilbert<sup>1</sup>, Samuel Mandin<sup>1</sup>, Fanny Bosson<sup>1</sup>, Nicolas Hengl<sup>1</sup>, Mohamed Karrouch<sup>1</sup>, Bruno Jean<sup>2</sup>, Jean-luc Putaux<sup>2</sup>, Thomas Gibaud<sup>3</sup>, Sébastien Manneville<sup>3</sup>, and Theyencheri Narayanan<sup>4</sup>

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Although CNCs have a high potential as elementary bricks in biosourced composites, achieving optimal performance of their functional properties is still difficult. Crucial challenges are first to control the orientation and organization of CNCs over a broad spatial scale, and second to control the balance between the colloidal interparticle forces and the external forces induced by the processing. Recently, we were able to evidence the ability of the ultrafiltration processes to develop well-defined layered structures of CNCs [1]. Furthermore, we discovered by using ultrasound waves, it was possible to align the CNCs along the wave propagation direction [2]. In this work, A novel processing method combining frontal ultrafiltration (FU) and ultrasound (US) has been developed. A dedicated channel-cell was designed to simultaneously generate 1) a vertical acoustic force thanks to a vibrating blade at the top and 2) a transmembrane pressure force at the bottom. Time-resolved in situ small-angle X-ray scattering permitted to probe the dynamical structural organization/orientation of CNCs during the processing. For the first time, a typical three-layer orthotropic structure that resembles the articular cartilage organization was achieved in one step during the FU/US process: a first layer composed of CNCs with their director vertically oriented along the direction of US wave propagation direction [3]. We have interpreted our observations, in particular the length scales and time scales involved in the spectacular alignment of the CNCs along the ultrasonic wave direction of propagation in terms of Rayleigh acoustic streaming.

[1] Semeraro E. F. et al., Colloids Surf. A, 584,124030, 2020. [2] Pignon F. et al., J. Phys. Chem. C, 125, 18409-18419, 2021. (doi.org/10.1021/acs.jpcc.1c03506) [3] Pignon F et al. J Coll Interface Sci., under revision, 2023.

## Wednesday 4:40 LT 6 (Level 8) / Track 3

#### CG13

# The Contrasting Behavior of Strongly and Weakly Interfacially Active Asphaltenes on the Rheology of Model Waxy Oils

<u>Abdulraouf Ali</u><sup>1</sup>, Ghinwa Yaghy<sup>2</sup>, Laksha Parameswaran<sup>3</sup>, Chris S. Hodges<sup>4</sup>, Thibaut V.J. Charpentier<sup>5</sup>, Simon Connell<sup>6</sup>, Kevin J. Roberts<sup>7</sup>, and David Harbottle<sup>8</sup>

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Asphaltenes and waxes are two components of crude oil that cause flow assurance issues. Although the components coexist, few studies have considered the effect of asphaltenes on wax crystallization and gel-forming properties. Furthermore, the current understanding remains contradictory with both wax-alleviating and wax-aggravating behaviors observed. In this study, asphaltenes extracted from a heavy crude oil were fractionated into strongly and weakly interfacially active asphaltenes by partitioning at a water-oil interface. The two asphaltene fractions exhibited contrasting physicochemical properties, with the strongly interfacially active asphaltenes (IAA) being more polar due to their higher heteroatom content (particularly S and O) and forming larger aggregates in the solution compared to the weakly interfacially active asphaltenes (referred to as remaining asphaltenes, RA). The two asphaltene fractions lowered both the wax gelation temperature and wax appearance temperature; however, the effect was comparable. The unit cell lattice structure of the wax particle remained unchanged in the presence of asphaltenes, but the wax particles were found to be smaller with RA compared to IAA. However, the key finding of the study is how the two asphaltene fractions

affected the yield strength of the gelled wax. For RA, the yield strength was lowered with an increasing asphaltene concentration, whereas for IAA, the overall effect was to increase the gel yield strength. Because the properties of the wax particles were largely unchanged by the two asphaltene fractions, the result suggests that the asphaltene-asphaltene interaction contributes to the overall yield strength. It was shown that the interaction between RA and RA is repulsive with negligible adhesion, whereas that between IAA and IAA is attractive with strong adhesion. These structure-breaker and structure-maker properties of the two asphaltenes confirm that the asphaltene-asphaltene interaction significantly contributes to modifying strength of a waxy gel.

## Symposium SG

## **Suspensions and Granular Materials**

Organizers: Erin Koos, Wilson Poon and Olivier Pouliquen

Wednesday 1:50 LT 18 (Level 8) / Track 4

## Rheology of granular particles immersed in a molecular gas under uniform shear flow

<u>Rubén Gómez González</u><sup>1</sup> and Vicente Garzó<sup>2</sup>

<sup>1</sup>Departamento de Física, Universidad de Extremadura, Badajoz, Badajoz 06006, Spain; <sup>2</sup>Departamento de Física and ICCAEx, Universidad de Extremadura, Badajoz, Badajoz 06006, Spain

Non-Newtonian transport properties of a dilute gas of inelastic hard spheres immersed in a molecular gas are determined. We assume that the granular gas is sufficiently rarefied and hence, the state of the molecular gas is not disturbed by the presence of the solid particles. In this situation, one can treat the molecular gas as a bath (or thermostat) of elastic hard spheres at a given temperature. Moreover, in spite of the fact that the number density of grains is quite small, we take into account their inelastic collisions among themselves in its kinetic equation. The system (granular gas plus a bath of elastic hard spheres) is subjected to a simple (or uniform) shear flow (USF). In the low-density regime, the rheological properties of the granular gas are determined by solving the Boltzmann kinetic equation by means of Grad's moment method. These properties turn out to be highly nonlinear functions of the shear rate and the remaining parameters of the system. Our results show that the kinetic granular temperature and the non-Newtonian viscosity present a discontinuous shear thickening (DST) effect for sufficiently high values of the mass ratio \$m/m\_g\$ (sm\$ and \$m\_g\$ being the mass of grains and gas particles, respectively). This effect becomes more pronounced as the mass ratio \$m/m\_g\$ increases. In particular, in the Brownian limit (\$m\_g/m\to 0\$) the expressions of the non-Newtonian transport properties derived here are consistent with those previously obtained by considering a coarse-grained approach where the effect of gas phase on grains is through an effective force. Theoretical results are compared against computer simulations in the Brownian limit showing an excellent agreement.

## Wednesday 2:10 LT 18 (Level 8) / Track 4

SG8

SG9

SG7

# Effect of carboxymethyl cellulose and poly (acrylic acid) on the dispersion of silicon particles for lithium-ion batteries

## Kyeong Jin Kim and Kyung Hyun Ahn

School of Chemical and Biological Engineering, Seoul National University, Seoul 08826, Republic of Korea

Silicon, an attractive anode material for lithium-ion batteries, significantly enhances energy density when co-utilized with graphitic particles. However, challenges arise from its volumetric expansion of up to 300% during cycling, leading to stability issues in silicon-containing electrodes. Achieving homogeneous dispersion of silicon particles within the electrodes is imperative for stable cell performance, as silicon agglomerates can dramatically expedite cell degradation. In this context, this study delves into the influence of the degree of substitution (DS) and molecular weight of carboxymethyl cellulose (CMC) on the dispersion characteristics of silicon particles, comparing it with poly (acrylic acid) (PAA) to monitor the impact of polymer type. In Si/polymer model suspensions, CMC (250kDa, DS 0.7) results in significant agglomeration of silicon particles, while PAA (250kDa) and CMC (250kDa, DS 1.2) lead to weakly-agglomerated and well-dispersed silicon particles, respectively. The agglomeration mechanism is explored in terms of polymer-polymer and particle-polymer interactions, revealing the relevance of hydrophobic interaction between CMC (DS 0.7) chains and the affinity between silicon and polymers in the agglomeration behavior of silicon particles. We also examine the impact of the molecular weight of CMC on silicon dispersion, finding that a decrease in the CMC molecular weight improves silicon dispersion even with DS of 0.7. Furthermore, we evaluate how varying the length of CMC chains affects the electrical and mechanical characteristics of Si/C electrodes in terms of silicon dispersion. This study enhances our understanding of silicon dispersion, aiding the interpretation of dispersion characteristics of silicon in more complex slurries containing polymer additives and its consequential impacts on electrode properties.

Wednesday 2:30 LT 18 (Level 8) / Track 4

## From bouncing space probes to toner powders

#### Schütz Denis<sup>1</sup> and Natali Unterberger<sup>2</sup>

<sup>1</sup>Rheology, Anton Paar GmBh, Graz, Styria 8054, Austria; <sup>2</sup>Rheology, Anton Paar GmBh, Graz, Styria 8054, Austria

Ever since the near-failure of the Philae lander of the Rosetta spacecraft mission in 2004, the behavior of Granular Materials (Bulk Solids) under conditions of low- or even microgravity has been an object of intense research scrutiny. Considering the planned future manned missions to the

Moon and Mars, it is also likely to continue. Due to the difficulty of actual experiments in microgravity, this work is so far mostly calculative and theoretical. The applied stress towards a powder column is the most important parameter in the study of the flow (and non-flow) behavior of bulk solids, whereby the significance of the total applied force cannot be understated. In the framework of the Mohr-Coulomb failure criterion applied normal stress is the key determinator of the behavior, likened to temperature in soft matter rheology. The key word here is applied stress, as the theory as well as the employed measurement techniques (Jenike or ring type shear cells) do not factor in artifacts that are contingent upon the powder's own weight. In this fashion a common problem is shared between space physics and chemical engineering as this leads to artifacts, especially in the measurement of bulk solid of high density such as metal powders. Utilizing sub-fluidized system to selectively counteract gravity in a fluidization column and measured by a modern Rheometer, we aim to reconcile experimental data with published calculative data obtained by various model systems of different density. In contrast using applied vacuum under the powder column we intend to simulate applied stress within the same system and compare it to shear cell data.

## Wednesday 2:50 LT 18 (Level 8) / Track 4

## Dense suspensions under novel unsteady shear protocols

Naveen K. Agrawal<sup>1</sup>, Zhouyang Ge<sup>2</sup>, Martin Trulsson<sup>3</sup>, Outi Tammisola<sup>1</sup>, Luca Brandt<sup>1</sup>, and Luca Brandt<sup>4</sup>

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Dense suspensions are ubiquitous in everyday life, yet accurately predicting and controlling their flow properties remains challenging due to the complex interplay between external flow and the underlying microstructure. Even if the suspending fluid is Newtonian, suspensions can exhibit rich rheological behaviors such as non-monotonic complex viscosity and non-zero normal stress differences. Particle dynamics under periodic shear can also demonstrate reversible-irreversible transition (RIT). Previous works on dense suspensions have primarily focused on oscillatory shear protocols, which are periodic and involve shear reversals. However, complex shear protocols are yet to be investigated. This work explores the connections between the rheology and dynamics of non-Brownian suspensions under unsteady shear protocols such as rotary shear (RS) and reversible rotary shear (RRS) and compares them to the oscillatory shear (OS). These new shear protocols are similar to simple shear, where the magnitude of the shear rate remains constant, but its direction rotates continuously. Both the RS and RRS protocols are periodic, but the former does not involve sudden shear reversal, whereas the latter is similar to OS. We have found that the suspension rheology for both RS and RRS is similar to OS: there is a volume fraction-dependent strain amplitude at which the relative viscosity attains a minimum value. However, suspension dynamics are qualitatively different. While suspensions under RRS show a dynamical phase transition from 'absorbing' states to 'diffusive' states past a volume fraction-dependent critical strain amplitude, similar to OS, the same suspension remains diffusive in RS. Finally, we discuss how the rheological response and dynamics of suspensions are linked to the microstructure evolution in our novel complex shear protocols.

#### Wednesday 3:40 LT 18 (Level 8) / Track 4

## The approach of a sphere to a wall in a dense suspension

Antoine Seguin<sup>1</sup>, Koceila Zidi<sup>2</sup>, Baptiste Darbois-Texier<sup>3</sup>, and Georges Gauthier<sup>4</sup>

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The rheology of dense particle suspensions has been extensively studied in simple shear configurations and its validity for describing suspension flows in complex configurations is now crucial. In this context, we study experimentally the dynamics of a sphere that sediment under its own weight in a dense suspension as it approaches a horizontal wall and squeezes the suspension. Away from the bottom wall, at low Reynolds numbers, we observe that the sphere settles at the same velocity in a Newtonian fluid and in a suspension of equivalent effective viscosity according to Marron and Pierce law. In the near-wall region, at a distance considerably less than the sphere's radius, we observe that the sedimentation velocity of the sphere in a suspension differs from that in a Newtonian fluid. In a Newtonian fluid flowing at low Reynolds number, the settling velocity of a sphere evolves linearly with the distance from the bottom wall, with zero impact velocity at the moment of contact. This behavior is in agreement with lubrication theory. In the case of a suspension flowing at low Reynolds number, the sedimentation velocity of a sphere evolves a function of suspension volume fraction, sphere diameter and grain size. Our data adopt the same general behavior and follow a unique scaling law. We show that the characteristic velocity is the Stokes velocity and determine a characteristic length dependent on the parameters studied. The non-linear aspect of the approaching dynamic of the sphere cannot be approached with a continuous description of the suspension.

#### Wednesday 4:00 LT 18 (Level 8) / Track 4

## A minimal continuum model of clogging in spatio-temporally varying channels

Anushka Herale<sup>1</sup>, Duncan Hewitt<sup>2</sup>, and Philip Pearce<sup>1</sup>

<sup>1</sup>Dept. of Mathematics, University College London, London, United Kingdom; <sup>2</sup>DAMTP, University of Cambridge, Cambridge, United Kingdom

Particle suspensions in confined geometries exhibit rich dynamics, including flowing, jamming, and clogging. It has been observed that jamming and clogging in particular are promoted by variations in channel geometry or fluid material properties - such variations are often present in industrial systems (e.g. local confinements) and biological systems (e.g. stiffening of red blood cells in deoxygenated conditions in sickle cell disease). The aim of this talk is to shed light on the macroscopic dynamics of particulate suspensions in these conditions. To this end, we present

SG12

a continuum two-phase model of particle suspensions based on granular rheology that accounts for spatio-temporally varying material properties or channel geometries. The model comprises a continuous particle phase which advects with flow and has material properties dependent on the particle volume fraction, and a suspending fluid which flows through the particle phase obeying Darcy's law. We solve the system using a finitevolume method and simulate the evolution of an initially uniform particle density. We find that varying material properties and varying geometry can induce heterogeneity in particle volume fraction. We are able to show the emergence of high and low particle density regions in volumedriven flows. These results clarify how spatial variation in material and channel properties can contribute to clogging of particle suspensions.

## Wednesday 4:20 LT 18 (Level 8) / Track 4

SG13

## Rheological Exploration of Irregular Porous Particle Suspensions in Newtonian Solvents: Characterization, Interactions, and Microstructural Evolution

Johanna Vargas<sup>1</sup>, Claudia Carotenuto<sup>2</sup>, Farid B. Cortés<sup>3</sup>, Camilo A. Franco<sup>4</sup>, and Mario Minale<sup>5</sup>

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Porous particles are widely used in the field of drug delivery, adsorption process, and energy storage. The present work focuses on the rheology of non-Brownian suspensions of porous silica particles immersed in a Newtonian Polyisobutene (PIB). Three different porous particles are used, and the results are compared with those obtained on suspensions of irregular silica particles and of hollow glass spheres. Hollow glass sphere suspensions show a Newtonian behaviour for volume fractions up to 0.4, thus indicating a nice dispersion of the silica spheres in the PIB, thanks to a nice solvent particle interaction. Conversely, above a critical concentration, the porous particles and irregular silica particles suspensions show shear thinning behaviour and the presence of a yield stress. The critical concentration, volume fraction, of the irregular silica particles suspensions is much higher than that of the porous particles, 0.2 vs. 0.02. The irregular suspensions behaviours collapse on a master curve, if the volume fraction is corrected to account for the polymer adsorbed on the particles. Two regimes are identified, a collisional, at high shear rates, and an adhesional one, at low shear rates, where a percolated microstructure is detected. In the collisional regime, where particle contact forces dominate, porous particle suspensions, the adhesional regime is enhanced because the lubrication forces are reduced by and additional drainage mechanism: the fluid flow through the porous particles. This additional mechanism is also responsible of the reduction of the critical volume fraction for the shear thinning behaviour of the porous particle suspensions.

## Wednesday 4:40 LT 18 (Level 8) / Track 4

**SG14** 

# Ultrasonic investigations of magneto-rheological fluids for use in ultrasonic metamaterial systems <u>Richard L. Watson</u>

#### School of Engineering, University of Warwick, Coventry CV4 7AL, United Kingdom

Magneto-Rheological Fluids (MRF's) are a suspension of magnetic particles in a carrier fluid. Application of a magnetic field to them induces the magnetic particles to align and form structures along magnetic field lines. These induced structures result in significant rheological changes to the fluid and thus the response of the fluid. Application of sufficiently strong magnetic fields can result in an MRF behaving as an elastic solid. MRF's are active fluids which respond to an external stimulus to control their properties. Ultrasonic metamaterials are structures composed of many component parts, which as an ensemble result in properties which are typically not found in nature. They can be resonant structure or scattering structures to achieve this. This work focusses on Phononic crystal structures, which are arrays of scattering structures with the diameter and spacing of the structure controlling the frequency response of the metamaterial. The other component of the system is the fluid through which the ultrasound propagates. The acoustic impedance difference (c.f. refractive index in optics) between the fluid and scattering structures controls the metamaterial frequency response of the metamaterial swill allow control of the acoustic impedance difference, and as such allow external tuning of the frequency response of the metamaterial system. Characterisation of the MRF suspension and its properties when under magnetic fields and the impact on its ultrasonic properties are an interesting and potentially useful approach in the development of active metamaterial systems.

## Symposium BL

## **Bio-rheology, Living and Active Matter**

Organizers: Laura Casanellas, Alexander Morozov and Christian Wagner

Wednesday 1:50 LT 23 (Level 8) / Track 5

Understanding the rheology of semi-interpenetrating hydrogels

Itziar Insua, Mercedes Fernández, Itxaso Calafel, and Robert Aguirresarobe

POLYMAT and Department of polymers and advanced materials, University of the Basque Country, San Sebastián, Guipúzcoa 20018, Spain

This study explores the rheological behaviour of semi-interpenetrating hydrogels composed of polyacrylamide (PAM) and hyaluronic acid (HA). The synthesis of these hydrogels involves the network formation of PAM and HA, resulting in a unique combination of mechanical and biological properties. The incorporation of hyaluronic acid, known for its biocompatibility and bioactivity, introduces biofunctionality to the hydrogels, making them suitable for biomedical applications. This combination opens up the catalogue of hydrogels for biomedical applications, especially those that need greater mechanical properties, which is usually the drawback of common hydrogels. The rheological response of the semi-interpenetrating hydrogels investigated under various conditions, including linear and non-linear tests. The interplay between the PAM and HA networks is analysed to understand the internal structure of the hydrogels rheological response, influenced by the concentration of acrylamide, crosslinking density, and, more interestingly, the HA concentration. High molecular weight HA in water has three different regions: diluted, semidiluted unentangled, and semidiluted entangled regions. Formulations have been prepared in each one of these regions to study the effect of the HA physical network on the formation of the semi-interpenetrated structure. The synthesis of the hydrogels involves additive manufacturing by VAT 3D printing. Overall, this investigation contributes to the fundamental understanding of the rheology of semi-interpenetrating hydrogels, offering insights into their design principles.

#### Wednesday 2:10 LT 23 (Level 8) / Track 5

BL8

## Adapting a Gaussian networking theory to model the dynamics of cross-linking within the cytoskeleton Nadine du Toit and Kristian K. Müller-Nedebock

Department of Physics, Stellenbosch University, Stellenbosch, Western Cape 7602, South Africa

Cytoskeletal filaments are characterised, not only by their individual mechanical properties and roles within the cell, but also according to the manner in which they assemble and couple to one another to form networked structures. These networks contribute to transport and mechanical properties within cells [see e.g., 1], as well as (via coupling through the cell wall or membrane) to the general properties of the tissues in which they are embedded. A thorough understanding of the network dynamics of the cytoskeleton could therefore lead to significant insights pertaining to a variety of related properties and processes. This talk will focus on one particular mechanism by which these networks may form, namely, cross-linking of filaments to one another. An analytical approach towards modelling a system consisting of filaments or polymer chains and generic cross-linker particles, consisting of two beads joined by a spring, will be presented. The discussion will outline the adaptations introduced into an existing Gaussian networking theory [2] in order to dynamically model the attachment of the cross-linkers to the beads of the polymer chains. Throughout this discussion, various possible implementations of the model will be considered and various physical quantities and averages that are calculable within the formalism will be highlighted. It will be shown that the adapted Gaussian networking theory can be readily combined with field theoretical formalisms that model the dynamics of the cross-linkers and of the chains themselves. This approach thereby couples the Langevin dynamics of the filaments to one another via the dynamics of the bead-spring style cross-linkers. The talk will be concluded with a look at initial computer simulations of the system, complementing the analytical approach.

S. Fürthauer and M. J. Shelley Annu. Rev. Condens. Matter Phys. 13, 365 (2022).
S. F. Edwards, J. Phys. Fr. 49, 1673 (1988).

#### Wednesday 2:30 LT 23 (Level 8) / Track 5

**Impact of disassembling factor - cofilin - on dense branched actin cytoskeletal networks mechanics** Magdalena Kopec<sup>1</sup>, Julien Heuvingh<sup>2</sup>, and Olivia du Roure<sup>3</sup>

<sup>1</sup>PMMH, ESPCI Paris, PARIS, France; <sup>2</sup>PMMH, ESPCI Paris, PARIS, France; <sup>3</sup>PMMH, ESPCI Paris, PARIS, France

Actin is a protein which self-assembles into highly dynamic filaments organized within the cells in different kind of networks. The combination of mechanics and dynamics of these networks enables the cell to achieve essential processes like migration, deformation or integration of external mechanical cues. Here we investigated the impact of cofilin, one of the main disassembly factor, on the mechanical properties of actin networks using superparamagnetic cylinders [1]. These cylinders allow rheology of dense actin meshwork - with architecture similar to the ones found in cells - to be studied. Cofilin renders the networks much softer but does not induce full breakage of the structure. Instead, the networks come back to their original length after mechanical compression. The mechanical response is highly non linear, independently of the presence of cofilin. However, in its presence, some filaments break as proven by the regrowth of the networks after deformation. This property of repolymerization after application of mechanical stress could be of great use for different cellular processes.

BL7

BL9

## Wednesday 2:50 LT 23 (Level 8) / Track 5

Time Resolved SAXS and Rheology Reveal Key Formation Mechanisms of Folded Protein Networks

<u>Matt D. Hughes</u><sup>1</sup>, Sophie Cussons<sup>2</sup>, Ahmad Boroumand<sup>3</sup>, David J. Brockwell<sup>4</sup>, Arwen Tyler<sup>5</sup>, and Lorna Dougan<sup>6</sup> <sup>1</sup>School of Physics and Astronomy, University of Leeds, Leeds, West Yorkshire LS2 9JT, United Kingdom; <sup>2</sup>Astbury Centre for Structural Molecular Biology, University of Leeds, Leeds, West Yorkshire LS2 9JT, United Kingdom; <sup>3</sup>School of Physics and Astronomy, University of Leeds, Leeds, West Yorkshire LS2 9JT, United Kingdom; <sup>4</sup>Astbury Centre for Structural Molecular Biology, University of Leeds, Leeds, West Yorkshire LS2 9JT, United Kingdom; <sup>5</sup>School of Food Science and Nutrition, University of Leeds, Leeds, West Yorkshire LS2 9JT, United Kingdom; <sup>6</sup>School of Physics and Astronomy, University of Leeds, Leeds, United Kingdom

The correct self-assembly and formation of hierarchical structures of biomolecules is fundamental to life, from the construction of cell walls to the spontaneous formation of fibrin blood clots. Despite the ubiquity of these networks throughout living systems, the physical mechanisms behind their formation and self-assembly remain poorly understood. Here, we use photochemically cross-linked folded protein hydrogels as a model protein network system, with a combined time-resolved rheology and SAXS approach to gain a cross-length scale understanding of the key formation mechanisms of network formation. Using bespoke *in situ* illumination rigs at the Diamond Light Source, UK and on a pseudo-controlled rheometer, to initiate photo-chemical crosslinking during both SAXS and rheological measurements we can directly monitor the structural/mechanical formation of the protein networks. Rheological time-sweeps during formation mode is shown to have a lag time of  $6.87 \pm 0.02$  mins with a max growth rate of  $2.8 \pm 0.1$  Pa/s, while the second mode exhibits slower kinetics with a lag time of  $10.48 \pm 0.03$  mins and a max growth rate of  $1.29 \pm 0.05$  Pa/s. Time-resolved SAXS experiments reveal that these two distinct formation mechanisms are: a primary formation, where new protein molecules form new, irreversible chemical crosslinks to create the initial network scaffold; and a subsequent secondary formation phase, where additional new chemical crosslinks form within the initial scaffold, leading to a denser network structure. Understanding these two mechanisms as the origin of the structural and mechanical properties of protein networks opens novel routes to altering network formation, such as in the bundling of fibrin fibers in blood clots or the clustered architecture of folded protein biomaterials.

## Wednesday 3:40 LT 23 (Level 8) / Track 5

## Protein-Functionalised DNA Nanostar Hydrogels

BL11

Giorgia Palombo<sup>1</sup>, Yair Augusto Gutierrez Fosado<sup>2</sup>, and Davide Michieletto<sup>3</sup>

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Active gels are innovative materials that consume energy to actuate and change their material properties. The Watson-Crick base pairing makes DNA an ideal building block for engineering intelligent biomaterials that are easy to manipulate mechanically and topologically[1]. Recently there has been an increasing interest in hydrogels made of DNA star-like molecules called DNA nanostars (DNAns). They have been widely studied as prototypical examples of reversible gels, attracting interest in fundamental[2][3] and applied Soft Matter[4]. Nevertheless, their functionalisation using proteins has been overlooked so far.

In this talk, I will share my recent experimental and computational efforts to create a DNAns hydrogel whose degradation and viscoelastic properties can be modulated in time using different enzymes. More specifically, I will discuss our DNAns design and (micro)rheology experiments to characterise the time-varying viscoelasticity of these hydrogels for different restriction enzymes. Additionally, I will discuss our modelling using oxDNA to link the microscopic choice of DNA sequence and DNAns design to the mesoscopic material behaviour of the gel.

Surprisingly, we discover that specific restriction endonucleases are not able to digest and degrade the gel phase, despite the presence of their recognition sequence on the DNAns arms. On the contrary, unspecific endonuclease, such as DNase, can degrade the gel within hours. Our results should be important to design DNA-based scaffolds and drug-delivery systems with continuous time-varying rheology.

## References

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- [2] S. Biffi et al., PNAS, 110, 39 (2013).
- [3] N. Conrad et al, PNAS, 116, 15 (2019).

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### Wednesday 4:00 LT 23 (Level 8) / Track 5 Challenges of injectable hydrogel characterization

BL12

## Anna Ryl and Piotr Owczarz

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One of the challenges of today's world is the search for modern methods of treatment and recovery from disease and injury. A promising solution is hydrogel polymer matrices, whose potential applications include injectable drug carriers, scaffolds for cell culture, and matrices fabricated using 3D printing. Surprisingly, despite the many papers devoted to these systems, the effect of high shear rates occurring during injection application on the structure and spatial conformation of the lattice is very rarely addressed. Ultimately, the only criteria ensuring the injectability of shear-thinning hydrogels and thermosensitive sols are: shear-thinning nature defined in the range of moderate shear rates and remaining in the liquid sol phase during administration, respectively. For this reason, based on our own research and a few literature sources presented for shear-thinning

hydrogels and polymer sols undergoing thermoinduced sol-gel phase transition, multi-interval research procedures combining capillary and rotational rheometry techniques will be proposed, which will reproduce the conditions of actual application of injectable hydrogels. Instrumental injectability tests using testing devices will also be presented, the results of which will be compared with panel tests presented for injectable medical formulations. Moreover, the risk of incorrect characterization of thermosensitive polymer sols will be discussed if the conditions of actual injection application as well as the aggregation conditions in the peri- and orthokinetic aggregation regime are not taken into account. Finally, the impact of injection application on the mechanical properties of the produced hydrogels will be discussed through the analysis of multi-interval thixotropic tests as well as texture profile analysis of hydrogels after injection through needles.

#### Wednesday 4:20 LT 23 (Level 8) / Track 5

## Human semen viscoelasticity and its relation to male infertility

<u>Giovanna Tomaiuolo<sup>1</sup></u>, Valentina Preziosi<sup>1</sup>, Stefano Guido<sup>1</sup>, Giovanna Tomaiuolo<sup>2</sup>, and Stefano Guido<sup>2</sup>

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Infertility is a growing worldwide problem affecting 15% of couples of reproductive age worldwide and bringing a number of psychological and economic burdens. Although infertility has been mostly attributed to female factors for cultural reasons, about 30% of infertility cases are related to male factors. In addition, a progressive decline of male fertility as well as semen quality, in terms of sperm concentration and motility, has been found in the last years (1). In spite of this, advances in male infertility diagnosis and treatment have been rather limited, with only few techniques translated from research to clinical practice. Although the rheological behavior of human semen is considered as relevant in the assessment of male fertility, the clinical practice is focused on a qualitative measure of semen viscosity, while its elastic properties remain overlooked and essentially unexplored in the scientific literature (2). Here, we provide for the first time a quantitative experimental evidence that normospermic human semen behaves as a viscoelastic fluid and can be characterized by oscillatory shear rheometry in terms of the elastic and viscous moduli measured in the linear regime. In spite of the large variability among different samples, the shear moduli can be scaled according to a weak-gel model, providing a new insight on semen rheological behavior (3). The impact of these findings is highlighted by the negative correlation between sperm motility and both the elastic and the viscous modulus at an oscillatory frequency comparable to that of spermatozoa flagellar beating. This dependence suggests a more rigorous and physiologically-relevant approach to characterize semen rheology and opens up new avenues for future research on the role played by viscoelasticity on male fertility analysis. (1) G. Mendeluk, et al., Journal of andrology, 21 (2000). (2) G. Tomaiuolo, et al., Interface focus, 12 (2022) (3) G. Tomaiuolo et al., Soft Matter, 19 (2023)

#### Wednesday 4:40 LT 23 (Level 8) / Track 5

#### The study of the human tear film: a microrheological approach

<u>Andres Cardil Tornos</u><sup>1</sup>, Javier Ramos<sup>1</sup>, Mercedes Fernández<sup>2</sup>, Itxaso Calafel<sup>2</sup>, Arantxa Acera<sup>3</sup>, and Juan Francisco Vega<sup>1</sup> <sup>1</sup>BIOPHYM, Instituto de Estructura de la Materia, CSIC, Madrid, Spain; <sup>2</sup>POLYMAT and Department of Advanced Polymers and Materials, University of the Basque Country, San Sebastián, Gipuzkoa 20018, Spain; <sup>3</sup>Department of Cell Biology and Histology, University of the Basque Country, Gipuzkoa, Spain

We have investigated the rheological properties of natural human tears and a variety of artificial eye drops containing hyaluronic acid (HA). Dynamic light scattering-based passive microrheology ( $DLS\mu R$ ) was employed to determine the viscoelastic properties of these samples. This technique enables the exploration of the viscoelastic properties of soft materials by tracking the motion of tracer nanoparticles embedded within the sample [1]. Our work demonstrates the applicability of  $DLS\mu R$  in studying the viscoelastic properties of low-volume and complex bio-fluids as natural tears. Utilizing the Cox-Merz rule, we obtained the Newtonian viscosity and characteristic relaxation time for each sample, providing insights into the hydrodynamic size of macromolecules, their concentration in the solution, and an approximation of the complexity of relaxation mechanisms.

We conducted a comprehensive analysis of the frequency dependence of complex viscosity at temperatures ranging from 278 K to 313 K to understand the viscoelastic behaviour across the entire applicability range. Both natural and artificial tears exhibit non-Newtonian behaviour, with significant distinctions observed among the characteristics of different systems. This study serves as a gateway to characterizing the viscosity of the human tear film, offering insights into various eye conditions such as dry eyes [2,3].

1 T. M. Squires, T. G. Mason, Annual Review of Fluid Mechanics. 42, 413 (2010).

- 2 P. Aragona, et al., Translational Vision Science & Technology. 8, 2 (2019).
- 3 A. Blasco-Martinez, et al., Optometry and Vision Science. 99, 455 (2022).

**BL13** 

**BL14** 

## Symposium SV

## **Soft Solids and Viscoplastic Fluids**

Organizers: Maria Charalambides, Maziyar Jalaal and Ian Wilson

## Wednesday 1:50 LT 11 (Level 10) / Track 6 Keynote Memory effects and delayed yielding in amorphous materials Suzanne M. Fielding

Physics, Durham University, Durham, United Kingdom

Amorphous solids include soft materials such as emulsions, foams, colloids, granular matter, and gels, as well as harder metallic and molecular glasses. In contrast to conventional crystalline solids, the internal arrangement of their constituent microstructures (emulsion droplets, foam bubbles, etc.) lacks any intrinsic order. Understanding the rheological (deformation and flow) properties of these materials thus poses a considerable challenge. Typically, they behave elastically at low loads then yield plastically at larger loads. In this talk I shall summarise recent progress in understanding the rheology of these amorphous materials, with a particular focus on complex material memory effects, and on the yielding transition between an initially solid-like state and a finally fluidised one.

## Wednesday 2:30 LT 11 (Level 10) / Track 6

SV8

SV9

SV7

Elasticity Meets Viscoplasticity: A tensorial, Yield-Stress free model for simple yield stress fluid

Gabriele Pagani, Theo Tervoort, and Jan Vermant

Materials Department, ETH Zürich, Zürich, Switzerland

Elasto-viscoplastic (EVP) fluids are characterised by a solid dominated behaviour at small stresses and a strain-rate dependent fluid behaviour at high shear rate. It is custom to model this behaviour with models that contain a binary threshold called yield-stress, e.g. the Herschley-Bulkley model. However, the physical existence of this threshold can be questioned as soft solids can deform elastically [1]. A new elasto-viscoplastic model without a yield stress was recently proposed [2]. The formulation resembles a single mode Maxwell model for viscoelastic materials, with the introduction of a stress-dependent viscosity to capture viscoplastic behaviour. A Carbopol 940 dispersion was chosen as typical material of simple-yield stress fluids (i.e. non-tixotropic EVP fluids that exhibit simple plasticity). The transition from elastic to plastic was properly described by a stress activated relaxation time using a Ree-Eyring model. Shear start-up experiments were performed to fit the model and obtain the steady state behaviour. To assess the tensorial response of the material we first studied the evolution of the first normal stress difference in shear flow. The first normal stress difference follows the predicted simple plastic response and can be only partially captured by the model, due to noisiness in the measurements. Uniaxial compressions at constant strain rate were used to further test the model in different deformation modes. Whereas kinematic hardening was observed, the Von Mises criterion for plasticity was found to hold for the studied material and this transition was also predicted nicely by the constitutive model, using only shear data as input. [1] D.Bonn et al., Yield stress materials in soft condensed matter, Rev.Mod.Phys., 2017 [2] G.Pagani et al., No yield stress required: stress-activated flow in simple yield-stress fluids, in press, JOR

## Wednesday 2:50 LT 11 (Level 10) / Track 6

#### Yielding and double yielding through the lens of recovery rheology

Simon A. Rogers, James J. Griebler, and Krutarth Kamani

University of Illinois at Urbana-Champaign, Urbana, IL 61801-3602, United States

Yield stress fluids are often thought about in a piecewise manner as behaving like elastic solids below the yield stress and like generalized Newtonian liquids above it. Accurate determination of the yield stress is therefore crucial to understanding and predicting the behaviour of yield stress fluids such as inks for 3D printing, foods and cosmetics, muds and soils, and many industrially and biologically relevant materials. Despite the centrality of the yield stress concept, there exist multiple methods by which researchers determine 'the' yield stress, and they can provide values that are orders of magnitude apart. Such discrepancies have even led to some asking the question of whether the yield stress exists at all. In this work we study a series of yield stress fluids using recovery rheology concepts, where strain is decomposed into recoverable and unrecoverable components, and present evidence that yielding takes place gradually over a wider range of stresses than previously thought. It is shown that the overshoot in the loss modulus that has often been used as a measure of yielding is due to the acquisition of unrecoverable strain. It's shown how these measurements led to the development of a model that describes spatially heterogeneous yielding in a mean-field manner and how accurately this model predicts the rheology of simple yield stress fluids. In more complex materials, responses are observed with two overshoots in the loss modulus, a phenomenon often referred to as "double yielding". It is shown that these dual features are really a combination of two distinct processes, only one of which is yielding. Recovery rheology is then used to critically assess the question of how best to define the yield stress, and it is shown that all common metrics can be obtained from our simple model, with a single yield stress, taking into account the kinematics of the particular experiments.
# Wednesday3:40LT 11 (Level 10) / Track 6SV10Insights intoHydroxyPropyl(Methyl)Cellulose (HPMC) hydrogels: investigating degree and molar substitutioneffects via SAOS and LAOS experiments

Saray Perez-Robles, Claudia Carotenuto, and Mario Minale

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Hydroxypropyl(methyl)cellulose (HPMC) is made by etherification of natural cellulose, giving the polymer the ability to form films, bind, retain water, and provide thickening properties. Its solubility, viscosity, and gelation behavior can be adjusted based on the molar and degree of substitution (MS and DS, respectively) of hydroxypropyl (HP) and methoxy (Me) groups per anhydroglucose ring along the cellulose backbone. Due to its versatile properties, it is commonly used in various industries such as pharmaceuticals, food, construction, and cosmetics. Aqueous HPMC solutions undergo reversible thermogelation, i.e. become a gel when heated and transform back to sol state upon cooling. With temperature, the interactions between the HP and Me functional groups increase leading to polymer dehydration, phase separation, and gelation. Thus, we studied the effects of DS (ranging from 1.7 to 2.1) and MS (from 0.1 to 0.3) on the thermogelation of HPMC aqueous solutions using an ARES-G2 rheometer (TA Instrument) equipped with a Couette geometry. Classical SAOS experiments were first run to find the differences between five HPMC polymer samples. Frequency sweeps, time sweeps, and multiwave thermal ramps were conducted between 30 and 90 °C heating the solution at 1°C/min. Furthermore, LAOS tests were performed at different key temperatures to delve deeper into the microstructure evolution of the hydrogels. The Winter-Chambon and Fredrich-Heymann criteria were used to estimate the gel strength at the gel point and gel phase using the multiwave data. The Ewoldt et al.[1] theory was used to define strain stiffening/softening and shear thickening/thinning zones through the thermogelation. Our results show differences in the transition temperatures (beginning of the phase separation and gelation), gelation kinetics, and gel microstructure due to different DS and MS content. Reference [1] R.H. Ewoldt, A. Hosoi, G.H. McKinley, Journal of Rheology, 52 (2008) 1427-1458.

#### Wednesday 4:00 LT 11 (Level 10) / Track 6

SV11

# Scaling laws for near-wall flows of thixo-elasto-viscoplastic fluids based on high resolution OCT-based velocimetry

Ases Akas Mishra<sup>1</sup>, <u>Kasra Amini</u><sup>2</sup>, Amit Kumar Sivakumar<sup>3</sup>, Dragana Arlov<sup>4</sup>, Outi Tammisola<sup>2</sup>, Fredrik Lundell<sup>2</sup>, and Roland Kádár<sup>1</sup>

<sup>1</sup>Chalmers University of Technology, Göteborg, Sweden; <sup>2</sup>Dept. of Engineering Mechanics, KTH Royal Institute of Technology, Stockholm, Sweden; <sup>3</sup>Department of Chemistry and Chemical Engineering, Chalmers University of Technology, Gothenburg, Sweden; <sup>4</sup>Tetra Pak Processing Systems, Lund, Sweden

The recovery process from the previous breakdown of microstructure varies depending on the physiochemical properties of the material and unfolds over timescales. In instances where this timescale equals or surpasses that of the relevant flow, the system's current state dependence on its shear history is termed thixotropy. This research employs Doppler Optical Coherence Tomography (D-OCT) as an innovative, non-intrusive flow velocimetry method suitable for investigating the influence of complex rheological properties on the flow characteristics of three thixoelasto-viscoplastic (TEVP) fluids, Yogurt, Carbopol, and Laponite, within a millifluidic rectangular channel. Velocity profiles, with a depth resolution of 2.58 µm, are extracted from the 2D velocity field reconstructed based on the Doppler shift in the back-scattered incoherent light processed by the Michelson interferometer integrated into the apparatus. Furthermore, D-OCT measurements are conducted on pre-stirred fluid samples to examine the impact of structural breakdown on the flow field. The results indicate an expansion of the plug region, extending close to the surface for samples with higher yield stress. Additionally, a more pronounced yielding of the structure occurs with an increase in thixotropy, resulting in a shrinkage of the plug region. Two scaling power laws are established among four proposed non-dimensional number groups describing the state of the fluid/flow system in near-wall regions. These scaling laws are presented as an approach for a priori prediction of near-wall flow properties, specifically wall shear stress and slip velocity, parameters traditionally challenging to measure, solely using the rheological material functions of the fluids. Additionally, an ex-situ rheological measurement protocol, aligned with the flow conditions in the millifluidic channel, provides insights into

Wednesday	4:20	LT 11 (Level 10) / Track 6	SV12

A simple experiment to measure interfacial tension of yield stress materials <u>Paulo R. de Souza Mendes</u>, Eliana P. Marín, Priscilla R. Varges, and Cynthia R. Barreto *Mechanical Engineering, PUC-Rio, Rio de Janeiro, RJ 22793-410, Brazil* 

The measurement of interfacial tension in materials that possess a yield stress is still a scientific challenge to be overcome. The reason is that the yield stress impairs the action of interfacial tension, typically resulting in irregular interfaces. Most methods used to measure interfacial tension rely on a force balance at the contact line that heavily depends on the interface shape. Therefore, the interface shape modification due to the existence of a yield stress precludes the utilization of such methods. In this paper we present a simple experimental method to overcome this challenge. The method consists of employing vertical capillary tubes partially immersed in a yield stress material such that it traverses the interface. This configuration has been used to measure the interfacial tension when none of the fluids possess yield stress, and the measurable quantities are the height of the capillary rise and the contact angle. In our method, we use the same analysis, except that, instead of measuring the contact angle, we impose the interface shape with the aid of a piston that fits into the capillary. Three different "contact angles" are imposed, namely 00, 450 and 900. This last angle eliminates the contribution of interfacial tension to the force balance at the contact line, so that the weight of the column is

supported by the yield stress effect at the column bottom, which is the same for the other "contact angles." Experiments were performed at room temperature for Carbopol aqueous solutions at two different concentrations, in contact with air. The rheology of the Carbopol solutions was previously determined with the aid of rotational rheometry. Wall slip eliminates the viscoplastic contribution to the force balance. The results showed that the method presents good repeatability and is quite simple to be set up and employed.

# Wednesday 4:40 LT 11 (Level 10) / Track 6

#### Effect of aeration on the rheology of viscoplastic fluids

<u>D. Ian Wilson</u><sup>1</sup>, Bart Hallmark<sup>1</sup>, Douglas Gibson<sup>1</sup>, Rubens Rosario Fernandes<sup>1</sup>, Juan Manuel Peralta<sup>2</sup>, and Susana E. Zorrilla<sup>2</sup> <sup>1</sup>Chemical Engineering and Biotechnology, University of Cambridge, Cambridge CB3 0AS, United Kingdom; <sup>2</sup>Instituto de Desarrollo Tecnoogico para la Industria, Universidad Nacional del Litoral, Santa Fe, Santa Fe 3000, Argentina

In a bubbly liquid gas is dispersed as individual bubbles within a viscous liquid phase. Our interest lies in bubbly liquids where the liquid phase is a viscoplastic fluid, so that the addition of gas decreases the density of the material but it still displays viscoplastic characteristics. Examples of such materials include aerated concrete and buttercream icings. We report experimental investigations of the aeration and rheological characterisation of two viscoplastic fluids. The first is a commercial Carbopol suspension, which features polymeric 'sponges' dispersed in water, representative of a large family of elasto-viscoplastic soft solids which are often used as model viscoplastic fluids in the fluid mechanics community. The second is buttercream icing, which is a highly filled suspension of sugar crystals dispersed within a butter matrix (itself an emulsion), at a volume fraction of solids which is associated with jamming behaviour. In both cases we aerate the base fluids using a planetary mixer fitted with a whisk. The aeration profiles exhibit asymptotic behaviour, with the maximum air volume fraction being strongly dependent on the shear rate associated with the whisk element-wall gap. X-ray microtomography imaging of the aerated icings indicate the presence of large voids attributed to the wake of an element passing through the mass. Bubble size and shape distributions are reported. Rheological characterisation included vane tool tests to gauge yielding behaviour. The flow behaviour of the Carbopol suspensions could be studied in standard rotational rheometers, whereas that of the the buttercream icings was investigated using lubricated squeeze flow and upsetting testing. The dependency of the rheological model parameters (Herschel-Bulkley and Bingham, respectively) on the air volume fraction of each is established and compared.

# Symposium NF

# Non-Newtonian Fluid Mechanics and Flow Instabilities

Organizers: Marco Ellero, Stylianos Varchanis and Helen Wilson

# Wednesday 1:50 LT 19 (Level 10) / Track 7

**Unraveling the existence of asymmetric bubbles rising in viscoelastic fluids** Pantelis Moschopoulos, Alexandros Spyridakis, Yannis Dimakopoulos, and <u>John Tsamopoulos</u>

Department of Chemical Engineering, University of Patras, Patras, Greece

The peculiar dynamics of rising bubbles in viscoelastic solutions has stirred a lot of interest in the scientific community. The three phenomena that have been identified over the ongoing 60 years of research are: (a) the velocity jump discontinuity [1,2], (b) the negative wake structure [2], and (c) the knife-edge bubble shape [3]. The knife edge is an astonishing asymmetric bubble shape which forms a sharp, two-dimensional, cusp-like tail in one view, but it transforms to a broad side in the orthogonal view. The first two have been thoroughly investigated. However, the notorious knife edge shape has not been reproduced numerically. Thus, we undertake a novel, fully 3D computational study to examine the buoyancy-driven rise of a bubble in a viscoelastic solution without assuming axial symmetry to allow the development of three-dimensional disturbances. The governing equations are solved numerically using PEGAFEM-V [5]. For the first time, we capture numerically the so-called knife-edge shape obtaining excellent agreement with experiments. We visualize the flow kinematics and dynamics behind the bubble when the knife edge is formed. Our results indicate that an elastic instability is triggered on the locality of the trailing edge altering the flow field, which now resembles the four-roll mill configuration. We define two different quantities, one kinematical and one geometrical, to determine the onset of the elastic instability. Our novel numerical results allow us to postulate a physical mechanism that leads to the breakdown of axial symmetry and the appearance of the asymmetric bubble shape. Supported by HFRI FM17-2309, and EU Horizon 2020, MSCA, N 955605. [1] Astarita and Apuzzo, A.I.Ch.E., 11 (1965) [2] Pilz and Brenn, JNNFM, 145 (2007) [3] Liu et al., JFM, 304 (1995) [4] Fraggedakis et al. JFM, 789 (2016) [5] Varchanis et al. JNNFM, 284 (2020)

Wednesday 2:10 LT 19 (Level 10) / Track 7

# Bursting bubbles in a viscoelastic medium

<u>Vatsal Sanjay</u><sup>1</sup>, Ayush Dixit<sup>2</sup>, Konstantinos Zinelis<sup>3</sup>, Alexandros Oratis<sup>4</sup>, and Detlef Lohse<sup>5</sup> <sup>1</sup>University of Twente, Enschede, The Netherlands; <sup>2</sup>University of Twente, Enschede, The Netherlands; <sup>3</sup>Imperial College London, London, United Kingdom; <sup>4</sup>University of Twente, Enschede, The Netherlands; <sup>5</sup>University of Twente, Enschede, The Netherlands

Bursting bubbles in a viscoelastic medium are ubiquitous, from transporting pathogens through contaminated water to inkjet and the food processing industry. These flows subject the fluid to large deformations, exhibiting a rich phenomenology and resulting in properties intermediate

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**SV13** 

between simple liquids and elastic solids. In the continuum description of these flows, the conformation tensor is the order parameter for polymeric stretch. These polymers stretch and compress following the flow, evolving to their base state at long times. In leading order, a convenient way to describe this evolution is using the Oldroyd-B model comprising a linear relaxation formulation. As the polymers evolve with the flow, they apply restorative stress. Here, we extend the direct numerical simulations with the volume-of-fluid method to include polymeric flows. We build upon the highly efficient open-source free software program Basilisk C to investigate capillary-driven bursting bubbles with polymers. As the bubble bursts, it releases surface energy, resulting in inertial flow focusing on the Worthington jet and ultimately to droplets through the breakup of this jet. For rheologically complex viscoelastic fluids, this inertio-capillary driving can be offset by elastic stresses suppressing the Worthington jet at high enough elasto-capillary numbers. On the other hand, for low elasto-capillary numbers, we dictate the drops ejected from this Worthington jet.

# Wednesday 2:30 LT 19 (Level 10) / Track 7

NF43

# A three-equation shallow-flow model for Herschel–Bulkley fluids. Application to the simulation of natural hazards

Danila Denisenko, Gael Richard, and Guillaume Chambon

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Natural hazards such as wet snow avalanches or mud flows involve free-surface flows of yield-stress materials. Some of these phenomena could be considered as Herschel-Bulkley fluid flows. A new shallow-flow model for this type of material is derived with a consistent asymptotic method in order to simulate disaster events like a dam break upon an inclined topography. The development of the model is based on two steps. At first, the variables of the primitive equations are expanded up to the first order of accuracy both in the sheared and the pseudo-plug layers. A specific regularization of the rheology is used, allowing us to implement a regular perturbation method in the whole domain. However, unlike in classical regularization methods, the material is here characterized by a true yield stress, with a perfectly rigid behavior below the yield point. The expansions thus built provide a smooth velocity field and show a good agreement with experimental data. In the second step, the mass, momentum and energy balance equations are averaged over the depth of the flow. This results to a fully hyperbolic model of three equations for the fluid depth, the average velocity, and a third variable named enstrophy related to the internal shearing of the flow. This model admits an exact work energy theorem. A dam break problem upon an inclined plane is simulated . It is found that the propagation of the fluid front after the dam break demonstrates three main regimes: an inertia-dominated regime, a plasticity-dominated regime, and final stoppage. Comparison with experimental results shows a good agreement, especially for the inertia-dominated regime and for the material shape after stoppage.

#### Wednesday 2:50 LT 19 (Level 10) / Track 7

Contact line instabilities in non-Newtonian drops impacting on a solid surface

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Drop impact on solid surfaces is a phenomenon of great significance in nature and present in a wide range of applications such as heat transfer, inkjet printing, anti-icing, painting and coating. Numerous studies have considered Newtonian drops' impact on different surfaces, from hydrophilic to superhydrophobic. Depending on the wettability, the impact outcome can be divided into deposition, rebound and splashing. Especially at high speeds, the impact can lead to instabilities like fingering, breakup and deposition of small drops on the surface as the contact line recedes. However, instabilities of non-Newtonian drops impacting solid surfaces have still not been fully characterized and understood, especially at higher impact speeds. Several studies have focused on droplet impact of viscoelastic shear-thinning fluids. It has been proposed that during impact, the viscosity of the drop initially decreases, and then returns to its zero-shear viscosity when maximum diameter has been reached. This behavior can affect the extension of the spreading diameter and reduce contact line, which has been proposed as the cause for the suppression of drop rebound. Despite all this insight, the variety of non-Newtonian liquids studied until now on drop impact experiments is very limited. Here, we experimentally study the impact of viscoelastic drops on hydrophobic and superhydrophobic surfaces at higher speeds, triggering three-dimensional instabilities of the interface and contact line, especially in the receding phase. Parameters like the polymeric concentration, Weber number and rheological characteristics of the fluid are varied to identify the instability regimes.

# Wednesday 3:40 LT 19 (Level 10) / Track 7

Rheology of polymer solution determines the early stage of the drop spreading

Peyman Rostami<sup>1</sup> and Günter K. Auernhammer<sup>2</sup>

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Spreading droplets over a solid substrate plays an important role in many industrial and natural systems. These systems are important for applications ranging from printing and coating to agriculture. For low viscosity liquids, such as water, it is well known that spreading consists of two regimes; the inertial dominated regime and the viscous dominated regime. In the inertial regime, the spreading radius increases with the square root of time  $(r \sim t^{(1/2)})$ . After that, the spreading rate decreases to the viscous regime  $(r \sim t^{(1/10)})$  [1]. By increasing the viscosity, e.g. by adding glycerine, the exponent of the drop spreading deviates slightly from half (square root of time law). This early phase of spreading occurs in a few milliseconds, resulting in a high spreading velocity and a high shear region near the contact line. We correlate the shear rate-dependent rheological properties of the drop with the dynamics of drop spreading. We use solutions of water and PEO as shear viscoelastic fluids and compare them to Newtonian fluids. For the same zero shear viscosity, viscoelastic (shear thinning) drops spread faster than a Newtonian drop. We discuss the

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influence of molar mass and concentration of the polymer on the dynamics of drop spreading and give a qualitative model to describe the influence of shear rate dependent viscosity on drop spreading. [1] First steps in the spreading of a liquid droplet, Anne-Laure Biance, Christophe Clanet, and David Quéré, Physical Review E, 2004. [2] Spreading of a viscoelastic drop on a solid substrate, Peyman Rostami, Mathis Fricke, Simon Schubotz, Himanshu Patel, Reza Azizmalayeri, Güunter K. Auernhammer, arXiv:2308.14515 [cond-mat.soft], 2023.

# Wednesday 4:00 LT 19 (Level 10) / Track 7

# When is an Elastocapillary Worthington jet formed?

Konstantinos Zinelis<sup>1</sup>, Uddalok Sen<sup>2</sup>, Vatsal Sanjay<sup>3</sup>, Thomas Abadie<sup>4</sup>, Omar Matar<sup>5</sup>, Detlef Lohse<sup>6</sup>, and Maziyar Jalaal<sup>7</sup> <sup>1</sup>Imperial College London, London, United Kingdom; <sup>2</sup>Wageningen University and Research, Wageningen, The Netherlands; <sup>3</sup>University of Twente, Enschede, The Netherlands; <sup>4</sup>School Of Chemical Engineering, University Of Birmingham, Birmingham B15 2TT, United Kingdom; <sup>5</sup>Imperial College London, London, United Kingdom; <sup>6</sup>University of Twente, Enschede, The Netherlands; <sup>7</sup>Van Der Waals-Zeeman Institute, University Of Amsterdam, Amsterdam, The Netherlands

The Worthington jet forms when a liquid droplet impacts on a non-wetting substrate. For low viscosity liquids, the formation of such a jet can lead to a complete rebound of the droplet, which is critical for several deposition processes such as spray cooling, spraying pesticides, and inkjet printing. In recent years, the addition of polymers to the liquids, rendering them viscoelastic, has been a widely applied strategy to supress the undesirable rebound. In this work, we perform drop impact experiments on hydrophobic solid surfaces to investigate the pivotal role of viscoelasticity on the post-impact Worthington jet. Specifically, for dilute polymeric solutions, we observe a characteristic transition from an inertia-capillary to a hitherto unobserved elasto-capillary regime upon impact. Additionally, we study various impact scenarios controlling for inertia and elasticity, and we uncover the key role of arising pinned contact line on the non-wettable substrate due to viscoelasticity, for the development of beads-on-a-string (BOAS) structures. We use direct numerical simulations using the open-source volume-of-fluid code, Basilisk to demystify the experimental observations. We capture the temporal variations of the axial strain-rate and the extra elastic stress contribution. We also provide an analysis of the impact forces for the polymeric drops. Finally, we use the experimental and numerical observations to construct a simplified one-dimensional model to predict the formation of the elasto-capillary Worthington jet in viscoelastic fluids.

# Wednesday 4:20 LT 19 (Level 10) / Track 7

# A Computational Rheology study of the Dripping onto a Substrate (DoS)

Thomas Abadie<sup>1</sup>, Konstantinos Zinelis<sup>2</sup>, Gareth H. McKinley<sup>3</sup>, and Omar Matar<sup>4</sup>

<sup>1</sup>School Of Chemical Engineering, University Of Birmingham, Birmingham B15 2TT, United Kingdom; <sup>2</sup>Imperial College London, London, United Kingdom; <sup>3</sup>Hatsopoulos Microfluids Laboratory, MIT, Cambridge, MA 02139, United States; <sup>4</sup>Imperial College London, London, United Kingdom

The Dripping-on-Substrate (DoS) is a conceptually-simple, but dynamically-complex, technique for the study of the filament thinning dynamics of low viscosity non-Newtonian fluids. It incorporates the capillary-driven thinning of a liquid bridge which forms when a fluid sample of small volume is dispensed from a vertical needle onto a horizontal solid partially-wettable substrate. DoS allows experimentalists to measure the extensional properties, i.e. the extensional viscosity and the relaxation time of the sample, of lower viscosity solutions than is possible with commercially-available capillary break-up extensional rheometers (i.e. CaBER). In this work, we develop axisymmetric numerical simulations of DoS experiments, with the open-source Eulerian code, Basilisk, to optimise and extend the operational limits of this protocol. Specifically, we examine the dependence of the relaxation time measurements on the gravitational effects, expressed through the Bond number, and the substrate wettability, characterised by the macroscopic contact angle. In addition, we highlight the effect that the polymer finite extensibility for weakly-elastic fluids can have on measuring the polymer relaxation time. We also suggest a fitting methodology based on the analytical solution for FENE-P fluids developed by Wagner et al (2015) for improved predictions. Finally, we investigate the role of viscosity and the shear-thinning in simple Power-Law (i.e. Carreau-Yasuda), as well as Giesekus viscoelastic fluids.

# Wednesday 4:40 LT 19 (Level 10) / Track 7

# Flow characteristics of microjets into viscoplastic fluids

Seyedpedram Mousavi<sup>1</sup>, Hossein Hassanzadeh<sup>2</sup>, Faical Larachi<sup>3</sup>, and Seyed Mohammad Taghavi<sup>4</sup>

<sup>1</sup>Laval University, Quebec, Quebec, Canada; <sup>2</sup>Department of Chemical Engineering, Laval University, Québec, QC GIV 0A6, Canada; <sup>3</sup>Laval University, Quebec, Canada; <sup>4</sup>Department of Chemical Engineering, Laval University, Quebec, QC GIV 0A6, Canada

In this study, we investigate the microjet flow dynamics of a miscible jet injected into a tank filled with either a Newtonian or viscoplastic fluid. Due to the significant dimensions of the tank, wall effects are negligible, allowing the jet flow to be assumed to be a free jet. Our research employs various methods, including high-speed imaging, time-resolved tomographic particle image velocimetry, and a simplified model to predict jet penetration depth. Through these techniques, we explore the influence of three key dimensionless numbers, namely, the Reynolds number, the Bingham number, and the viscosity ratio. We identify three distinct flow regimes: anchor, mixing, and fingering, and propose a regime classification based on the Reynolds number and the viscosity ratio. Furthermore, we examine the impact of these parameters on jet penetration depth, jet laminar length, and jet radius. Overall, our findings indicate that increasing the apparent viscosity of the ambient fluid leads to a reduction in jet penetration depth. However, the yield stress of the viscoplastic fluid may alter this trend in specific cases. We also present a comparison between the results of experiments and a model that can crudely predict the depth of a jet penetration while considering the effects of the viscoplastic medium and turbulence in the jet flow domain.

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# Symposium NF

# Non-Newtonian Fluid Mechanics and Flow Instabilities

Organizers: Marco Ellero, Stylianos Varchanis and Helen Wilson

# Wednesday 1:50 LT 24 (Level 10) / Track 8 **Purely-elastic linear instabilities in parallel flows** <u>Helen J. Wilson</u>

#### Department of Mathematics, University College London, London WC1E 6BT, United Kingdom

When we look at purely-elastic instabilities in polymeric fluids, there are several key mechanisms that are fairly well understood. Perhaps the most important is the curved-streamline instability, driven by the coupling between streamline curvature and the first normal stress difference. Equally, the second normal stress difference can cause issues in rheometric flows because of a coupling to curvature. We have a reasonable understanding of shear banding, in which regions of differing microstructure can form. And there is a large body of work on interfacial instabilities, in which some difference of physical properties across a fluid interface can trigger instability. In this talk I will look at instabilities in flows with straight streamlines, with and without interfaces, and highlight some areas where instability mechanisms are really not fully understood.

## Wednesday 2:10 LT 24 (Level 10) / Track 8

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NF7

#### A New Instability in Rectangular Duct Flows of Viscoelastic Fluids: a Time-Resolved Doppler OCT Approach Kasra Amini, Fredrik Lundell, and Outi Tammisola

Dept. of Engineering Mechanics, KTH Royal Institute of Technology, Stockholm, Sweden

A new instability has been experimentally observed in a duct flow of shear-thinning non-dilute solutions of polyacrylamide. The streamwise velocity has been measured by Doppler-OCT at the center of the longer sides of the duct and shows asymmetry with respect to the duct centerline: two maxima closer to the walls and one minimum in its central regions. Moreover, the slower regions near the center of the duct exhibit highly fluctuating velocity variations indicating elastic instabilities. The rectangular cross-section of the duct exposes the flow field to higher shear rates near its longer sides. Due to strong shear-thinning attributes of the fluids a positive-feedback loop leads to an accelerated decrease in local viscosity, along with a capacity of the shear flow to reach higher velocities, and thereby higher shear rates in the vicinity of the longer sides. The heterogeneity of the flow field in terms of the governed local shear rates lead to segmentation of the cross-section, with quasi-nematic phases near the longer sides, reaching towards the central regions, and distinguishable micro-structures, using Polarized Optical Microscopy (POM). The change in the material structure results in an alteration in refractive index for each segment, detectable in the OCT intensity field capturing the tomographic section of the point-wise contrast sources. The footprint of this heterogeneity is observable to the unassisted eye, as a brighter line along the duct in the center of the longer sides, indicating different proportions of the high- and low refractive index regions along various optical paths. The complex interplay between the shear-thinning, elasticity, and inertia leads to formation of whale-fluke resembling curvatures as the boarders of the nematic zones at steady bulk flow regimes. It is shown that with an increase in elasticity, outside these regions, the lower inertia allows for the velocity field to undergo viscoelastic fluctuations and asymmetries, hereby termed the Whale Fluke Instabilities.

Wednesday 2:30 LT 24 (Level 10) / Track 8

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# Fluid fingers formation and Oscillatory Kelvin-Helmholtz Instability in stratified miscible systems under sinusoidal perturbation

Luigi Davide Gala<sup>1</sup>, Daniele Tammaro<sup>1</sup>, Marco De Corato<sup>2</sup>, Gerald Fuller<sup>3</sup>, and Pier Luca Maffettone<sup>1</sup>

<sup>1</sup>Dipartimento di Ingegneria Chimica, Università degli Studi di Napoli Federico II, NAPOLI, Italy 80125, Italy; <sup>2</sup>Aragon Institute of Engineering Research, University of Zaragoza, Zaragoza 50009, Spain; <sup>3</sup>Chemical Engineering, Stanford University, Stanford, CA 94305, United States

Hydrodynamic instabilities, such as the Oscillatory Kelvin-Helmholtz (OKHI) one, can take place both in stratified immiscible and miscible fluid systems, showing qualitatively the same properties. Its onset is related to interfacial properties, and it is governed by imposed perturbation, viscosity ratio between the two fluids, and interfacial tension. The latter has a dumping role for both systems, indeed, in miscible systems, the interfacial concentration gradient generates non-equilibrium stresses, called Effective Interfacial Tension (EIT), capable of mimicking the immiscible interfacial tension. It is not an equilibrium force and tends to disappear while diffusion proceeds. When an oscillatory force unsettles two immiscible/miscible overlaid fluids, the OKHI occurs deforming the initially unperturbed interface as waves. Perturbed miscible interfaces have received little attention so far and are the topic of this work. We here report the observations of a novel radial fingers instability, occurring when a stratified system, formed by polydimethylsiloxane (PDMS) and Acetone, is perturbed by sinusoidal rotation around the central container axis, inducing azimuthal shear stresses on the common horizontal interface. The initial destabilization scenario shows OKHI onset, creating the typical superficial waves. Due to centrifugal force and because the PDSM layer does not cover the whole substrate, the formed waves start to move radially, forming PDMS fingers in Acetone bulk. In our opinion, such dynamics are quite different from those reported to happen during the spin-coating process. Theoretical predictions also support the destabilization scenario, finding a clear relationship between the OKHI and the number of fingers.

Wednesday 2:50 LT 24 (Level 10) / Track 8

Effects of viscoelasticity on the inertial instability and mixing performance in a T-channel geometry

Rebecca J. Hill<sup>1</sup>, Mahdi Davoodi<sup>2</sup>, Claudio P. Fonte<sup>3</sup>, and Robert J. Poole<sup>1</sup>

<sup>1</sup>University of Liverpool, Liverpool, United Kingdom; <sup>2</sup>Schlumberger Cambridge Research, Cambridge, United Kingdom; <sup>3</sup>University of Manchester, Manchester, United Kingdom

The T-channel is known to be one of the most efficient microfluidic mixing geometries. For a three dimensional T-channel geometry consisting of two opposing planar inlet channels that join and turn through 90°, it is well known that, at a critical Reynolds number, we encounter an instability where the flow breaks symmetry but remains steady [1]. Within the symmetric regimes, the mixing is minimal. However, for the engulfment regime, that is, beyond the critical Reynolds number, a significant increase in the mixing quality is observed [2]. Hence, understanding the instability is of utmost importance in this context.

For a Newtonian fluid, the parameters of a T-channel flow have been well considered [3]. Yet the added complexities that arise with non-Newtonian effects are the subject of far fewer studies. In this study, we simulate four non-Newtonian models in a T-channel geometry using a finite volume method. The models considered are Oldrovd-B. Oldrovd-A. Giesekus and Carreau. We note that for Giesekus and Carreau, the parameters are chosen to match the shear viscosity of the models.

From the simulations, it is found that for the Oldroyd-B model, we observe the instability at a slightly lower Reynolds number than that of the Newtonian fluid. Whereas for the Oldroyd-A model, we see an increase in the critical Reynolds number. For the Giesekus and Carreau models, the conclusion is less clear as both may be shown to be stabilising or destabilising depending upon the precise definition of the Reynolds number. Whilst these models exhibit a clear difference in the onset of the instability, a negligible difference is observed in the mixing performance of each fluid for a low elasticity number. However, increasing the elasticity number leads to a variation in the mixing performance across all non-Newtonian models.

[1] N. Kockmann, et al., Proc. SPIE. 4982, 319 (2003).

- [2] S. Thomas, T.A. Ameel, Experiments in Fluids. 49, 1231 (2010).
- [3] S. Camarri, et al., Ind. & Eng. Chemistry Research. 59, 3669 (2020).

#### Wednesday 3:40 LT 24 (Level 10) / Track 8

Recent Advances in Polymer Viscoelasticity From General Rigid Bead-Rod Theory

Alan Jeffrey Giacomin<sup>1</sup> and Mona Kanso<sup>2</sup>

<sup>1</sup>University of Nevada, Reno, NV, United States; <sup>2</sup>Massachusetts Institute of Technology, Cambridge, MA, United States

One good way to explain the elasticity of a polymeric liquid, is to just consider the orientation distribution of the macromolecules. When exploring how macromolecular architecture affects the elasticity of a polymeric liquid, we find general rigid bead-rod theory to be both versatile and accurate. This theory sculpts macromolecules using beads and rods. Whereas beads represent points of Stokes flow resistances, the rods represent rigid separations. In this way, how the shape of the macromolecule affects its rheological behavior in suspension is determined. Our work shows the recent advances in polymer viscoelasticity using general rigid bead-rod theory, including the discovery of the first new materials functions from general bead-rod theory since the first, the complex viscosity of Hassager (1974). These include the steady shear material functions [1], largeamplitude oscillatory shear flow material functions [2], and the steady uniaxial [3], biaxial [3] and planar extensional viscosities [4]. We find each of these material functions to depend upon the same molecular feature: the ratio of the macromolecular moment of inertia about the molecular axis to that about the axes transverse to the molecular axis. We then use these new material functions to bridge the Oldroyd 8-constant framework (and thus all of its many special cases) to general bead-rod theory [5]. [1] Giacomin, A.J. et al., "General Rigid Bead-Rod Theory for Steady-Shear Flow," Phys. Fluids, 35(8), Part 1 of 4, 083111 (2023), pp. 1-9. [2] Myong Chol Pak et al., "Large-Amplitude Oscillatory Shear Flow from General Rigid Bead-Rod Theory," Phys. Fluids, 35(8), Part 2 of 4, 083120 (2023), pp. 1-15. [3] Myong Chol Pak et al., "Steady Elongational Flow from Rotarance Theory," Phys. Fluids, 35(10), Part 2 of 4, 103116 (2023), pp. 1-15. [4] Unpublished. [5] Poungthong, P. and A.J. Giacomin, "Bridging macromolecular theory of polymeric liquids to Oldroyd 8-constant constitutive framework," Phys. Fluids 35(11), 111705 (2023).

# Wednesday 4:00 LT 24 (Level 10) / Track 8

The shear rheology of a suspension of flat elastic particles via molecular dynamics

Luke R. Debono, Helen J. Wilson, and Luke K. Davis

Department of Mathematics, University College London, London, London WC1H 0AY, United Kingdom

We consider a recently developed model for flat elastic particles [1], comprised of three Stokes beads coupled via linear springs attached to sliplinks along the perimeter of the triangle formed by the beads. The model yields an analytic solution for the stochastic equations of motion which allows the computation of a closed constitutive equation for the stress tensor of a suspension of these particles. In particular, the constitutive law is governed by a lower-convected derivative: pointing to the lesser-known Oldroyd-A rheology for deforming surface elements.

We explore, for the first time, the shear rheology of this system via multi-particle collision dynamics implemented in the open-source molecular dynamics package LAMMPS[2]. We measure the dependence of the shear viscosity  $\eta$ , N<sub>1</sub> and N<sub>2</sub> on the shear rate. Following this, we make comparisons to the Oldroyd-A and B rheologies and provide particle-level insights about the evolution of the conformation.

[1] Eggers, J., Liverpool, T. B., & Mietke, A. (2023). Rheology of Suspensions of Flat Elastic Particles. Physical Review Letters, 131(19), 194002. https://doi.org/10.1103/PhysRevLett.131.194002

[2] Thompson, A. P., Metin Aktulga, H., Berger, R., Bolintineanu, D. S., Brown, W. M., Crozier, P. S., in 't Veld, P. J., Kohlmeyer, A., Moore,

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S. G., Dac Nguyen, T., Shan, R., Stevens, M. J., Tranchida, J., Trott, C., Plimpton, S. J., Thompson, A. P., Aktulga, H. M., & Berger, R. (2022). LAMMPS - a flexible simulation tool for particle-based materials modeling at the atomic, meso, and continuum scales. Computer Physics Communications, 271, 108171. https://doi.org/10.1016/J.CPC.2021.108171

#### Wednesday 4:20 LT 24 (Level 10) / Track 8

# **Transitory complex flows in simple shear motion – the influence of the Reynolds number** <u>Corneliu Balan</u>

Hydraulics, NUST Politehnica Bucharest, Bucharest 060042, Romania

The paper is concerned with the numerical modelling of generalized Newtonian and viscoelastic fluids in transitory shear motions. The spacetime dependencies of velocity and shear stress in the planar Couette and Poiseuille flows are studied and compared. The influences of: (i) the Reynolds number, and (ii) the initial and boundary conditions on the unsteady and steady solutions are investigated using the Mathematica software. The focus of the study is the analysis of flow instabilities generated by inertia (Reynolds number dependence) and/or by the presence of yield stress or elasticity in the fluid model (so called: intrinsic material instability). The analyses of the strain- and stress-controlled simulations performed at different initial and boundary conditions offer possible explanations of some spurious data recorded in shear measurements of complex viscoelastic fluids. Comparison with experimental results and numerical simulations performed with Ansys-Fluent code are also presented.

#### Wednesday 4:40 LT 24 (Level 10) / Track 8

NF14

**NF13** 

#### A comparison between the FENE-P and sPTT constitutive models in large-amplitude oscillatory shear Thomas John<sup>1</sup>, Robert J. Poole<sup>2</sup>, Adam Kowalski<sup>3</sup>, and Claudio P. Fonte<sup>1</sup>

<sup>1</sup>University of Manchester, Manchester, United Kingdom; <sup>2</sup>University of Liverpool, Liverpool L69 3GH, United Kingdom; <sup>3</sup>Unilever R&D, Bebington, United Kingdom

The simplified linear form of the Phan-Thien-Tanner (sPTT) and FENE-P models are popular viscoelastic constitutive models for a wide range of materials. These models are interesting since, although they are derived from distinct microstructural theories, they become mathematically identical in steady and homogenous flows when specific model parameters are chosen. Therefore, if fitting either of these models to experimental data using viscometric data (i.e. steady simple shear flow (SSSF) or steady extensional flow), it is impossible to determine which model is more appropriate for the given material. However, even in an Eulerian steady flow, the behaviour of these models can differ from one another due to Lagrangian transients in complex geometries. Therefore, it is important that we understand the differences in the rheological behaviour of these models under LAOS for a range of Deborah (*De*) and Weissenberg (*Wi*) numbers (relating to non-dimensional frequency and amplitude, respectively). We observe that the FENE-P model exhibits strong stress overshoots, whereas the sPTT model does not. We explain the stress overshoots using the evolution equations for the conformation tensor. Moreover, we are able to show that the sPTT model in SSSF. However, the FENE-P LAOS response only scales with its SSSF scaling parameter for large enough values of the extensibility parameter  $L^2$ . We then analyse the responses of both models using the Sequence of Physical Processes (SPP) framework. The SPP results show that FENE-P (and FENE-CR) models exhibit viscous backflow (negative instantaneous viscous modulus) just before the stress overshoot.

# **Thursday Morning**

# Symposium OL

# **Oldroyd Lecture**

# Thursday 8:40 Conference Auditorium / Oldroyd Lecture OL1 Influence of macromolecular architectures and chemical structures on extensional rheology of polymer melts Oint Oint Oian Huang Oint Oint Oint Oint Oint

#### Polymer Research Institute, Sichuan University, Chengdu, China

Measuring the nonlinear rheology of well-defined polymer melts is of great importance to establish the link between macroscopic rheological properties and microscopic molecular structures. Polymers are complex materials. While they may have different chemical structures in their monomers, the monomers can be further organized into many different macromolecular architectures like linear, ring, star, comb, and dendritic shapes during synthesis. It is known that the rheological behaviour of polymer melts is very sensitive to macromolecular architectures. However, experimental data of nonlinear rheology especially the extensional rheology at large strains for model branched polymers is still rare. In addition, the effect of chemical structures of the monomers is not considered in the tube model. A few experiments have already shown that chemical structures may influence the nonlinear extensional rheology significantly. But different explanations such as the flow-induced friction reduction and flow-induced friction enhancement still exist.

In the first part of this work, we study the influence of macromolecular architectures using model polystyrene (PS) melts. We start from the simplest branched architecture, i.e., star PS with one branch point only, and investigate the effect of arm numbers on nonlinear extensional rheology. Then the star PS melts are also compared with H and pom-pom PS melts which have two branch points. In the second part, we fix the macromolecular architectures to linear polymers, and study the influence of chemical structures. We start from comparing PS with other polymers containing aromatic rings in their side groups, where p-p stacking between neighbouring polymer chains may affect the rheological behaviour in extensional flow. Then the PS melts are further compared with other polymers without p-p interactions.

# Symposium PL

# **Plenary Lectures**

# Weissenberg Lecture

# Thursday 9:10 Conference Auditorium / Plenary Lectures How grains flow? The rheology of granular systems, from sand to sticky grains <u>Olivier Pouliquen</u>

PL2

#### IUSTI, CNRS-Aix Marseille University, Marseille, France

Knowing the rheology of granular media submitted to steady, uniform shear is the first step towards developing a hydrodynamics approach of granular flows of interest in many industrial and geophysical applications. In this talk, I will first review the progress made in our understanding of the rheology of simple granular media, introducing the concept of pressure-imposed rheology for dry grains and suspensions. This framework provides a useful basis for tackling more complex granular systems. The case of powders, for which the concept of flowability remains unclear, represents a real challenge. The main difficulty arises from the presence of adhesive forces between the grains, which prevent smooth and continuous flows. To start addressing the challenge of powder rheology, we have studied model cohesive granular materials composed of sticky grains, both in discrete simulations and in experiments. Our results reveal unexpected and complex behaviors compared to simple granular media, showing how a slight change in interparticle interaction can have strong consequences on the bulk rheology.

# Symposium EF

# **Emulsions, Foams and Interfacial Rheology**

Organizers: Simon Cox, Thibaut Divoux and Nick Jaensson

 Thursday
 10:40
 LT 1 (Level 7) / Track 1
 EF7

 Bijel: soft matter or miniature universe? Effects of orientational order on modulated cylindrical interfaces
 R Mike L. Evans<sup>1</sup>, Jason Klebes<sup>2</sup>, and Paul S. Clegg<sup>3</sup>

<sup>1</sup>Mathematics, University of Leeds, Leeds, United Kingdom; <sup>2</sup>Mathematics, University of Leeds, Leeds, United Kingdom; <sup>3</sup>Physics and Astronomy, University of Edinburgh, Edinburgh, United Kingdom

Oil and water don't mix ...or do they? Nanoparticles can sculpt the oil-water interface into a 3D labyrinth, to form a "bijel" (Bicontinuous Interfacially Jammed Emulsion geL). Similarly, amphiphilic molecules can form lipid monolayers when mixed with oil and water, leading to a bicontinuous "sponge phase". Bijels are recently discovered materials with potential applications ranging from energy storage to surgical implants, while lipid-induced bicontinuous emulsions have been known for longer. More excitingly (for a mathematical physicist) the internal structure of any of these labyrinthine interfaces can be modelled using ideas borrowed from the physics of the 4D curved space we call home. So, bijels and sponge phases can give insights into the workings of spacetime and vice versa. We have modelled the tubular interfaces within bicontinuous mixtures. By examining the equilibrium energetics of the structured 2D fluid at the interface, we predict low-temperature morphologies. Like the corresponding dispersed phases (Pickering emulsions and vesicles), these tubular surfaces may host orientational order with discrete rotational symmetries, for example in the pairwise positions of the interfacial particles. We examine how that order interacts with shape modulations of the interface. While on spherical droplets defects are topologically required to exist, the cylindrical topology is compatible with a defect-free patterning. Nevertheless, once an axial modulation is introduced by a mechanism such as spontaneous curvature, nontrivial patterns of order emerge and modify the shape of the tube.

#### Thursday 11:00 LT 1 (Level 7) / Track 1

#### The role of proteins for stabilizing lung surfactant thin liquid films

EF8

Maria Clara Novaes-Silva<sup>1</sup>, Benjamin Buhl<sup>1</sup>, Ainhoa Collada<sup>2</sup>, and Jan Vermant<sup>1</sup>

<sup>1</sup>Materials Department, ETH Zürich, Zürich, Switzerland; <sup>2</sup>Faculty of Biology, Complutense University of Madrid, Madrid, Spain

The alveolar walls are covered by a thin liquid layer, where densely packed structures composed of lipids and proteins are found. These structures unravel at the interface, forming a complex assembly composed of a monolayer attached to multilamellar surfactant reservoirs [1]. The stability of this assembly and its lipid composition are believed to be regulated by lung proteins. They are claimed to create a DPPC-enriched monolayer that can withstand high surface pressures while removing other unsaturated lipids to the surfactant reservoirs, forming a stable 3-D structure [2]. The importance of this mechanism is twofold: on one hand it decreases the energy required for breathing by lowering the surface stress in the alveolar layers; on the other hand, it stabilizes the interfacial structure, preventing monolayer collapse upon the breathing cycles. To probe the stability of lung surfactant thin liquid films and to further investigate this complex interfacial structure, we used a combination of dynamic thin film balance experiments with atomic force microscopy under physiological conditions. We compared a commercial lung surfactant replacement (Infasurf) with pure lipids and lipid-protein mixtures to evidence the role of each component. Our findings show that Infasurf thin films are extremely stable under breathing-like drainages, and we attribute this phenomenon to a line tension driven elasticity. Additionally, we identified that lung proteins promote the formation of structured aggregates after breathing oscillations, that are responsible for this line tension. [1] J. Perez-Gil, Biochimica et Biophysica Acta. 1778, 1676-1695 (2008). [2] J.C. Castillo-Sanchez et al., Archives of Biochemistry and Biophysics, 703, 108850 (2021).

# Thursday 11:20 LT 1 (Level 7) / Track 1

EF9

**Complex Thin Film Morphologies via Multiple Motions of a Bubble Decorated with Surface-Active Molecules** Lorenzo Lombardi, Daniele Tammaro, and <u>Pier Luca Maffettone</u>

# University of Naples Federico II, Naples, Italy

Thin films are present in a diverse array of systems and applications such as foams, coatings, biosensors, and solar cells. The problem of stability and patterning of thin films is central to these applications and extensive research has explored their underlying principles. We show that controlled motion of bubbles against the air-liquid surface enables the creation of complex and stable up to coalescence thin film structures. The experiments are carried out with pure silicone oil and with Bovine Serum Albumine (BSA) in water. In the latter case partial mobility of the interface is expected while in the former case the interface is fully mobile. Only in the case of surface decorated samples we found the formation of either a stable wimple or an unstable wimple that transforms into a dimple by properly adjusting the bubble approach velocity. The wimple morphology features a film with a relatively thin central area surrounded by a thicker rim, which gradually thins near the bubble base. We employ a model based on lubrication theory to elucidate the mechanism behind the formation of these structures. To achieve quantitative agreement between experimental results and model predictions, the model is formulated by incorporating a single adjustable parameter to account for the mobility of the interface. The simulation outcomes indicate the crucial role of a partially mobile interface in the formation and stabilization of the observed complex structures. Patterned surfaces could be manufactured by quenching the film once the desired morphology has been achieved.

# Thursday 11:40 LT 1 (Level 7) / Track 1 **Thin liquid film rupture: Interplay between Marangoni flows and film break up.** <u>Lucas Bidoire</u> and Jan Vermant

Materials Department, ETH Zürich, Zürich, Switzerland

Foams are present in many industrial domains such as food technology, energy, and health care. These are bi-phasic materials separated by thin liquid films. The macroscopic material behavior is related to the thin film lifetime and often drainiage is the rate limiting step. However, despite extensive studies on drainage and rupture, especially the latter process is not fully understood [1]. Clearly there is a complex interplay between hydrodynamic, capillarity, molecular forces (disjoining pressure) and interfacial rheology [2]. Literature reports that, in many surfactant stabilized systems, this complex balance will lead to instabilities and surface property inhomogeneities through the Marangoni effect. Yet, most experimental studies assume that interfaces are homogeneous and Marangoni stresses negligible. Here, using the Dynamic Thin Film Balance apparatus (DTFB), we provide quantitative experimental evidence of the Marangoni effect in draining thin films. Systems studied are air/water interfaces stabilized by simple surfactants such as palmitic acid, hexadecanol and sodium palmitate. The use of a fluorescent probe surfactant (Palmitic acid NBD) leads to visual observation of the surfactant spatial gradient and Marangoni number estimations. The time to rupture is investigated focusing on the applied Capillary number and the measured Marangoni number. In addition, surfactant coverage inhomogeneities and thin film thickness variations are studied through a non-homogeneous disjoining pressure. We believe that this study is the first of its kind and may give an experimental picture of both lateral and normal stresses coupling leading to thin film collapse. [1] E. Chatzigiannakis, et al., Current Opinion in Colloid & Interface Science. 53, 101441 (2021). [2] F. Janssen, et al., Food Hydrocolloids. 116, 106624 (2021).

# Thursday 12:00 LT 1 (Level 7) / Track 1

Coalescence dynamics on a colloid-laden interface - Effect of particle wettability

Simona Migliozzi<sup>1</sup>, Loic Chagot<sup>1</sup>, Correia L. Elton<sup>2</sup>, Eduarda Barros De Oliveira<sup>2</sup>, Sepideh Razavi<sup>2</sup>, and Panagiota Angeli<sup>1</sup> <sup>1</sup>Department of Chemical Engineering, University College London, London, United Kingdom; <sup>2</sup>School of Chemical, Biological and Materials Engineering, University of Oklahoma, Norman, OK, United States

The use of colloids is emerging as a promising strategy to stabilize interfaces. In these systems, the stabilization mechanism relies on the formation of particle networks, which depend on the interparticle interactions taking place. By tuning particle properties such as size, shape and wettability, the interparticle interactions can be modified, inducing complex rheological responses of the particle-laden interfaces. Investigating the dynamic response of these interfaces as they come together is therefore crucial to understand the role of interfacial rheology in the stabilisation mechanism of complex multiphase formulations. In this work, we experimentally investigated the coalescence dynamics of a pendant drop on a liquid-liquid interface stabilised with solid particles with different surface modifications. Using high-speed imaging, we captured the coalescence dynamics at increasing particle concentrations, while details of the microscructure and mechanical dilational response of the coalescence time with particle concentration is dependent on different particle wettability, where more hydrophobic particles appeared to delay the coalescence at increasing concentrations, as opposed to more hydrophilic particles, which accelerated the coalescence dynamics, also inducing a shift from total coalescence to partial coalescence. For all particle types, an increase in the concentration of particles deposited at the interface generated stronger particle climbing at the mother droplet interface, changing the thinning of the liquid neck during the last stages of the coalescence. Overall, our findings demonstrate that the thinning dynamics are strongly influenced by the mechanical properties of the particle network, thus confirming the key role of interfacial rheology in the fast dynamics of Pickering interfaces.

# Thursday 12:20 LT 1 (Level 7) / Track 1

**EF12** 

# Effect of interfacial nanoparticles on the coalescence process of model drop-in-matrix emulsions by rheo-optics and microfluidics

Angela Marotta, Valentina Preziosi, Giovanna Tomaiuolo, and Giovanni Filippone

Dipartimento di Ingegneria Chimica, dei Materiali e della Pr, University of Naples Federico II, Naples 80125, Italy

Understanding how small solid particles alter the morphology of two-phase fluids such as emulsions and polymer blends at the microscale is crucial for controlling the final microstructure and thus most macroscopic properties of such ubiquitous systems. The final morphology of particlecontaining biphasic liquids is the result of the competition between break up and coalescence phenomena that occur during flow. Coalescence is responsible for often undesired coarsening phenomena and there is a great interest in preventing this phenomenon to preserve fine drop-in-matrix microstructures. Coalescence is the result of a sequence of events that starts with the approach of a pair of droplets due to velocity gradients and ends with the drainage and finally the breakup of the thin matrix film between the droplets. The presence of solid particles much smaller than the average size of the droplets can influence each of these steps. Depending on where the particles accumulate (e.g. in one of the two liquid phases or at the drop-matrix interface), different effects have been observed, such as coalescence suppression, drop clustering, disruption of coalescence as well as promotion of coalescence. The microscopic mechanisms underlying these phenomena are still under debate and fundamental studies are needed. Here, we present a thorough rheo-optical study on the effect of small amounts (=5 wt%) of zinc oxide (ZnO) nanoparticles on the coalescence process of dilute emulsions based on polydimethylsiloxane (PDMS) and poly(iso)butylene (PB). An unexpected promotion of coalescence processes. The role of different parameters (nanoparticle content, shear rate, initial drop size) was investigated. It was found that the coarsening is the result of mechanical interlocking between the nanoparticles at the interface connecting the drop size) was investigated. It was found that the coarsening is the result of mechanical interlocking between the nanoparticles at the interface connecting the drop size) was

**EF11** 

# Symposium PF

# **Polymeric Fluids**

Organizers: Francesco Del Giudice, Richard Graham and Evelyne van Ruymbeke

# Thursday 10:40 LT 17 (Level 7) / Track 2 PF11 Universality in the Alignment-Induced Variation of Inter-Chain Interaction in Polymer Melts under Flows Nuofei Jiang<sup>1</sup> and Evelyne van Ruymbeke<sup>2</sup>

<sup>1</sup>IMCN, BSMA, Université catholique de Louvain, Louvain-la-Neuve 1348, Belgium; <sup>2</sup>IMCN, BSMA, Université catholique de Louvain, Louvain, Louvain-la-Neuve, Belgium

Polymer dynamics is universal in the linear viscoelastic regime, while the recent experiments show that the nonlinear extensional rheology of polymer melts is non-universal. In order to understand this transition, in this work we investigated the variation of inter-chain interaction in the coarse-grained (CG) molecular dynamics (MD) simulation systems of unentangle polymer melts under flows in terms of the frictional coefficient, which is thought to be the key factor in explaining the nonuniversality. The frictional coefficient in the simulation system under shear and planar extensional flows are quantified from the two expressions we proposed very recently (Jiang, N.; van Ruymbeke, E., Macromolecules 2023, 56 (8), 2911-2929.). After presenting the comparison between the CG MD simulation and the experimental data, and proving the effectiveness of our expressions, we show that those frictional coefficients can be universally related to the projection areas of the polymer coils, regardless of the large-scale and Kuhn-scale properties of the polymer chains as well as the types of deformation. Our results suggest the universal nonlinear rheology could still exist in different chemistries by carefully taking account of the Kuhn-scale properties.

# Thursday 11:00 LT 17 (Level 7) / Track 2

# Fundamentals of the Oldroyd-B model revisited: Tensorial vs. vectorial theory

Burkhard Duenweg<sup>1</sup>, Aaron Brunk<sup>2</sup>, Joydip Chaudhuri<sup>1</sup>, and Maria Lukacova-Medvidova<sup>2</sup>

<sup>1</sup>Theory Group, Max Planck Institute for Polymer Research, Mainz, Rheinland-Pfalz 55128, Germany; <sup>2</sup>Institute of Mathematics, Johannes Gutenberg University Mainz, Mainz, Rheinland-Pfalz 55128, Germany

The standard derivation of the Oldroyd-B model starts from a coupled system of the momentum equation for the macroscopic flow on the one hand, and Fokker-Planck dynamics for molecular dumbbells on the other. The constitutive equation is then derived via a closure based upon the second moment of the end-to-end vector distribution. We here present an alternative closure that is rather based upon the first moment, and gives rise to an even simpler constitutive equation. We establish that both closures are physically sound, since both can be derived from (different) well-defined non-equilibrium ensembles, and both are consistent with the Second Law of thermodynamics. In contrast to the standard model, the new model has a free energy and a dissipation rate that are both regular at vanishing conformation tensor. We speculate that this might perhaps alleviate the well-known high Weissenberg number problem, i.e. severe numerical instabilities of the standard model at large flow rates. As the new model permits a trivial solution (vanishing conformation tensor, vanishing polymer stress), an extension may be needed, which includes Langevin noise in order to model thermal fluctuations.

# Thursday 11:20 LT 17 (Level 7) / Track 2

# Extensibility governs the flow-induced alignment of polymers and rod-like colloids

<u>Vincenzo Calabrese</u><sup>1</sup>, Tatiana Porto Santos<sup>1</sup>, Carlos G. Lopez<sup>2</sup>, Lettinga Minne Paul<sup>3</sup>, Simon J. Haward<sup>1</sup>, and Amy Shen<sup>1</sup> <sup>1</sup>Okinawa Institute of Science and Technology, Onna-son, Okinawa 904-0495, Japan; <sup>2</sup>RWTH Aachen University, Aachen, Germany; <sup>3</sup>Institut für Weiche Materie Forschungszentrum Jülich, Jülich, Germany

Polymers and rod-like colloids (PaRC) adopt a favorable orientation under sufficiently strong flows. However, how the flow kinematics affect the alignment of such nanostructures according to their extensibility remains unclear. By analysing the shear- and extension-induced alignment of chemically and structurally different PaRC, we show that extensibility is a key determinant of the structural response to the imposed kinematics. We propose a unified description of the effectiveness of extensional flow, compared to shearing flow, at aligning PaRC of different extensibility.

# Thursday 11:40 LT 17 (Level 7) / Track 2

**PF14** 

**PF13** 

**PF12** 

# A variable entanglement density constitutive model for entangled polymer systems from nonequilibrium thermodynamics

Pavlos S. Stephanou<sup>1</sup>, Dimitrios G. Tsalikis<sup>2</sup>, Dimitrios G. Tsalikis<sup>3</sup>, Vlasis G. Mavrantzas<sup>4</sup>, and Vlasis G. Mavrantzas<sup>5</sup> <sup>1</sup>Department of Chemical Engineering, Cyprus University of Technology, Limassol 3036, Cyprus; <sup>2</sup>Department of Chemical Engineering, University of Patras, Patras, Greece; <sup>3</sup>Department of Mechanical and Process Engineering, ETH Zurich, Zurich, Zurich 8092, Switzerland; <sup>4</sup>Department of Chemical Engineering, University of Patras, Patras, Greece; <sup>5</sup>Department of Mechanical and Process Engineering, ETH Zurich, Zurich, Zurich 8092, Switzerland

Detailed nonequilibrium molecular dynamics simulations have shown that under flow, there is a strong reduction in the average number of entanglements per chain, a mechanism known as convective constraint release (CCR). Initially Ianniruberto and Marrucci, and more recently Dolata and Olmsted proposed new theoretical models that account explicitly for flow-induced disentanglement. In this work, we employ the

single-generator bracket formalism of nonequilibrium thermodynamics by Beris-Edwards to derive a similar constitutive model. In the new approach, entanglement destruction is obtained by coupling the entanglement density directly with the velocity gradient and not with the orientation tensor, thus the final set of evolution equations (one for the average number of entanglements per chain, and one for the orientation of entanglement strands) are closer to those reported by Ianniruberto and Marrucci. Direct comparison with nonequilibrium molecular dynamics simulations shows that the model can describe quite accurately the loss of entanglements due to the applied flow for a variety of polymer melts (molecular weights) and flow rates. The new set of equations performs well also in describing experimental rheological data for polystyrene using a more realistic value of the convective constraint release (CCR) parameter, although some discrepancies are observed in the description of the second normal stress coefficient

#### Thursday 12:00 LT 17 (Level 7) / Track 2

# The relationship between the CCR parameter $\beta$ and polymer melt structure

Peter D. Olmsted<sup>1</sup>, Benjamin E. Dolata<sup>2</sup>, Thomas O'Connor<sup>3</sup>, and Marco A. Galvani Cunha<sup>4</sup>

<sup>1</sup>Department of Physics & Institute for Soft Matter Synthesis, Georgetown University, Washington, DC 20057, United States; <sup>2</sup>National Institute for Standards and Technology, Gaithersburg, MD 20899, United States; <sup>3</sup>Materials Science and Engineering, Carnegie-Mellon University, Pittsburgh, PA 19104, United States; <sup>4</sup>Physics, University of Pennsylvania, Philadelphia, PA 15213, United States

We investigate the universality of entanglement kinetics in polymer melts. We compare predictions of a recently developed constitutive equation for disentanglement to molecular dynamics simulations of both united-atom polyethylene and Kremer-Grest models for polymers in shear and extensional flow. We find that the convective constraint release parameter  $\beta$  is independent of molecular weight and thus the number of entanglements, but that it increases with chain stiffness. We posit that  $\beta$ , which is related to the entanglement release rate, has a power-law dependence on the probability that dangling chain ends form binary contacts with the surrounding chains. We show that this implies that  $\beta$  is a function of the ratio  $C_{\infty}^2/N_{eK}$ , i.e. the characteristic ratio and the maximum stretch of an entanglement strand, and we find that the form  $\beta \sim$  $(C_{\infty}^2/N_{eK})^c$  is consistent with the data. These results may help shed light on which polymers are more likely to undergo shear banding.

#### Thursday 12:20 LT 17 (Level 7) / Track 2

PF16

**PF15** 

**Modelling the nonlinear shear rheology: from unentangled to entangled linear polymer melts** <u>Maxime Dalne<sup>1</sup></u>, Salvatore Costanzo<sup>2</sup>, Dimitris Vlassopoulos<sup>3</sup>, Dimitris Vlassopoulos<sup>4</sup>, Michael Rubinstein<sup>5</sup>, and Evelyne van Ruvmbeke<sup>1</sup>

<sup>1</sup>Institute of Condensed Matter and Nanosciences, Université Catholique de Louvain, Louvain-la-Neuve, Belgium; <sup>2</sup>Department of Chemical, Materials and Industrial Engineering, University of Naples Federico II, Naples, NA 80125, Italy; <sup>3</sup>Institute of Electronic Structure and Laser, Foundation of Research and Technology Hellas, Heraklion, Greece; <sup>4</sup>Department of Materials Science and Technology, University of Crete, Heraklion, Greece; <sup>5</sup>Duke University, Durham, NC, United States

The linear viscoelastic response of unentangled melts is well described by coarse grained model such as the Rouse model. However, in the high shear rate regime, the polymer chain tends to orient in the flow direction and the Rouse model is not valid anymore.

In a recent work [1], we proposed a new approach based on the shear slit model [2] to describe the nonlinear shear rheology of unentangled linear polymer melts. Our model introduces the shear strands, which represent the confinement effect induced by the nonlinear shear flow on the polymer chains, as the latter can only equilibrate at length scales shorter than these sections. According to this molecular picture, the overshoot observed in the stress growth coefficient is seen as a transition from the linear envelope to the steady regime, triggered by the progressive confinement of the chains in shear slits. By assuming that the chain confinement starts at the time corresponding to the boundary between affine and non-affine deformation of chain, quantitative agreement was obtained between the experimental data and the predicted curves. We now extend this approach, which is valid when the chain has no or very few entanglements, to describe the flow properties of entangled polymer melts. Based on the same concepts, we consider that, in the steady regime, the inner part of a chain is disentangled and therefore, is no longer confined at a scale of Me but at the scale of the shear strands. This assumption allows us to quantitatively predict the steady viscosity. We then investigate how to account for the progressive disentanglement of the chain with the flow to describe the transient regime.

M. Dalne, et al. "Nonlinear shear rheology of unentangled polystyrene melts", *In preparation*, (2023).
 D. Parisi, et al. "Nonlinear shear rheology of entangled polymer ring", *Macromolecules*, **54** (6), 2811 (2021).

# Symposium SV

# **Soft Solids and Viscoplastic Fluids**

Organizers: Maria Charalambides, Maziyar Jalaal and Ian Wilson

# Thursday 10:40 LT 6 (Level 8) / Track 3

SV14

Experimental and modelling investigation of the mechanical behaviour of chocolate at quasi-static and elevated strain rates

Thrasyvoulos K. Elezoglou<sup>1</sup>, Maria N. Charalambides<sup>1</sup>, and Sunil Rana<sup>2</sup>

<sup>1</sup>Mechanical Engineering, Imperial College London, London, United Kingdom; <sup>2</sup>Mondelez International, Birmingham, United Kingdom

Chocolate is a highly complex, multi-phase composite. Its mechanical behaviour strongly depends on the applied strain-rate. This study determines the behaviour at ambient temperature across a wide strain rate range, to reflect the need for building models for predicting fragmentation during the first oral bite as well as during normal handling and transportation of the product. The latter is important to prevent the development of cracks in the products before they reach consumers and reduce waste. Monotonic compression tests were conducted at constant speeds ranging from 0.001 m/s to 6 m/s, while monotonic tension tests were conducted at 0.001 m/s, and 0.01 m/s. For the high-speed tests, we present an analysis that considers dynamic effects on the recorded raw data and allows determination of the true response of the material. It was found that the strength of chocolate increases with strain rate, while there are different clusters of almost identical Young's Moduli corresponding to low (=0.1 m/s) and high speeds (>0.1 m/s). Cyclic tests showed that the Young's Modulus calculated from the unloading path is 1.5 to 4 times larger than the one calculated from the loading path. Comparisons between quasi-static compression and tension results demonstrated that chocolate is stiffer in tension but stronger in compression. Finite Element Analyses simulated the uniaxial compression tests using several models and showed that the Drucker-Prager hardening model captures adequately and in a unified way the different behaviour of chocolate in compression and tension.

# Thursday 11:00 LT 6 (Level 8) / Track 3

**SV15** 

**SV16** 

# Characterising the mechanical properties of soft solids through acoustics and rheology, exemplified by anhydrous milk fat

Megan J. Povey<sup>1</sup> and Daniel I. Hefft<sup>2</sup>

<sup>1</sup>School of Food Science and Nutrition, University of Leeds, Leeds, Yorkshire and the Humber ls2 9jt, United Kingdom; <sup>2</sup>Product Research Team, Campden Food BRI, Chipping Campden, None Selected GL55 6LD, United Kingdom

Foods vary in their elastic properties over a wide range of behaviours. In the case of mastication, textures vary from hard solid through brittle (chocolate bar) and crispy/crunchy (biscuits) to viscous and extensional flow (syrup) and finally very low viscosity fluid (water). Here we deploy an elastic description of soft solids which embraces all these behaviours to quantify the elastic behaviour of food, in particular through the use of sound. We illustrate the use of this mathematical description in the quantitative characterisation of the elastic and flow properties of food through orthodox measurement techniques and novel ultrasound methods. Measurement is complicated by human sensory capabilities that span the entire range from solid to fluid to gas in an integrated manner, during the appreciation of food. We use acoustic and rheological measurement techniques for the determination of the mechanical properties of soft solids, comparing oscillatory rheometry with acoustic parameters as exemplified by acoustic and oscillatory rheometry measurements in crystallising anhydrous milk fat (AMF). We conclude that acoustic and rheological measurements complement each other with acoustic techniques offering the possibility of inline, in process determination of mechanical and flow

Thursday 11:20 LT 6 (Level 8) / Track 3

# Rheology and yielding transitions in mixed kaolinite/bentonite suspensions

Ahmad Shakeel, Alex Kirichek, and Claire Chassagne

#### Hydraulic Engineering, TU Delft, Delft, The Netherlands

Kaolinite suspensions are known to have quite fast settling behaviour and Newtonian character, particularly at low concentrations, due to the nonswelling nature of their constitutive particles. On the other hand, bentonite is known for its swelling and interacting nature, which leads to the formation of a network structure, even at low concentrations. Therefore, both these clays are known to have significantly different rheological fingerprint. In this study, the yielding transitions in mixed kaolinite/bentonite suspensions have been investigated by varying the kaolinite/bentonite ratio and the total solid content. The detailed rheological analysis of these suspensions was carried out using amplitude sweep tests, frequency sweep tests, stress ramp-up tests and structural recovery tests. The results showed that the kaolinite suspensions exhibited a twostep yielding behaviour in stress and amplitude sweep tests at higher solid content (= 35 wt%). On the other hand, the bentonite suspensions displayed a single-step yielding even at higher concentrations. In mixed kaolinite/bentonite suspensions, a clear transition between a single and two-step yielding behaviour was observed in steady and oscillatory tests as a function of kaolinite/bentonite ratio, for a particular total solid content. The structural recovery experiments showed that for mixed kaolinite/bentonite suspensions at lower total solid content, where two-step yielding was present, the structural recovery was lowest. Furthermore, these results showed that the rheological properties (i.e., moduli) of stable kaolinite/bentonite suspensions were completely recovered to their initial state, even after multiple shearing cycles. The settling column analysis displayed that a very small amount of bentonite was needed to stabilize the kaolinite suspensions, which is necessary for preparing stable clay based formulations.

SV17

# Thursday 11:40 LT 6 (Level 8) / Track 3

Morphology of center plug for viscoplastic Poiseuille flows with superhydrophobic walls

Amir Joulaei, Rakshithbt Gowda, Hossein Rahmani, and Seyed Mohammad Taghavi

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In this study, the plane Poiseuille flow of a viscoplastic fluid in a microchannel featured by two superhydrophobic (SH) walls is numerically studied. The two walls of SH (including the lower and upper walls) contain a series of micro-grooves that may exhibit a relative misalignment. Therefore, diverse configurations of the two SH walls are defined by the offset number, which represents the relative misalignment of the grooves at the lower wall when compared to those at the upper wall. The air is trapped within the grooves, assuming that the liquid/air interface remains flat while pinned at the groove edges. In order to model the boundary condition at the liquid/air interface, the Navier slip law is implemented, while a no-slip condition is considered at the liquid/solid contacts. Moreover, the Bingham model via the Papanastasiou regularization technique is used to model the fluid's viscoplastic behaviour. Throughout this study, we focus on thin channels, where the channel height is considerably smaller than the groove period. Our investigation encompasses the effect of flow inertia, along with various dimensionless parameters, including the Bingham number (B), slip number (b), groove periodicity length (l), offset number ( $\epsilon$ ), and slip area fraction ( $\phi$ ), on the morphology of the unyielded center plugs. Here, we explore the influence of the velocity perturbation field on the breakage and yielding of the unyielded center plugs, while also examining how the offset number, in conjunction with the Reynolds number, affects this process.

#### Thursday 12:00 LT 6 (Level 8) / Track 3

Rheology of magnetic hydrogels for soft magnetic actuators

<u>Francisco J. Vazquez-Perez</u><sup>1</sup>, Cristina Gila-Vilchez<sup>1</sup>, Alberto Leon-Cecilla<sup>1</sup>, Luis Álvarez de Cienfuegos<sup>2</sup>, Dmitry Borin<sup>3</sup>, Stefan Odenbach<sup>3</sup>, James E. Martin<sup>4</sup>, and Modesto T. Lopez-Lopez<sup>1</sup>

<sup>1</sup>Departamento de Física Aplicada, Universidad de Granada, Granada, Granada E-18071, Spain; <sup>2</sup>Departamento de Química Orgánica, Universidad de Granada, Granada, Granada E-18071, Spain; <sup>3</sup>Chair of Magnetofluiddynamics, Measuring and Automation Tech, Technische Universität Dresden, Dresde, Saxony 01069, Germany; <sup>4</sup>Sandia National Laboratories, Albuquerque, NM 87059, United States

Soft magnetic actuators are soft materials able to respond to an external magnetic field by locomotion, by changing their temperature or some of their dimensions, or even by bending or twisting. Magnetic field is an attractive stimulus due to the ease of its application, the fast response of magnetic actuators, and the safe penetration in biological environments [1, 2]. For mechanical characterization of the hydrogels used in the elaboration of the different actuators, we used a Discovery HR-1 rheometer (TA Instruments, USA) equipped with a parallel plate geometry of 40 mm of diameter for oscillatory shear tests and a two-clamp geometry for tensile tests. In oscillatory shear tests we measured the viscoelastic moduli as a function of the shear strain amplitude at fixed frequency, as well as a function of frequency at fixed shear strain amplitude. In tensile tests we measured the length of the sample as a function of the applied force, obtaining the Young's modulus. These properties were first measured for our actuators, which were prepared using mould-casting technique while an external magnetic field was applied during curing to obtain a prealignment of the magnetic particles. These first gels were prepared with a double calcium curing and low particle concentration. Secondly, actuators that respond to the external field by twisting were prepared with hydrogels cured only once with calcium, but with 3 times more magnetic particles. The Young's modulus obtained for this second material was used in a theory of torsional deformation for the different structures formed by the particles along the material: i) circular structures and ii) helical structures. Although similar Young's moduli were obtained for both materials, the viscoelastic moduli show significant differences.

S. R. Goudu, I. C. Yasa, X. Hu, H. Ceylan, W. Hu, M. Sitti, Adv. Funct. Mater., 30(50) (2020) 2004975.
 Y. Kim, H. Yuk, R. Zhao, S. A. Chester and X. Zhao, Nature, 558(7709) (2018) 274 - 279.

# Thursday 12:20 LT 6 (Level 8) / Track 3

# Rheology and microstructure of silica colloidal dispersions concentrated by drying: adsorbed, confined, free water and channels formation

<u>Manon Pépin<sup>1</sup></u>, Manon Pépin<sup>2</sup>, Anne Davaille<sup>2</sup>, and Christiane Alba-Simionesco<sup>1</sup> <sup>1</sup>Laboratoire Léon Brillouin, Gif-sur-Yvette 91190, France; <sup>2</sup>FAST, Orsay 91400, France

We investigate the links between the rheological properties and the nanoparticles 'organization in aqueous silica colloidal dispersions. We use Ludox solutions of two different diameters (16 and 28nm) and vary the water concentration, while keeping the ionic content constant. Each concentration is obtained by solvent evaporation. In a first protocol, the solution is continuously stirred during evaporation, so that the whole solution remains homogeneous. In a second protocol, the solution is left alone and natural convection develops. The rheology is characterized using shear rate and oscillatory tests. The organization of the nanoparticles is probed with Small-Angle Neutron Scattering (SANS) and Small-Angle X-ray Scattering (SAXS) measurements, and the solvent (water) state thanks to thermal analysis (ThermoGravimetric Analysis, TGA, and Differential Scanning Calorimetry, DSC).

These measurements allow us to identify three types of water, (i) water adsorbed at the surface of the particles, (ii) water confined in nanocavities and (iii) free water, and how these waters evolve with increasing particle volume fraction. Rheology seems mainly controlled by the amount of free water. As the fraction of the latter decreases, the material becomes more heterogeneous at the mesoscale, with the formation of particle aggregates and free water channels. The rheology becomes more non-Newtonian with the apparition of a yield stress, which value increases with decreasing free water. In convection-evaporation experiments, that results in the formation of a skin floating on a less concentrated solution, and

SV18

SV19

strong localization of deformation on this heterogeneous skin that can induce its break-up. When the free water has completely evaporated, the samples become completely solid and are subject to classical fracture dynamics.

# Symposium SG

# **Suspensions and Granular Materials**

Organizers: Erin Koos, Wilson Poon and Olivier Pouliquen

#### Thursday 10:40 LT 18 (Level 8) / Track 4 Crowd evacuations can follow Suspensions' rules

Dariel Hernández-Delfin, Ander García, and Marco Ellero

Basque Center for Applied Mathematics - BCAM, BILBAO, BIZKAIA 48009, Spain

We revisit pedestrian crowd evacuation through a bottleneck from a rheological perspective. Pedestrian crowd dynamics exhibit complex behavior with the variation of the competitiveness of pedestrians. In highly competitive scenarios, the desire to escape rapidly leads to higher evacuation times. This outcome has been previously corroborated through numerical and experimental investigations, named the faster-is-slower phenomenon (FIS). In suspension rheology, a similar effect is obtained when submitted to an external perturbation. Namely, the system's viscosity increases as the shear rate or shear stress upon the suspension increases, called shear thickening (ST). ST is explained on the basis of the transition from frictionless (lubricated) interactions to frictional contacts. A similar transition experiences pedestrian crowds when the competitiveness of pedestrians is increased, causing a system transition from purely repulsive social to frictional contact forces. We simulate pedestrian egress from a room across a constriction formed by the room door based on the Social Force Model (SFM)[1]. We find that the viscosity of the pedestrian flow depends on the competitiveness, showing discontinuous shear thickening close to the values where FIS is observed. Competitiveness affects the resulting stress and density simultaneously. To parse out both contributions, we perform a recently proposed universal scaling analysis [2], proving crowd evacuations are governed by two distinct critical jamming densities[3], likewise DST, one frictional and another frictionless. The study finds that combined social and shear forces are necessary to trigger the frictionless-to-frictional transition that leads to FIS. Contact forces alone cannot activate FIS, and social force interaction is essential for avoiding contact at low speeds.

[1] Helbing, D., et al. Nature. 407, 487-490 (2000) [2] Ramaswamy, M. et al. J. Rheol. 67, 1189-1197 (2023) [3] Hernández-Delfin, D., García, A., Ellero, M., Under Review in Comms. Phys.

# Thursday 11:00 LT 18 (Level 8) / Track 4

# Impact of shear-thickening capsules

Philippe Bourrianne<sup>1</sup>, Etienne Jambon-Puillet<sup>2</sup>, Howard A. Stone<sup>3</sup>, and Pierre-Thomas Brun<sup>4</sup>

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Dense suspensions exhibit shear-thickening, denoting a reversible increase in viscosity under external shear. While the steady-state rheology of dense suspensions has been extensively studied, the dynamics of the shear-thickening transition remain to be described. By filling thin elastic shells with shear-thickening suspensions, we study the deformation of capsules during their impact on a solid substrate. Liquid-filled elastic capsules are known as an analogue of liquid drops during impact. We will first describe the influence of the liquid viscosity on the spreading of capsules filled with Newtonian liquids. We will then show how the dynamics of the deformation of a capsule is dramatically altered in the case of a shear-thickening content. Finally, we will demonstrate how those observations provide access to rheological measurements and rare insights into the dynamics of solidification during the shear-thickening transition.

# Thursday 11:20 LT 18 (Level 8) / Track 4

#### Continuum modelling of dense suspensions with evolving relaxed states and flow-induced anisotropy <u>Giulio G. Giusteri<sup>1</sup></u>, Luca Santelli<sup>2</sup>, and Ryohei Seto<sup>3</sup>

<sup>1</sup>Department of Mathematics, Università Degli Studi Di Padova, Padova, Padova 35121, Italy; <sup>2</sup>Basque Center for Applied Mathematics, Bilbao, Basque Country 48009, Spain; <sup>3</sup>Wenzhou Key Laboratory of Biomaterials and Engineering, Wenzhou Institute, University of Chinese Academy of Sciences, Wenzhou, Zhejiang 325000, China

The phenomena of shear thickening and shear jamming in suspensions of solid particles dispersed in a viscous fluid have been widely investigated. Continuum-level models have been traditionally proposed based on a viscous fluid framework. Nevertheless, suspensions are materials that feature both fluid-like and solid-like behaviours, especially at high particle concentration. For this reason, we aim at including elastic contributions in continuum models as a proxy for the stress generated by particle contacts. An effective elastic response has been recently employed within a tensorial model for dense suspensions that can be jammed by shear [1]. In that work, rate-dependent effects were neglected, but they must be present in models that aim at capturing shear thickening.

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In this talk, we will present a major extension of that model, featuring a variety of non-Newtonian effects. In this line of research, the momentum balance equation is coupled with equations for tensorial quantities that describe the current state of deformation and an evolving set of relaxed states, for which the elastic stress vanishes. Notably, changes in the evolution of relaxed states can describe both continuous and discontinuous shear thickening. Moreover, we will discuss the role of the elastic response and that of flow-induced anisotropy, that can be originated by hydrodynamics interactions among the particles, in reproducing the behaviour observed in transient flows with shear rotation experiments [2].

G. G. Giusteri and R. Seto, Phys. Rev. Lett. 127, 138001 (2021)
 F. Blanc et al., Phys. Rev. Lett. 130, 118202 (2023)

# Thursday 11:40 LT 18 (Level 8) / Track 4

**Non-monotonic granular rheology under imposed stress and imposed volume** Christopher Ness<sup>1</sup> and Suzanne M. Fielding<sup>2</sup>

<sup>1</sup>School of Engineering, University of Edinburgh, edinburgh, United Kingdom; <sup>2</sup>Physics, Durham University, Durham, United Kingdom

We study the rheology of dry and wet granular matter using particle-based simulations operating under imposed volume and imposed stress conditions. Our model predicts non-monotonicity under both protocols, namely discontinuous shear thickening under imposed volume and shear thinning under imposed stress. This result suggests an apparent contradiction both in the constitutive behaviour of the material, and in the presumed nature of the shear banding that the system is predisposed toward. We reconcile this by demonstrating the mapping between S-shaped \$\sigma(\dot{\gamma})\$ flow curves in the imposed volume rheology and non-monotonicity in \$\mu(I)\$ and \$\mu(J)\$ in the respective imposed stress rheology, which reveals that there two separate non-monotonicities occurring at distinct stress scales.

# Thursday 12:00 LT 18 (Level 8) / Track 4

Dynamics of flexible fibers in complex viscous flows in pillar arrays

Zhibo Li<sup>1</sup>, Clément Bielinski<sup>2</sup>, Blaise Delmotte<sup>2</sup>, Anke Lindner<sup>1</sup>, and Olivia du Roure<sup>1</sup>

<sup>1</sup>Laboratoire de Physique et Mécanique des Milieux Hétérogènes, CNRS, ESPCI PSL, Sorbonne Université, Université Paris Cité, Paris, Île-de-France 75005, France; <sup>2</sup>LadHyX, CNRS, École polytechnique, Institut Polytechnique de Paris, Palaiseau, Îlede-France 91120, France

The dynamics of flexible fibers within complex environments, such as porous media, underlie many critical processes in both industrial and biomedical contexts. These processes encompass diverse applications, including chemical reactions in porous flow reactors through mixing, water or air purification via filtration, and medical diagnosis involving bioparticle separation. Our research integrates experimental and numerical approaches, explicitly focusing on unraveling the transport and morphological dynamics of flexible fibers-illustrated by actin filaments-within regularly arranged pillar arrays with various flow angles. Our investigation allows us to verify the array's efficacy in sorting fibers.

Actin filaments exhibit a wide variety of morphological dynamics within such complex environments. These dynamics involve not only twodimensional buckling, resulting in U-, S-, and W-shaped conformations, but also three-dimensional deformations leading to folded, coiled, or helical structures. The primary governing factor in filament dynamics is the dimensionless elastoviscous number, which compares viscous drag forces to elastic bending forces. By meticulously examining flow properties, we establish a mapping relation between filament deformation and the distribution of flow types and strengths within the array.

By delving into the intricate interplay between fiber dynamics and flow characteristics, our findings extend to understanding the chromatographylike sorting of fibers within this design and elucidating the deterministic lateral displacement (DLD) separation mechanism.

# Thursday 12:20 LT 18 (Level 8) / Track 4

# Simulations of sheared frictional particles – rheology, contact networks and fluctuations John R. Melrose

# NA, Consultant, Church Close Great B, Oxfordshire OX17 1RW, United Kingdom

Simulation results are reported for monolayers of polydisperse spheres with repulsive interactions, lubrication forces and frictional contacts. The equations of motion are without inertia. The aim is to connect the rheology to the changing network of contact constraints. Above area fraction F=0.8, the rheology is fit by a yield stress and shear thinning, akin to granular plasticity; the number of sliding contacts increases with shear rate, G and the contact networks (CN) have contact coordination Z>3. On quenching to G=0, the systems arrest in a range of states of percolating contact networks with non-zero stress tensor. Below F=0.795 the system has 3 regimes, with increasing G , Newtonian, sub-isostatic and a shear rate induced super-isostatic regime. In the sub-isostatic regime, at low G , the small systems studied (N = 195 to 6000) have a Newtonian regime, followed by shear thickening to a peak in viscosity followed by a shear thinning in the super-isostatic regime. The peaks are strongly system size dependent. On quenching to G=0, the contact networks collapse, but at high G, can show a slow evolution with intermittent jumps. Under flow, there are spatial temporal fluctuations in structure and collective flow patterns. In the sub-isostatic regime, they drive jumps between a low stress state and an unstable high stress state; these correlate strongly with the contact network coordination and measures of cluster rigidity. The talk will examine the relationship between the rheology and the particle level structure - its kinetics, flow patterns and their changing length scales. Fluctuations up and down in stress involve changes in the relative portions of clusters of local cages in compression and extension and more localised regions associated with bursts of high dissipation.

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# Symposium BL

# **Bio-rheology, Living and Active Matter**

Organizers: Laura Casanellas, Alexander Morozov and Christian Wagner

Thursday 10:40 LT 23 (Level 8) / Track 5

#### Physics and slowdown of the erythrocyte sedimentation rate

Alexis Darras, Thomas John, Lars Kaestner, and Christian Wagner

Experimental Physics, Saarland University, Saarbruecken, Saarland 66123, Germany

Red blood cells (or erythrocytes) sedimentation rate (ESR) is a physical parameter of blood which is often checked in medical diagnosis. It is indeed well known that in case of inflammation, the increase in fibrinogen and other proteins induces a higher ESR. A higher ESR is clinically established as a disease marker. Recently, we demonstrated that Red Blood Cells (RBCs), when left at rest and suspended at physiological volume fractions, form percolating aggregates as wide as the container. It follows that they sediment following a so-called "gel collapse", governed by the geometry of the percolating aggregate acting as a porous material. In this talk, by comparing physical models to experimental sedimentation curves, we show how this knowledge can help to quantify physically meaningful parameters that characterize the details of the collapse dynamics. Amongst others, we provide a dependency of the maximal sedimentation velocity as a function of the initial RBC volume fraction (i.e. the hematocrit), which was a long-sought correction for ESR measurements from anemic patients. We also review how those parameters make it possible to experimentally distinguish between healthy samples and some conditions where the ESR is slowed down. In particular, this opens new perspective to use the ESR as an objective marker to detect diseases where the RBCs are deformed and/or rigidified.

#### Thursday 11:00 LT 23 (Level 8) / Track 5

A molecular-mechanical link in shear-induced self-assembly of a functionalized biopolymeric fluid

Galina E. Pavlovskaya and Thomas Meersmann

Sir Peter Mansfield Imaging Centre, Medicine, University of Nottingham, Nottingham, Nottingham NG7 2RD, United Kingdom

Na-23 multiple quantum filtered (MQF) rheo-NMR methods were applied to probe the molecular foundation for flow induced self-assembly in 0.5% k-carrageenan fluid. This method is sensitive enough to utilize an endogenous sodium ion concentration of approximately 0.02%. Rheo-NMR experiments were conducted at different temperatures and shear rates to explore varying molecular dynamics of the biopolymer in the fluid under shear. The temperature in the rheo-NMR experiments was changed from 288 K to 313 K to capture transition of k-carrageenan molecules from helices to coils. At each temperature, the fluid was also tested for flow and oscillatory shear behaviour using bulk rheometry methods. It was found that the Na-23 MOF signals were observed for the 0.5% k-carrageenan solution only under shear and when the fluid demonstrated yielding and/or shear-thinning behaviour. At temperatures of 303 K and above, no Na-23 MQF signals were observed independent of the presence or absence of shear as the molecular phase transition to random coils occurs and the fluid becomes Newtonian. References: Soft Matter, 2023, 19, 3228

#### Thursday 11:20 LT 23 (Level 8) / Track 5

# Ordering of red blood cells in viscoelastic confined flows

Yazdan Rashidi<sup>1</sup>, Ian Graham<sup>2</sup>, Paulo E. Arratia<sup>2</sup>, Francesco Del Giudice<sup>3</sup>, Christian Wagner<sup>1</sup>, and Steffen M. Recktenwald<sup>1</sup> <sup>1</sup>Department of Experimental Physics, Saarland University, Saarbruecken 66123, Germany; <sup>2</sup>Department of Mechanical Engineering and Applied Mechanics, University of Pennsylvania, Philadelphia, PA 19104, United States; <sup>3</sup>Department of Chemical Engineering, Swansea University, Swansea SA1 8EN, United Kingdom

Blood primarily consists of red blood cells (RBCs), which significantly affect its flow behavior in the circulatory system. RBCs high deformability allows them to squeeze through micro-vessels that are much smaller than their equilibrium size. In microfluidic flows with channel dimensions similar to their size, RBCs exhibit characteristic shapes such as croissants and slippers. Understanding RBC single-cell flow has advanced the development of lab-on-a-chip technologies for the assessment of RBC deformability as a biomarker for specific pathologies. However, the tendency of cell clustering in commonly employed Newtonian buffer solutions hinders the analysis throughput of individual RBCs. Consequently, such approaches are often limited to very dilute samples to avoid cell clustering. In this work, we investigate the effects of fluid rheology on the ordering (or lack thereof) of RBCs in strongly confined channels in experiments using single-cell shape classification and in simulations. Ordering of rigid spheres in viscoelastic polymer solutions has recently been explored. Nevertheless, our understanding of such ordering phenomena for anisotropic and deformable objects is limited. Here, we probe RBC ordering along the channel centerline in microchannels with a cross-section similar to the RBC size. We use different polymer solutions displaying shear-thinning and viscoelasticity or near-constant viscosity and viscoelastic features. We first describe the effect of the non-Newtonian fluid as a surrounding medium on the well-known croissant shape and the RBC clustering in capillary flow. Second, we demonstrate, for the first time, that equally spaced RBCs emerge in different shear-thinning, viscoelastic polymer solutions, independent of the molecular architecture of the polymer molecule. Complementary numerical simulations reveal that fluid viscoelasticity plays an important role in obtaining repulsion between two RBCs, which leads to equally spaced RBCs, while shearthinning seems to modulate this effect.

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**BL16** 

**BL15** 

**BL18** 

# Thursday 11:40 LT 23 (Level 8) / Track 5

Shear-Induced Accelerated Coagulation and Fibrinolysis in Oscillating Tube Flow

Sehyun Shin<sup>1</sup>, Jikang Wang<sup>2</sup>, SungSoo Na<sup>3</sup>, Chang-Soo Han<sup>4</sup>, and chenhui Zhang<sup>5</sup>

<sup>1</sup>Mechanical Engineering, Korea University, Seoul, Republic of Korea; <sup>2</sup>Mechanical Engineering, Korea University, Seoul, Republic of Korea; <sup>3</sup>Mechanical Engineering, Korea University, Seoul, Republic of Korea; <sup>4</sup>Mechanical Engineering, Korea University, Seoul, Republic of Korea; <sup>5</sup>Mechanical Engineering, Korea University, Seoul, Republic of Korea

Background: Rapid and precise assessment of viscoelastic hemostatic assays is crucial in emergency settings for making timely medical decisions. However, conventional thromboelastography (TEG) analyzers, which require long test times of up to 1 hour, are often unsuitable for emergency patients. Recently, we introduced Global Thrombosis Express (GTX), an extremely rapid thromboelastography device that completes the entire test in just 10 minutes at relatively elevated shear rates. Thus, it is necessary to determine whether the parameters of GTX are interchangeable and comparable to those of conventional TEG devices such as the TEG5000. Materials and Methods: To address this issue, we conducted a comparative analysis of basic kaolin assays using GTX and TEG5000. Due to the fast testing characteristic of  $\mu$ -TEG, we defined new parameters for GTX including R-time, K-time, angle a, maximum amplitude (MA), time to MA (tMA), Ly30, and CL30. Assays were repeated in duplicate. Reliability (TEG 5000 vs. GTX) and repeatability (interdevice comparison) was quantified. Linear regression was used to define the relationship between GTX and TEG 5000 but showed high correlation (R2 = 0.86, 0.69, and 0.87, respectively). MA exhibited the highest correlation (R2 = 0.95) between GTX and TEG5000. According to the normal ranges of MA, the diagnostic sensitivity and specificity of GTX were 93% and 100%, respectively. Conclusion: Despite the significantly reduced test time, all parameters assessed with GTX would be beneficial for emergency and operating rooms in analyzing hemostasis and fibrinolysis. Given the potential advantages of using the GTX device at the site of care, confirmation of agreement between the devices represents an important advance in diagnostic testing.

# Thursday 12:00 LT 23 (Level 8) / Track 5

BL19

**BL20** 

# Biomicrofluidic analysis of hematological diseases by means of mathematical biomechanical models and statistical analysis

Aurora Hernández-Machado

Física de la Materia Condensada, Universitat de Barcelona, Barcelona, Barcelona 08028, Spain

We have developed microfluidic devices for precise characterization of hematological diseases. By means of one drop of blood and combining front microfludics with mathematical models based on biomechanics, we analyze the properties of red blood cells and microrheological properties such as the viscosity of blood. We detect the advancement of microfronts of fluid inside microchannels. By means of statistical analysis we improve the diagnosis of the hematological diseases. We predict if a sample of blood corresponds to healthy blood or to blood with an hematological disease. We have obtained different performance for the different methods, some of them with very good results and an accuracy of 94%.

Thursday 12:20 LT 23 (Level 8) / Track 5

# Development of active all-enzyme hydrogels from glucose oxidase

Harrison Laurent<sup>1</sup>, Matt D. Hughes<sup>1</sup>, David J. Brockwell<sup>2</sup>, and Lorna Dougan<sup>1</sup>

<sup>1</sup>School of Physics and Astronomy, University of Leeds, Leeds, West Yorkshire LS2 9JT, United Kingdom; <sup>2</sup>Astbury Centre for Structural Molecular Biology, University of Leeds, Leeds, West Yorkshire LS2 9JT, United Kingdom

Folded protein hydrogels offer an exciting class of biomaterials with tunability of their viscoelastic properties while maintaining protein functionality, providing powerful opportunities in biomedical and industrial settings. Our group has previously employed combined rheology, neutron/x-ray scattering, and modelling techniques to obtain cross-lengthscale understanding of the mechanical and structural properties of various photochemically cross-linked monomeric protein hydrogels [1,2]. In this work we focus on the dimeric enzyme glucose oxidase (GO) from *Aspergillus Niger*, which catalyses the reaction of glucose into gluconic acid and hydrogen peroxide. This enzyme has been widely researched several applications, including diabetic monitoring and treatment, biobatteries, cancer cell culture, and nanoparticle synthesis [5]. As GO can dissociate into folded monomers, reassociate into dimers, and unfold leading to biopolymer entanglement, the time-resolved rheological profile of network formation is complex, and has a non-trivial dependence on volume fraction. This is in stark contrast to previous work on monomeric protein systems, in which *G'* rapidly increases with hydrogel formation, followed by a slow relaxation due to network rearrangement and protein unfolding. By employing time resolved, linear and non-linear rheology, we quantify the characteristic timescales of these processes for GluOx hydrogels. Complimentary spectroscopic techniques are employed to monitor structural and protein relaxations directly. Finally, we measure the functional response of the hydrogel using an enzymatic activity assay and applying Michaelis-Menten kinetics. This multi-modal cross-lengthscale approach allows us a deep insight into the design space of both dimeric and all-enzyme hydrogels, providing new avenues in biomedical applications. 1. Hughes, M. D. G. et al. ACS Nano 15, 11296-11308 (2021) 2. Hughes, M. D. G. et al. Soft Matter 16, 6389-6399 (2020) 3. Salehipour, M. et al. Polym. Bull. 80, 5861-5896 (2023)

# Symposium GR

# **Geo-rheology**

Organizers: Janine Kavanagh, Edward Llewellin and Sandra Piazolo

# Thursday10:40LT 11 (Level 10) / Track 6KeynoteGR1Rheology of crystalline materials during diffusion creep: influence of grain coarsening and grain boundarysliding

#### John Wheeler<sup>1</sup>, Sandra Piazolo<sup>2</sup>, Lyn Evans<sup>3</sup>, and Robyn Gardner<sup>4</sup>

<sup>1</sup>Earth, Ocean and Ecological Sciences, University of Liverpool, Liverpool, Merseyside L69 3GP, United Kingdom; <sup>2</sup>University of Leeds, Leeds, United Kingdom; <sup>3</sup>Monash University, Melbourne, Australia; <sup>4</sup>Macquarie University, Sydney, Australia

Diffusion creep is a major contributor to superplasticity, an industrially important deformation style in metals, and plays a key role in rock deformation deep in the Earth. The wet low temperature version, pressure solution, is a major deformation mechanism in the upper 10s of km of the Earth. The mechanism involves dissolving grains at high stress interfaces, diffusing the material and precipitating it at low stress interfaces. Given the wide range of circumstances under which it operates, it is important to understand the rheology of diffusion creep. Based on well-established simplified analysis, this is Newtonian and smaller grain sizes give lower viscosities. I discuss here a more detailed analysis where rheology of interlocking grain aggregates is calculated from their geometries, and modelling of subsequent microstructure evolution shows how the rheology evolves.

These are some key and perhaps surprising predictions to be discussed.

- 1. The diffusion must be accompanied by sliding at grain boundaries. Models show that strain localises along slip surfaces formed by aligned grain boundaries on all scales, thus strongly modifying overall strength.
- 2. Diffusion creep is predicted to produce elongate grains and then the overall aggregate has intense mechanical anisotropy. Thus strength during diffusion creep, and localisation on weak zones, is influenced not just by grain size but by other aspects of microstructure.
- 3. Grain coarsening increases grain size and thus strength. Our most recent work shows how it interacts with ongoing deformation; in particular grain growth can lead to particular grain shapes which are directly related to strain rate, and influence strength.

Consequently, understanding the rheology of diffusion creep must encompass the effects of diffusion itself, grain boundary sliding and grain coarsening. Interactions between these processes merit further study.

http://pcwww.liv.ac.uk/johnwh/

# Thursday 11:20 LT 11 (Level 10) / Track 6

# Unravelling the internal dynamics of low-temperature, wet pyroclastic density currents

# <u>Nemi Walding</u><sup>1</sup>, Rebecca Williams<sup>2</sup>, Pete Rowley<sup>3</sup>, Natasha Dowey<sup>4</sup>, Mark Thomas<sup>5</sup>, and Sara Osman<sup>5</sup>

<sup>1</sup>Energy and Environment Institute, University of Hull, Hull, United Kingdom; <sup>2</sup>School of Environmental Sciences, University of Hull, Hull, United Kingdom; <sup>3</sup>University of Bristol, Bristol, United Kingdom; <sup>4</sup>Sheffield Hallam University, Sheffield, United Kingdom; <sup>5</sup>University of Leeds, Leeds, United Kingdom

Pyroclastic Density Currents (PDCs) are hazardous, rapidly moving and often high-temperature particle-laden dense currents that exhibit behaviours on a continuum from turbulent (i.e., fluid-dominated) to granular (i.e., particle-dominated) regimes. PDCs are fluidised by the upward movement of gas, which increases pore pressure and promotes mobility of the flow. The internal dynamics of these deadly volcanic phenomena are incredibly difficult to observe and measure at the field scale. Therefore, our understanding of PDCs relies on a combination of stratigraphic field work analysis, analogue experiments, and numerical modelling. PDCs can become cold, wet, and unpredictable with the introduction of moisture. In phreatomagmatic eruptions, rising magma interacts explosively with external water. In phreatic eruptions, water beneath the ground or on the surface is heated by magma generating steam driven explosions. Both types of eruptions have the potential to produce violent PDCs. The devastating phreatic eruption on Mount Ontake, Japan (September 2014) caused 60 fatalities from low-temperature PDCs, inundating tourists visiting the volcano, and the 1888 phreatic eruption of Bandai (Japan) killed 461 people from PDCs at temperatures less than 100°C, but at extreme levels of humidity. Previous experimental works by Walding et al. (2023) show how the presence of water in a pyroclastic material can significantly reduce flowability and fluidisation (i.e., the mobility and transport behaviours of a PDC), highlighting how the introduction of moisture can fundamentally impact flow dynamics and run-out distance of PDCs. However, there has been little study into the role that moisture plays in PDC rheology. Understanding wet PDCs will be pivotal in enhancing our grasp of their flow behaviours and thus the hazard that they pose. This research aspires to provide innovative data points that can directly impact hazard management practices and strengthen the ability to predict and manage PDCs with greater fidelity.

GR2

#### Thursday 11:40 LT 11 (Level 10) / Track 6

**Particles sinking in non-Newtonian seawater: the role of exopolymers in modifying sedimentation processes** <u>Magdalena M. Mrokowska<sup>1</sup></u>, Anna Krzton-Maziopa<sup>2</sup>, and Tomasz Chablo<sup>2</sup>

<sup>1</sup>Hydrology and Hydrodynamics Department, Institute of Geophysics, Polish Academy of Sciences, Warsaw 01-452, Poland; <sup>2</sup>Faculty of Chemistry, Warsaw University of Technology, Warsaw 00-664, Poland

Natural waters including seas are commonly considered as Newtonian media. However, microalgae and bacteria can modify the rheology of water by excessive secretion of exopolymers (EPSs), which in their hydrated forms create gel-like structures. Despite field observations, the non-Newtonian properties of EPSs in saline environments and the implications to geophysical processes such as sedimentation of marine particles (minerals, aggregates, microplastics), are poorly understood. Meanwhile, marine polymers have been widely studied in industrial applications. To tackle this issue, we investigated to what extent the rheology of seawater affects the sedimentation of model particles. Settling experiments combined with rheological tests, utilising two commercial polysaccharides: xanthan gum (XG) and kappa-carrageenan (CR) dispersed in seawater and in solutions of single salts (e.g. NaCl, KCl), were conducted, and flow and viscoelastic characteristics of solutions were associated with the movement of particles throughout liquid medium. The settling of spheres, disks and rods in a transparent tank was captured with a camera. It has been revealed that the dynamics of particles varied in sinking velocities, orientations, and secondary motions depending on the type of polymer used as a rheology modifier. The observed changes can be well explained by different rheological properties of tested solutions originating from gel-like network created by mutual interactions between macromolecules. Even more pronounced rheological and hydrodynamic effects were observed in solutions containing dissolved electrolytes (e.g. Ca, K chlorides) demonstrating their crucial roles in saltwater mixture on structuring certain groups of polysaccharides. Our study provides valuable insights into the interactions between EPSs and electrolytes comprising seawater and the effects of modified rheology on the hydrodynamics of particles. These findings provide a link between understanding the role of EPSs in natural waters and industrial applications.

# Thursday 12:00 LT 11 (Level 10) / Track 6

GR4

# A high-pressure (250 MPa) high-temperature (1120°C) rheometer: measuring the viscosity of volatile rich magmas

<u>Francesca L. Haywood</u><sup>1</sup>, Richard A. Brooker<sup>1</sup>, Heidy Mader<sup>1</sup>, Laurence Breeze<sup>1</sup>, Clapham Charles<sup>1</sup>, Amanda Lindoo<sup>2</sup>, and Gerald Mwale<sup>1</sup>

#### <sup>1</sup>Earth Science, University of Bristol, Bristol, United Kingdom; <sup>2</sup>Durham University, Durham, United Kingdom

We introduce a new high-pressure high-temperature (HPHT) rheometer capable of direct viscosity measurements across the range 100-30,000 Pa s at conditions up to 250 MPa and 1200oC. This rheometer will allow testing of current models of magma rheology with direct viscosity measurements, particularly at hydrous conditions. Magma rheology is a crucial focus of present-day volcanological research due to its primary control over a wide range of volcanic processes (from the extraction of magma in the deep earth, its transport to the surface, and finally, its sub-aerial eruption). Throughout this journey from source to deposition, the magma will experience significant changes in both temperature and pressure, driving extreme changes in melt viscosity measurements of high-temperature magmas are generally limited to anhydrous, ambient pressure conditions. This limitation is due to the difficulty posed in adding a pressurised chamber to a rheometer. Indirect HPHT viscosity measurements have been achieved using the falling-sphere method, however these are limited to low-viscosity materials, non-dynamic conditions and are prone to larger uncertainties (>15%). Our new rheometer design combines a cold-seal high-pressure apparatus and a concentric cylinder rheometer. It is able to achieve HPHT conditions due to a magnetically coupled measuring system which links an internal spindle within a sealed pressure chamber to an external rheometer head (supplied by Anton Paar). The HPHT rheometer produces viscosity measurements with an uncertainty of  $\pm 5\%$ , the technical features and challenges that contribute to this uncertainty will be presented.

# Thursday 12:20 LT 11 (Level 10) / Track 6

GR5

# The rheology of bubble suspensions up to the wet foam limit, with application to magma and lava <u>Ceri Allgood</u>, Mélissa J. Drignon, and Edward W. Llewellin

Department of Earth Sciences, Durham University, Durham, United Kingdom

Magma and lava comprise a continuous Newtonian liquid phase suspending a variable fraction of gas bubbles. Bubble volume fractions are typically within the range  $0 \le \Phi \le 0.8$ , so modelling the flow of magma and lava requires an accurate model for the rheology of bubble suspensions up to the wet foam limit. The rheology of a bubble suspension depends strongly on its bubble volume fraction and shear conditions, quantified by the capillary number Ca. This dimensionless number describes the balance between viscous stress deforming the bubbles, and surface tension stress restoring them towards spheres. When Ca  $\ll 1$ , surface tension stress dominates, bubbles are close to spherical, and the apparent viscosity of the suspension decreases with increasing  $\Phi$ . Here, we present the results of analogue rheometry experiments that explore the transition between these regimes, for bubble volume fractions in the range  $0 \le \Phi \le 0.74$ . We use suspensions of CO<sub>2</sub> bubbles in golden syrup in a concentric cylinder geometry, at low Reynolds number and low Stokes number. We access both the high and low capillary number regimes by running paired experiments at 10 & degC and 25 & degC, exploiting the temperature dependence of syrup viscosity to control the viscous stress. Flow curves are collected for the low capillary number regime, whereas oscillatory rheometry is used for the high capillary number regime to minimize bubble breakup and coalescence . Our data support the Cox-Merz rule, with the viscosities from flow curves overlapping the complex viscosities from frequency sweeps. We also find that the dependence of viscosity on Ca is well captured by a Cross model. Our data greatly expand the range of  $\Phi$  covered by previous experiments, and provide a new model for estimating the viscosity of magmas and lavas based on their bubble content.

# Symposium EM

# Experimental Methods and New Rheometric Techniques

Organizers: Christian Clasen, Dan Curtis and Manlio Tassieri

Thursday 10:40 LT 19 (Level 10) / Track 7

EM1

EM2

EM3

# High-frequency optimally windowed chirp rheometry for rapidly evolving viscoelastic materials: application to a crosslinking thermoset

<u>Thanasis E. Athanasiou</u><sup>1</sup>, Michela Geri<sup>2</sup>, Patrice Roose<sup>3</sup>, Gareth H. McKinley<sup>2</sup>, and George Petekidis<sup>1</sup> <sup>1</sup>IESL-FORTH, Heraklion 70013, Greece; <sup>2</sup>Hatsopoulos Microfluids Laboratory, MIT, Cambridge, MA 02139, United States; <sup>3</sup>allnex, Drogenbos, Belgium

The knowledge of the evolution of mechanical properties of the curing matrix is of great importance in composite parts or structure fabrication. Conventional rheometry, based on small amplitude oscillatory shear is limited by long interrogation times. In rapidly evolving materials, time sweeps can provide a meaningful measurement albeit at a single frequency. To overcome this constraint we utilize a combined frequency and amplitude-modulated chirped strain waveform in conjunction with a home-made sliding plate piezo-operated (PZR) and a dual-head commercial rotational rheometer (Anton Paar MCR 702) to probe the linear viscoelasticity of these time-evolving materials. The direct controllability of the PZR resulting from the absence of any kind of firmware and the microsecond actuator-sensor response renders this device ideal for exploring the advantages of this technique. The high frequency capability allows us to extend the upper limits of the accessible linear viscoelastic spectrum and most importantly, to shorten the length of the interrogating strain signal (PZR-Chirp) to sub-second scales, while retaining a high time-bandwidth product. This short duration ensures that the mutation number is kept sufficiently low, even in fast curing resins. The method is validated via calibration tests in both instruments and the corresponding limitations are discussed. As a proof of concept the technique is applied to a curing vinylester resin. The linear viscoelastic (LVE) spectrum is assessed every 20 seconds to monitor the rapid evolution of the time- and frequency-dependence of the complex modulus. Comparison of the chirp implementation in a commercial rotational rheometer with the PZR provides further information on the applicability of this technique and its limitations. Finally, FTIR spectroscopy is utilized to gain insights on the evolution of the chemical network while the gap-dependence of the evolving material properties in these heterogeneous systems is also investigated.

# Thursday 11:00 LT 19 (Level 10) / Track 7 High-Frequency Linear Rheology via Ultrasound-Driven Microbubble Dynamics: Methodology and Experimental Validation

Yu-Fan Lee<sup>1</sup>, Claude Oelschlaeger<sup>2</sup>, and Valeria Garbin<sup>1</sup>

<sup>1</sup>TU Delft, Delft 2629 HZ, The Netherlands; <sup>2</sup>Institute for Mechanical Process Engineering and Mechanics, Karlsruhe Institute of Technology, Karlsruhe 76131, Germany

High-frequency rheology provides access to fast dynamics in soft materials, encompassing a broad frequency range from 1 to 10^8 Hz. Among the available high-frequency rheometry methods, the ultrasound-driven bubble dynamics approach holds promise for high-frequency measurements, probing viscoelasticity within a wide range, from 10^3 Pa to 10^7 Pa, across a potential frequency spectrum from kHz to MHz. This approach holds relevance to applications in diagnostic and therapeutic ultrasound. Theoretical studies have proposed various models for bubble dynamics within viscoelastic materials. Building upon it, experimental studies have demonstrated the practical feasibility of employing bubble dynamics as a rheological measurement technique. In this study, we further improved the precision of the ultrasound-driven bubble dynamics approach and validated the extracted rheological properties of the viscoelastic media at high frequency with measurements from diffusing-wave spectroscopy (DWS). Soft agarose gel was chosen to fit in within the overlapping measurement window of bubble dynamics approach and DWS. We meticulously demonstrate and refine the calibration protocol, allowing us to obtain resonance curves that contain rheological information. The extracted rheological properties are compared with those obtained by DWS, revealing reasonable agreements between the two independent measurement techniques. This finding serves as a significant validation step for the bubble dynamics approach. The accuracy of the proposed method and avenues for future improvements are also discussed.

# Thursday 11:20 LT 19 (Level 10) / Track 7

# Beyond bio- and micro-rheology: an innovative method to investigate protein condensates viscosity and surface tension through correlative atomic force and fluorescence microscopies

<u>Andreas Santamaria</u><sup>1</sup>, Luca Costa<sup>1</sup>, Christine M. Doucet<sup>1</sup>, Stephanie Hutin<sup>2</sup>, Chloe Zubieta<sup>2</sup>, and Pierre-Emmanuel Milhiet<sup>1</sup> <sup>1</sup>Centre de Biochimie Structurale - CNRS, Montpellier 34090, France; <sup>2</sup>Laboratoire de Physiologie Cellulaire and Végétale-CEA-CNRS, Grenoble, France

Biomolecular condensates, formed by liquid-liquid phase separation (LLPS) play a crucial role in subcellular compartmentalization, contributing to the formation of membrane-less organelles and various cellular pathways (Gao et al., 2022). Their transition to a solid state, which impacts rheological and mechanical properties, is often linked to the onset of pathologies (Darling and Shorter, 2021). Therefore, viscosity and surface tension between condensed and dilute phases are key parameters to investigate condensate dynamics, function and aging. Common techniques employed to investigate such properties include fluorescence recovery after photobleaching (FRAP) (Alshareedah et al., 2021), optical tweezers (OT) (Jawerth et al., 2018), coalescence (Fisher and Elbaum-Garfinkle, 2020) and microrheology (Alshareedah et al., 2021). In particular, passive

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microrheology relies on tracking the movement of beads embedded within condensates, determining their diffusion coefficient, hence the surrounding viscosity (Fisher and Elbaum-Garfinkle, 2020; Alshareedah et al., 2021). However, results are influenced by bead size, functionalization, and local inhomogeneities (Feric and Brangwynne, 2013; Alshareedah, et al., 2021). Regarding OT, the high laser power employed can hinder the characterization of thermosensitive condensates, also inducing additional phase transitions. To address these limitations, we introduce an innovative method combining atomic force and fluorescence microscopies, which allows, in a one-shot experiment, to determine condensate surface tension and viscosity. Initially, the condensate is confined between a spherical AFM probe and the substrate to obtain surface tension. Subsequently, the probe induces dry spots (circular dewetting) within large condensates. The fluorescent mass flow during rewetting is quantified in terms of contact line speed, allowing the assessment of the condensate viscosity (Bankoff et al., 2003). This method is named FRAP-ID: Fluorescence Recovery After Probe-Induced Dewetting.

#### Thursday 11:40 LT 19 (Level 10) / Track 7

#### Puff rheometer for fast and contactless measurements of viscosity and surface tension

Coen R. van der Gracht<sup>1</sup>, Nick O. Jaensson<sup>1</sup>, <u>Ruth Cardinaels<sup>1</sup></u>, and Ruth Cardinaels<sup>2</sup>

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Characterization of the rheology and interfacial properties of materials typically requires different measurement setups and involves different manipulations of the sample. To enable in-situ and in-line characterization of the viscoelastic and interfacial properties of soft materials, we are developing a fast and non-manipulative apparatus. An air puff with a known pressure profile is applied to the surface of the material and the resulting surface deformation depends on the rheological, interfacial and physical properties of the material. This principle was initially introduced for characterizing the firmness of fruits (1) and previously, a qualitative correlation between surface deformation and viscosity of viscous food products was shown (2). Our puff rheometer combines measurements of the air-deformed surface and a numerical model within an optimization procedure. The numerical model, based on the finite element method (FEM), is used to model the deformation of the fluid caused by the air puff. This numerical problem is assumed to be axi-symmetrical, the top surface is free to deform and a range of constitutive models are considered to represent different types of (complex) fluids. The experimental setup uses pressurized air, a pressure controller, and a solenoid valve to control the air puff, and measures the deformation of the fluid surface with a laser sensor. With an optimization procedure, the numerical model can be matched to the live measurements, giving access to the surface tension and rheological properties. We analyze the sensitivity of the measurement results to the various input parameters and material characteristics to quantify the potential of this method. Moreover, a simple analytical model capturing the main physical mechanisms is shown to accurately describe the experimental and numerical data for Newtonian fluids, which facilitates data extraction. (1) Hung et al. (1999) Postharvest Biology and Technology, 16(1), 15-25. (2) Morren et al. (2015) Journal of Texture Studies, 46(2), 94-104.

#### Thursday 12:00 LT 19 (Level 10) / Track 7

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# **Measurement of viscoelastic properties of a liquid using a rotating body subjected to oscillatory shear** <u>Wook Ryol Hwang<sup>1</sup> and Hye Jin Ahn<sup>2</sup></u>

<sup>1</sup>School of Mechanical Engineering, Gyeongsang National University, Jinju-si, Gyeongsangnam-do 52528, Republic of Korea; <sup>2</sup>School of Mechanical Engineering, Gyeongsang National University, Jinju-si, Republic of Korea

We propose a systematic approach for measuring linear and non-linear viscoelastic properties of a liquid by the oscillatory motion of an immersed rotating body in a vessel. The shape of a rotating object is general and we tested four different types of impellers in the present work: a disk, an anchor, and two different flat bladed turbines. The effective shear stress was properly scaled with a torque through the expression of complex viscosity and the strain magnitude was scaled by the deflection angle with an effective shear rate coefficient (the Metzner-Otto constant). Three different concentrations of aqueous polyethylene oxide (PEO) solutions were tested and linear viscoelastic responses of storage and loss moduli were measured as a function of the oscillation frequency. In spite of the presence of non-rheometric and highly non-uniform flow field, comparison with the data from the conventional cone-and-plate and parallel-plate fixtures of a rheometer shows remarkably accurate measurement with at most 7% deviation within the frequency range from 0.01 [rad/s] to 100 [rad/s] for all the impeller geometries, except for the pitched bladed turbine. In addition, we show that this method can be applied to large amplitude oscillatory shear experiments for non-linear viscoelastic properties. The proposed method provides a versatile measurement for viscoelasticity, eliminating complicated issues in the conventional rheometry such as the wall slip, free surface evaporation/solidification and the edge fracture without a significant loss of accuracy. Furthermore, the method may facilitate the in-situ measurement of viscoelastic properties of a fluid within an industrial reactor/agitator as a tool for in-situ/on-line monitoring of microstructures.

Referecne: W. R. Hwang, H. J. Ahn. (2023). System and method for measuring rheological property of fluid using a rotating body of an arbitrary shape (Korean Patent No. 10-2023-0055682).

# Thursday 12:20 LT 19 (Level 10) / Track 7

#### Strategy to advance creep measurements of polyolefin melts

Georg M. Scheutz<sup>1</sup>, Sylvie Vervoort<sup>2</sup>, and Elin Goedhart-Smet<sup>2</sup>

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Creep and creep/recovery measurements are particularly meaningful for the characterization of polymer melts. They reveal the behaviour of the polymer at low deformation rates or long times and allow to determine zero-shear viscosity and longest relaxation time, which are powerful probes

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of polymer structure. Creep measurements to steady state often require long instrument times, especially if the molecular weight of the polymer is high. Besides long lead times to data and low productivity, this entails a risk of polymer degradation, inducing erroneous results. To address this problem, we developed an advanced creep testing strategy to reduce the number of inconclusive tests and improve measurement efficiency. The improved workflow involves real-time first-derivative analysis of log J(t) and a practical a priori creep duration estimation based on a standard SAOS measurement.

# Symposium NF

# Non-Newtonian Fluid Mechanics and Flow Instabilities

Organizers: Marco Ellero, Stylianos Varchanis and Helen Wilson

Thursday 10:40 LT 24 (Level 10) / Track 8

# **Revisiting Expansion Flows of Viscoelastic Fluids**

Joshua L. Cummings<sup>1</sup>, Mónica S. Oliveira<sup>1</sup>, and Konstantinos Zografos<sup>2</sup>

<sup>1</sup>James Weir Fluids Laboratory, University of Strathclyde, Glasgow, United Kingdom; <sup>2</sup>Altair, Edinburgh, United Kingdom

This research reexamines planar sudden expansion flows of viscoelastic fluids, expanding previous numerical work [1] to higher Weissenberg numbers. Unlike the more commonly investigated sudden contraction flows, which have benefited from the implementation of numerical advancements, the sudden expansion has been comparatively overlooked due to its complexity in numerical simulation. The study utilises the log-conformation approach to build upon existing findings related to creeping flow in viscoelastic fluids described by the upper-convected Maxwell model and investigate elasticity beyond previous numerical limitations in the literature. It is shown that the corner vortices exhibit a strong non-monotonic change with Weissenberg number, with the existence of large vortices in the inlet channel demonstrated and investigated. The influence of expansion ratio is also studied and its influence on vortex lengths is considered. The study is extended to include Oldroyd-B fluids and the implications of a Newtonian solvent are considered. Inertial flows are also investigated, contrasting the numerical studies of inertial flows through sudden expansions present in the literature which are limited to simulations with FENE models [2, 3]. The study is then further expanded to include sudden expansions with multiple inlets, a configuration with direct industrial application, where the influence of changes to geometric parameters such as inlet spacing is studied.

[1] Poole, R. J., et al. "Plane sudden expansion flows of viscoelastic liquids." Journal of Non-Newtonian Fluid Mechanics 146.1-3 (2007): 79-91.

[2] Oliveira, P.J. "Asymmetric flows of viscoelastic fluids in symmetric planar expansion geometries." Journal of Non-Newtonian fluid mechanics 114.1 (2003): 33-63.

[3] Rocha, G.N., et al. "Bifurcation phenomena in viscoelastic flows through a symmetric 1: 4 expansion." Journal of Non-Newtonian fluid mechanics 141.1 (2007): 1-17.

# Thursday 11:00 LT 24 (Level 10) / Track 8

# Theoretical analysis of viscous and viscoelastic flow through a cross-slot <u>Xintong Ji</u> and Helen J. Wilson

Department of Mathematics, University College London, London WC1E 6BT, United Kingdom

A cross-slot (or cross-channel) is a flow geometry with four 'arms'. The inflow comes from two opposite (horizontal as default) 'arms' and the outflow goes away from the other two opposite (vertical as default) 'arms'. A slow flow through this geometry produces a pattern with reflectional symmetry and produces a good approximation to a pure planar extension near the stagnation point at the centre.

For viscoelastic flow through the cross-slot, there is substantial evidence of a symmetry-breaking event as the flow rate is increased. A body of experimental evidence follows from the original work of Arratia et al. (2006) and numerical evidence (e.g. Poole et al. (2007)) to show an initial bifurcation to a steady asymmetric flow that results from purely elastic effects (generating) and inertial effects (stabilising); however, as yet we have no analytical insight into this complex fluid behaviour.

In this work we begin by studying Stokes flow in the cross-slot geometry, in order to provide a theoretical foundation on which to base an analysis of viscoelastic flow. We use a conformal mapping to transform the cross-slot geometry (relatively complex geometry) into a unit disk (relatively simple geometry). The velocity and the stream function in the cross-slot can be calculated and the curved streamlines can be drawn. We will build on this solution to address linear instability and the effects of viscoelasticity.

# Thursday 11:20 LT 24 (Level 10) / Track 8

On the Astarita flowfield and stress responses in 2D steady, homogeneous viscoelastic flows Robert J. Poole

University of Liverpool, Liverpool L69 3GH, United Kingdom

The two-dimensional steady, homogeneous flowfield proposed by Astarita (1991, Journal of Rheology, 35(4), pp.687-689) is studied for the Phan-Thien and Tanner (simplified, linear form) {sPTT} viscoelastic model. As the flow is steady and homogeneous, the sPTT model results also give

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the FENE-P model solutions via a simple transformation of parameters. The flowfield has the interesting feature that a scalar parameter may be used to vary the flow "type" continuously from solid-body rotation to simple shearing to planar extension whilst the rate of deformation tensor D remains constant (i.e. independent of flow-type). The response of the models is probed in order to determine how a scalar "viscosity" function may be rigorously constructed which includes flow-type dependence. By defining a frame-invariant coordinate system with respect to the eigenvectors of D, we associate a "viscosity" for each of the flows to a deviatoric stress component and show how this quantity varies with the flow-type parameter. For elliptical motions rate-thinning is always observed. For strong flows, i.e. flow-types containing at least some extension, thickening occurs and only a small element of extension is required to remove the rate-thinning inherent in simple shear flow in the model. Finally, a functional form of a viscosity equation which could incorporate flow-type, but be otherwise inelastic, the so-called GNFFTy (Generalized Newtonian Fluid incorporating Flow Type pronounced "nifty") model, is proposed.

#### Thursday 11:40 LT 24 (Level 10) / Track 8

#### Tensorial models for viscoelastic materials based on evolving relaxed states

Muhanna A. Alrashdi and Giulio G. Giusteri

#### Department of Mathematics, Università Degli Studi Di Padova, Padova, Padova 35121, Italy

Viscoelastic materials are ubiquitous in industrial processes involving polymers and mixtures of macromolecules with various suspending fluids. We present a model for viscoelastic materials [1] based on the observation that the microscopic arrangement of molecules determines the state at which the system would converge in the absence of applied forces. We incorporate concepts originated in the theory of solid plasticity and introduce an elastically-relaxed deformation tensor. The modelling effort focuses on the evolution of the relaxed state. We take as a basic request that, if we keep the material in a static configuration, then the relaxed deformation tensor should converge to the current deformation tensor.

Within this class of models, even the simple case with constant material parameters is able to qualitatively reproduce a number of experimental observations in both simple shear and extensional flows, including linear viscoelastic properties, the rate dependence of steady-state material functions, the stress overshoot in incipient shear flows, and the difference in shear and extensional rheological curves. Furthermore, by allowing the relaxation time of the model to depend on the total strain, we can reproduce the non-attainability of steady flows in uniaxial extension, and link this to a concept of polymeric jamming. Our framework is quite effective in reproducing experimental data of wormlike micellar solutions.

Another distinctive feature of our approach is the fundamental use of logarithmic strains, that leads to a proper generalisation to finite elastic deformations of the Maxwell model. Indeed, we can recover the upper-convected Maxwell model and the Giesekus model for the elastic stress evolution as different truncations for small elastic strains of the stress evolution implied by our model.

1. M. A. H Alrashdi, G. G. Giusteri, Evolution of local relaxed states and the modelling of viscoelastic fluids, preprint, https://arxiv.org/abs/2306.17242

#### Thursday 12:00 LT 24 (Level 10) / Track 8

## Tensorial viscosity models for non-Newtonian fluids

Emily Cook<sup>1</sup>, Duncan Hewitt<sup>2</sup>, and Mahdi Davoodi<sup>3</sup>

<sup>1</sup>University College London, London, United Kingdom; <sup>2</sup>DAMTP, University of Cambridge, Cambridge, United Kingdom; <sup>3</sup>Schlumberger Cambridge Research, Cambridge, United Kingdom

Many industrial fluids are engineered to exhibit viscoelastic behaviours which aid in industrial processes. As a result of elasticity, such fluids can exhibit a whole zoo of complex behaviour; in particular, they can experience normal stresses in flows where inelastic fluids would experience none. It is therefore of interest to efficiently capture these viscoelastic effects during modelling and simulation, as they could have a meaningful impact on the industrial application.

Relatively 'simple' generalised-Newtonian-fluid models are usually considered to be insufficient for modelling viscoelastic phenomena, because they impose an instantaneous relationship in which the stress and strain-rate tensors are always aligned. On the other hand, viscoelastic models, such as the Oldroyd B model, are implicit equations of the stress tensor, and are therefore difficult to analyse and can be computationally expensive or unstable.

This talk will outline and explore a class of constitutive equation which relates stress and deformation rate via a viscosity tensor. Such models allow for guaranteed control of normal stresses, without the requirement of implicit derivatives of the stress tensor, improving numerical tractability. The efficacy of such models in emulating some complex viscoelastic behaviour, and their association with traditional viscoelastic models, will be discussed.

# Thursday 12:20 LT 24 (Level 10) / Track 8

**NF20** 

# Comparison of Methodologies for the Application of the Onsager Variational Principle to Rheology Kwang Soo Cho

#### Department of Polymer Science and Engineering, Kyungpook National University, Daegu, Republic of Korea

The original Onsager variational principle is a mathematical generalization of classical linear irreversible thermodynamics formulated by an inductive manner. Doi, J. Phys.: Condensed Matter (2011) proposed a modification of the Onsager variational principle for isothermal process and applied it to various complex fluids such as viscoelastic fluid, liquid crystal and gel. We find some shortcommings of Doi's modification. The first is the methodology is applicable to only incompressible fluid. The second is the relation between stress and internal variables, maybe called

**NF19** 

stress equation, is based on poor approximation. In this talk we introduce deductive derivation of the Onsager variational principle and remedy for the shortcommings of Doi's modification.

# Thursday Afternoon

# Symposium MR

# **Microrheology and Microfluidics**

Organizers: Sepideh Khodaparast, Anke Lindner and Monica Oliveira

Thursday 2:00 LT 1 (Level 7) / Track 1

#### **Exploring Microfluidic-Small Angle Neutron Scattering for Soft Matter Physics**

Shivani Sathish<sup>1</sup>, Thomas Moragues<sup>2</sup>, Amy Shen<sup>1</sup>, and Viviane Lutz-Bueno<sup>3</sup>

<sup>1</sup>Micro Bio Nanofluidics Unit, Okinawa Institute of Science and Technology Graduate Univers, Okinawa, Japan; <sup>2</sup>ETH Zurich, Department of Chemistry and Applied Biosciences, Zurich, Switzerland; <sup>3</sup>Laboratory of Neutron Scattering and Imaging, Paul Scherrer Institute, Zurich 8032, Switzerland

Here we contribute to the fundamental understanding of non-equilibrium soft matter physics by offering an in situ experimental methodology for studying structural changes under flow, as well as their impact on materials properties and processing. Our research covers diverse model systems, ranging from wormlike micelles to 3D printing ink development. We address challenges in studying structural changes under flow by developing microfluidic-small-angle neutron scattering (microfluidic-SANS), overcoming limitations in sample volumes and time scales caused by low neutron fluxes. Microfluidic devices are typically made from polydimethylsiloxane elastomer (PDMS) due to its exceptional optical properties. However, PDMS is unsuitable for SANS sample cells, because of the high incoherent background signal induced by the hydrogen content. To address this, we employ selective laser-induced etching (SLE) to fabricate neutron-transparent fused silica microchannels. By parallelizing the flow into 16 microchannels, we increase sample volume exposed to the beam by overexposing multiple channels, while maintaining high spatial and temporal resolution provided by microfluidics. This approach enhances visibility and control of fluid processing during SANS experiments, facilitating in situ contrast matching, mixing, and time/spatial-resolved experiments with small sample volumes, opening opportunities for innovative science cases and experiments. Beyond device fabrication and usage, we explore methods to reduce reflection signals from microfluidic edges and present in situ contrast matching, as well as spatially and time-resolved flow mapping experiments. These efforts significantly contribute to our understanding of soft matter structures under flow conditions, which are stimulated with the use of contrast matching SANS. In conclusion, this research not only advances fundamental non-equilibrium soft matter studies but also holds practical applications in industries like pharmaceuticals, cosmetics, and 3D printing.

# Thursday 2:20 LT 1 (Level 7) / Track 1

**MR15** 

# Multidirectional analysis of multiple particle tracking data for simultaneous velocimetry and microrheology Maria Chiara Roffin<sup>1</sup>, Cheng Xuanhong<sup>2</sup>, Kelly M. Schultz<sup>2</sup>, and James F. Gilchrist<sup>2</sup>

<sup>IN</sup> Nottingham Trent University, Nottingham, United Kingdom; <sup>2</sup>Lehigh University, Bethlehem, PA 18015, United States

Colloidal suspensions are the basis of many products commonly used in everyday life, but their characterization is non-trivial especially for complex formulations. In addition, many of these materials undergo processes that include flow during end-use. When studying these materials it important to know the fluid properties and the external forces applied to the system. However, during experimentation it is often impossible to separte those two measurements. The work presented here develops a new microrheologocal technique that can characterize simultaneously the properties of the fluid and the velocity of the tracer particles, when undergoing unidirectional flow. The fluid properties are calculated using multiple particle tracking microrheology (MPT). MPT measures the Brownian motion of the tracer particles and it relates it to the rheological properties of the fluid. When tracer particles move unidirectionally due to the external force, they still undergo Brownian motion in the direction perpendicular to the force. Here, 1D MPT is used to characterize the fluid properties. Simultaneously, the tracer motion parallel to the external force is used to quantify the particle velocity. One specific application of this analysis technique is shown here: thermophoresis. Thermophoresis is the migration of particle in a fluid driven by a temperature gradient. The applications of this phenomenon are numerous especially in biomolecular separation, despite no agreement has been found on the underlying physics. Experimentally, tracer particles are suspended in a fluid and a thermal gradient is applied to the system. With this technique we can characterize fluid properties such as viscosity from the particle motion perpendicular to the thermal gradient. While simultaneously calculate the thermophoretic velocity of the particles.

# MR14

# Thursday 2:40 LT 1 (Level 7) / Track 1 M Probing the Dynamic Viscoelasticity of the Cytoskeleton Using a Simple Microrheology and Microfluidic Approach

#### Abbie Davidson, John M. Girkin, and <u>Anders Aufderhorst-Roberts</u> Department of Physics, Durham University, Durham, United Kingdom

In living materials, changes in viscoelasticity emerge from regulated biological processes that are driven by highly local and specific chemical and mechanical stimuli. It remains challenging to recreate such complex changes in viscoelasticity during rheological testing. In this presentation I will discuss a simple approach that combines microrheology with diffusion-controlled microfluidics to probe the viscoelastic properties of dynamic biological materials in changing chemical environments. I will demonstrate how these methods can be applied to networks of a filament-forming protein known as vimentin, a key contributor to cell viscoelasticity that is responsible for maintaining the cell's mechanical integrity under large stresses and fast deformations. Through studies of cell-free vimentin networks we show that vimentin assembly can be controlled through simple diffusion of ions to in a manner reminiscent of processes in live cells. By tracking the Brownian diffusion of embedded probe particles and calculating viscoelasticity moduli we show that the rheological properties of these networks depend directly on the ionic concentration and diffusion rate. We also show that by exchanging ions of different valency, unambiguous structural transitions can be induced, where filaments form coarse bundled structures, which have an associated reduction in viscoelasticity. These structural changes are strikingly similar to those observed in living cells where vimentin filaments form dense "bundles" in regions close to the nucleus, suggesting that these living processes may be partially regulated by ionic strength. Finally, we map the chemical changes that induce this bundling using ion-sensitive fluorescent dyes, allowing a correlation of network viscoelasticity, structural changes, and chemical environment. Together this approach affords intriguing rheological insights into the mechano-structural properties of living cells while presenting a promising platform for other biological materials.

# Thursday 3:00 LT 1 (Level 7) / Track 1

Machine-learning-based measurement of relaxation time via particle ordering <u>Maurizio De Micco</u>, Gaetano D'Avino, Massimiliano M. Villone, and Pier Luca Maffettone *Università degli Studi di Napoli Federico II, Napoli, Italy* 

The rheological characterization of complex fluids is of great importance in many fields of application. Among the properties that can be measured, the relaxation time has great relevance, as it provides a measure of fluid elasticity. Controlling this attribute is crucial for various applications as droplet formation, coating, mixing, and flow cytometry. In this work, we propose a novel method to estimate the relaxation time of viscoelastic fluids by means of the application of machine learning to microfluidics. Specifically, we train a Long-Short Term Memory (LSTM) neural network to identify the Weissenberg number (Wi) characterising the dynamics of trains of rigid particles suspended in a viscoelastic liquid flowing along a cylindrical channel. By looking at how the distribution of the distances among the particles evolves along the channel, the network is able to capture the contribution of Wi to the dynamics of particle trains, thus being able to estimate quite precisely the relaxation time of the suspending fluid. To train, validate, and test the LSTM architecture, we built an in silico dataset according to the approach proposed by D'Avino and Maffettone [1] to simulate the dynamics of particle trains in microfluidic flows of viscoelastic liquids. In particular, we fixed the particle volume fraction and confinement ratio, and we imposed a uniform distribution of interparticle distances at the channel inlet, i.e., that there is no preferential value of interparticle distance. Each example of the dataset is composed of the distributions of interparticle distances at different sections of a cylindrical channel, which are fed to the network to retrieve the Weissenberg number. The predicted relaxation time fairly agrees with the expected one. In addition, the proposed method is non-intrusive and can, in principle, be used to measure other properties of fluids. [1] D'Avino G., Maffettone P.L. (2019). Numerical simulations on the dynamics of trains of particles in a viscoelastic fluid flowing in a microchannel

# Symposium FR

# Food Rheology

# Organizers: Jan Engmann, Peter Fischer and Brent Murray

# Thursday3:50LT 1 (Level 7) / Track 1KeynoteOrigins of polysaccharide conformation and viscoelasticity in miscible heterogeneous solventGleb E. Yakubov<sup>1</sup> and Pallab Kumar Borah<sup>2</sup>

<sup>1</sup>Soft Matter Biomaterials and Biointerfaces, University of Nottingham, Nottingham, United Kingdom; <sup>2</sup>Soft Matter Biomaterials and Biointerfaces, University of Nottingham, Nottingham, United Kingdom

Polysaccharide polymers constitute 21 the fundamental building blocks of life and display a diverse set of conformational states which results in complex viscoelastic behaviour of their solutions; the origins of which needs further understanding. Utilising a model high molecular weight, high Trouton ratio 'pectin' polysaccharide extracted from okra (*Abelmoschus esculentus*) mucilage, we combine computer simulations and experimental data to unveil the underlying microscopic hydrodynamic origins of polysaccharide conformation. In miscible heterogenous solvents of water and glycerol, the polysaccharide chain undergoes a conformational transition from swelled-to-collapsed configurations, resulting in marked viscoelastic response. The conformational transition is entropy driven. Molecularly adsorbed water molecules have increased presence within ca. 0.40 nm of the chain surface with increase of glycerol in the solvent composition, thus indicating the emergence of preferential solvation. This

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FR1

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FR2

preferential solvation elicits an entropically unfavourable dynamic solvent heterogeneity, which is lessened by swelling and collapse of polysaccharide chains. Altering the preferential solvation layer by adjusting solvent composition allows for precise control of chain conformation and viscoelastic parameters. Our results provide an essential missing piece of the puzzle that is inaccessible through mean-field assumptions and offer new fundamental insights applicable in biological, biomedical, and engineering applications, including microrheological flows, microfluidics, bio-inkjet printing, as well as in pharmacological and food formulations.

# Thursday 4:30 LT 1 (Level 7) / Track 1

# Tribology & interfacial rheology: Explaining protein-stabilised oil droplet lubrication by non-linear interfacial rheology (LAOD)

# Jack Yang<sup>1</sup>, Lei Ji<sup>2</sup>, Leonard Sagis<sup>1</sup>, and Elke Scholten<sup>1</sup>

<sup>1</sup>Wageningen University and Research, Wageningen, The Netherlands; <sup>2</sup>Arla, Aarhus, Denmark

There is a global transition ongoing: the transition from animal-based to plant-based ingredients, which aims to reduce the environmental impact of food production. One of such product are oil-in-water (O/W) emulsions, which require stabilization of surfactants; and a plant-based alternative are plant-based proteins. In this work, we study a particular property of the plant-based oil droplets; the ability to lubricate. Oil droplet lubrication is crucial in foods upon swallowing. We studied lubrication properties using tribology, where we analysed the lubrication properties of milk-(included as control), pea- and soy-protein-stabilised oil droplets. We show that milk protein-stabilised oil intact droplets can lubricate the surfaces by rolling between surfaces. The soy protein-stabilised oil droplets broke and lubricated by free oil. Interestingly, the pea protein-stabilised oil droplets showed an intermediate behaviour. The intactness of the droplets was analysed using CLSM. The lubrication behaviour of the droplets could be explained using interfacial rheology in a drop tensiometer. O/W interfaces stabilised by the proteins were created and subjected to large amplitude oscillatory dilatational (LAOD) rheology. Amplitude and frequency sweeps were performed, and deformations in the NLVE regime were analysed using Lissajous-Bowditch plots. The rheology data showed that the milk proteins formed a stiff viscoelastic layer. The pea and soy proteins formed weaker layers, and especially the soy proteins formed very mobile interfaces. This suggests that oil droplets with a weak interfacial layer tend to break upon lubrication (e.g. soy protein droplets), while droplets with a stiff layer (e.g. milk protein droplets) remain intact. Our work shows a strong link between lubrication behaviour and mechanical properties of an interfacial film. We show an exciting toolset to understand the lubricating properties of oil droplets, especially droplets that are stabilised by novel ingredients, such as plant-based ingredients.

# Thursday 4:50 LT 1 (Level 7) / Track 1

# Crystallization of the disperse phase influences the oral tribology of O/W-emulsions

Philipp Schochat, Lina Lepp, Heike P. Karbstein, and Nico Leister

Food Process Engineering, Karlsruhe Institute of Technology, Karlsruhe 76131, Germany

Suspo-emulsions are used for food, cosmetic and pharmaceutical products. This also includes emulsion-based food such as conventional dairy products and non-dairy alternatives. Product properties such as flow behavior or sensory perception of the non-dairy products do not always correspond to those of conventional products thus consumer perceive them as products with inferior quality. It is assumed, that one reason for the deviation in quality is the crystallization of the triglycerides leading to differences in the solid fat content. In the context of this publication, this is discussed in terms of the solidity as a parameter of suspo-emulsions. By using low and high melting triglycerides and by measuring at different temperatures the solidity was varied. The disperse phase content of the suspo-emulsions was 30%. Rheological frequency sweeps were carried out within a temperature range of 5 °C to 50 °C. As expected, all suspo-emulsions showed a decreasing dynamic viscosity with increasing temperature. The differences in solidity (hard, pasty or liquid state) of the disperse phase caused no differences in viscosity at each temperature. In contrast, oral tribology can detect differences in solidity of the disperse phase. With the one ball on three pins method, the suspo-emulsions were examined. The friction coefficient was determined for different rotational speeds (0.001-100 mm/s). With increasing solidity of the disperse phase, the friction coefficient increases. For each suspo-emulsion, a friction profile can be determined. Comparing the profiles of a butter fat emulsion with those of plant-based fat emulsions, different plant-based fats can be mixed, to mimic the friction profile of milk products in plant-based alternatives. For product development, the disperse phase content can be used to tune the viscosity and the solidity of the triglyceride phase can adjust the friction values.

#### Thursday 5:10 LT 1 (Level 7) / Track 1 **Real-time rheology of soft matter-Based Food**

Simona Russo Spena and Nino Grizzuti

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The physics of food flow and food cooking has received special interest in recent times [1], to the point that a special issue of a well-known scientific journal has been fully dedicated to "Kitchen flows" [2]. Among other experimental techniques, rheology is an important tool for the characterization of food and food process properties. On the one hand, this is due to the fact that many food species belong to the rheologist much-loved soft matter family. On the other hand, it is well established that rheological properties strongly correlate with many other processing and final (sensorial) properties. In this work we apply real time rheology to explore the behaviour of some types of food during the cooking process. To this end, we use a temperature controlled rheo-reactor apparatus [3] where a special mixing tool (a double helix rotor immersed in a cylindrical cup) is used with the dual function of measuring viscous and viscoelastic properties during food elaboration and guaranteeing at the same time continuous stirring and homogeneity. Examples of rheo-cooking include the preparation of lemon slush, custard, gelatin-based candies. The effect of processing and material parameters, such as temperature and thermal history and recipe formulation, are considered and discussed. The effect of such parameters on the final, sensorial properties is also analyzed by texture analysis of the produced food.

FR3

FR4

References [1] A.J.T.M Mathijssen, M. Lisicki V.N. Prakash, E.J.L. Mossige, (2023), Rev. Mod. Phys., 95, 025004. [2] G.G. Fuller, M. Lisicki, A.J.T.M. Mathijssen, EJ.L. Mossige, R. Pasquino, V.N. Prakash., L. Ramos, (2022), Phys. Fluids, 34, 110401. [3] E. Paruta-Tuarez, P. Marchal, L. Choplin, (2014) ", Int. J. Adhesion Adhesives, 50, 32-36

# Symposium PF

# **Polymeric Fluids**

Organizers: Francesco Del Giudice, Richard Graham and Evelyne van Ruymbeke

Thursday 2:00 LT 17 (Level 7) / Track 2

#### Rheological characterisation of concentrated biopolymer systems

Anand Raja<sup>1</sup>, Philipp K. Wilfert<sup>2</sup>, and Stephen J. Picken<sup>1</sup>

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Biopolymers, in many cases, may be classified as being linear or branched polydisperse polyelectrolytes with ionizable functional groups. These groups are susceptible to variations in pH, ionic strength, and temperature, and give rise to hydrogen bonding, Coulombic interactions and hydrophilic or hydrophobic interactions respectively. These interactions lead to variations in both, the solubility in solution as well as the viscous dissipative flow properties. It is this latter aspect that is of prime relevance for the extraction, processing, and application of biopolymer systems. The working concentration range in these cases is greater than 1 g/dL and often as high as 10 g/dL; far above the dilute or semi dilute concentration regime where the viscosity of the polymer solution may be described using the Einstein equation, or indeed the Huggins or Kraemer equation respectively.

Thus, with the help of the Herschel-Bulkley model, we have documented viscosity and consistency index values for polymer concentration spanning five orders of magnitude. These values can be reduced to a universal behaviour versus scaled polymer concentration and compared alongside theoretical predictions made using a Taylor series like expansion as well as scaling arguments. To do so, we sometimes require large changes to the value of the intrinsic viscosity upon (de)screening the ionic charged interactions. As the intrinsic viscosity pertains to the size of the polymer coils in solution, such changes may then be related to changes in the persistence length. Therefore, we would also like to present our effort to capture changes to the persistence length and the intrinsic viscosity using the wormlike chain model in combination with the Odijk - Skolnick - Fixman model.

# Thursday 2:20 LT 17 (Level 7) / Track 2

**PF18** 

**PF17** 

# Carbon dots-based biopolymer composites: carbon dots synthesis, elaboration and study of composites with modulated properties

Enrique Manso Castillo, Véronique Thévenet, and Alain Ponton

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Carbon dots (CD) are zero-dimensional carbon nanostructures with a size in the range of nanometers that present photoluminescent properties, a wide variety of functional groups and excellent biocompatibility and nontoxicity [1]. As a new type of fillers, CD were incorporated in different polymer matrices to design advanced structural and multifunctional materials with significantly improved properties allowing their use in a wide range of applications [2]. However, their properties, depending on many parameters as reported in the literature, are scattered. A key-challenge faced by CD-based polymer composites involve a better knowledge of the relationship between the synthesis of CD, their properties and the elaboration of composites with their properties in regard of applications.

In this context, we have studied the effect of carbon dots on the modulation of hydrogel composite properties. For this purpose, CD synthesized from glucose using a one-step ultrasonic method and then purified by dialysis were introduced inside a sodium alginate matrix which was physically crosslinked using  $CaCO_3$  as a crosslinker. The introduction of CD inside the sodium alginate matrix allowed the elaboration of an optically responsive material due to the photoluminescent properties of CD, modulating at the same time the sol-gel transition kinetics as well as the thermo-mechanical and structural properties of the totally gelled composites. The results can be explained by the interactions between the negatively charged CD and both the oxygen rich alginate functional groups and the positively charged  $Ca^{2+}$  ions.

[1] Li, H., Kang, Z., Liu, Y., & Lee, S.-T. (2012). Carbon nanodots: Synthesis, properties and applications. Journal of Materials Chemistry, 22(46), 24230-24253. https://doi.org/10.1039/C2JM34690G

[2] Sui, B., Li, Y., & Yang, B. (2020). Nanocomposite hydrogels based on carbon dots and polymers. Chinese Chemical Letters, 31(6), 1443-1447. https://doi.org/10.1016/j.cclet.2019.08.023

# Thursday Afternoon

# Thursday 2:40 LT 17 (Level 7) / Track 2

# Shear rheology of unentangled pom-pom polymers

<u>Giovanni Ianniruberto</u><sup>1</sup>, Giuseppe Marrucci<sup>1</sup>, Guilhem P. Baeza<sup>2</sup>, Valerian Hirschberg<sup>3</sup>, and Salvatore Costanzo<sup>1</sup> <sup>1</sup>Department of Chemical, Materials & Production Engineering, University of Naples Federico II, Naples, Italy; <sup>2</sup>Matéis

UMR5510, INSA Lyon, Villeurbanne 69621, France; <sup>3</sup>Institute of Technical Chemistry, Clausthal University of Technology, Clausthal-Zellerfeld, Germany

Polymers with a pom-pom shape have attracted the attention of the scientific community since the pioneering work of McLeish and Larson [1]. Differently from the existing literature where entangled pom-pom polymers are typically examined [1,2] we here focus on nearly unentangled systems that in recent times have attracted the attention of the scientific community in view of their unexpectedly complex rheological behaviour [3,4]. In particular, we here perform linear and nonlinear shear rheology experiments on newly-synthesised polystyrene pom-pom melts in the unentangled regime. The data mostly refer to the viscosity, during the transient shear start-up, and up to the steady state.

We also analyse the observed rheological phenomena by using dumbbell-like molecular models as well as Brownian dynamics simulations of chains coarse grained at the Kuhn segment level. As is now well known, however, agreement with data can only be attained by also accounting for the glassy contribution, which in unentangled systems obviously plays a much more important role than in entangled ones [3,4].

[1] T.C.B. McLeish and R.G. Larson, J. Rheol 42, 81 (1998).

[2] M.C. Röpert, et al., Macromolecules 55 5485 (2022).

[3] Y. Matsumiya, et al., Macromolecules 51 9710 (2018).

[4] G. Ianniruberto, et al., Macromolecules 52 4610 (2019).

Thursday 3:00 LT 17 (Level 7) / Track 2

# PF20

# Computational Fluid Dynamics of Polymer Flow Induced Crystallisation using the polySTRAND model

William P. Grant<sup>1</sup>, Oliver Harlen<sup>2</sup>, and Daniel J. Read<sup>3</sup>

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Many polymeric materials are semi-crystalline. Under flow conditions crystallisation rates increase dramatically, owing to the polymer chains becoming more aligned during flow, so reducing the entropic penalty for forming crystals. A key question concerns the effect of polydispersity on both rheology and crystal nucleation rate. Long chains are expected to contribute most strongly to the viscoelastic properties, but also to become more aligned and hence more strongly favour crystal formation. The Rolie Double Poly (RDP) [1] model allows non-linear rheology to be predicted for polydisperse linear polymers, whilst the polySTRAND [2] model is designed to address the effects of polydispersity on crystallisation rate (predicting an enhanced contribution of long chains to the critical nucleus). In this work we combine these two models in a computational fluid dynamics (CFD) framework, allowing predictions for flow induced cystallisation in a complex flow geometry. The simulations are run using the open-source CFD program openFOAM [3], with the rheoTool [4] extension for non-Newtonian flow. This offers full customizability of the source code to tailor to our specific needs. We focus on a contraction-expansion geometry. We have produced results for a fully polydisperse system for polymer flow induced crystallisation. Most recently, we have incorporated temperature dependence and demonstrated how this affects the crystal growth rate. We will show predictions of flow induced crystallisation for this and compare to experimental data in the literature.

[1] V.A. Boudara, et al., J. Rheol, 63, 71 (2019). [2] D.J. Read, et al., Physical Review Letters, 124, 147802 (2020). [3] C. F. D. Direct, OpenFOAM user guide, (2015). [4] F. Pimenta and M. A. Alves, J. Non-Newt. Fluid Mechanics, 239, 85 (2017).

Thursday 3:50 LT 17 (Level 7) / Track 2

# Rheology of Topologically Active DNA

Davide Michieletto<sup>1</sup> and <u>Yair Augusto Gutierrez Fosado<sup>2</sup></u>

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Classic polymer physics successfully describes most of the polymeric materials we see everyday. Yet, it relies on the assumption that polymers do not change topology (or architecture) in time or that if they do alter their morphology, they do so in equilibrium. This assumption spectacularly fails for DNA in vivo, which is constantly topologically re-arranged by ATP-consuming proteins within the cell.

Inspired by this, in my group we study entangled systems of DNA functionalised by certain proteins which can selectively alter DNA's topology and architecture in time and may expend energy to do so. Solutions of "topologically active" polymers are a new class of polymeric systems that display unconventional out-of-equilibrium viscoelastic behaviours and can be conveniently realised in vitro.

In this talk I will showcase some of our recent work in this field, using a combination of theory, simulations and experiments and spanning a range of length and time-scales from single molecule to bulk rheology. For instance, I will discuss our results on the microrheology of entangled DNA undergoing digestion by restriction enzymes [1], condensation by T4 ligase [2] and loop extrusion by condensin, topological gelation of recombining rings [3] and the thinning of DNA solutions by Nucleoid Associated Proteins [4]. Finally, I will present in detail new results on the "topological elasticity" of limited valence gels of DNA nanostars [5], in which we unexpectedly found that the onset of interlinked topological motifs within these hydrogels fully control their elastic properties.

PF19

PF21

#### References

- [1] Michieletto et al, Nature Comm, 13013, 4389 (2022)
- [2] Panoukidou et al, https://arxiv.org/abs/2210.14010
- [3] Bonato et al, PNAS, 119, 44 (2022)
- [4] Fosado et al, Phys Rev Lett, 130, 058203 (2023)
- [5] Palombo et al, https://arxiv.org/abs/2308.09689

#### Thursday 4:10 LT 17 (Level 7) / Track 2

**PF22** 

**PF23** 

**PF24** 

# **Predicting elongational rheology of polystyrene pom-poms from molecular topology and shear rheology** <u>Valerian Hirschberg<sup>1</sup></u>, Max G. Schussmann<sup>2</sup>, and Manfred Wilhelm<sup>2</sup>

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Understanding the effect of branching on the melt rheological properties in shear and elongation is a fundamental challenge in polymer science. Besides the number and the molecular weight of the arms, the exact location of the branching points, i.e., the exact topology is key to control shear and extensional rheological properties. Fifteen low-disperse, polystyrene (PS) pom-pom samples were synthesized with optimized synthetic routes via a combination of anionic polymerization and grafting onto, in the scale of up to 300 g per model system. The molecular parameters of the pom-poms such as molecular weight of the backbone ( $M_{w,b} = 100 - 400 \text{ kg/mol}$ ), of the arms ( $M_{w,a} = 7 - 300 \text{ kg/mol}$ ) and the arm number q at each end (q = 5-30) are systematically varied. Additionally, the data are compared with PS combs. The shear and elongational behaviour are investigated experimentally and modelled with the pom-pom model. Whereas the pom-pom model cannot predict well the SAOS mastercurve, it can well-describe the strain hardening and high strain hardening factors beyond 100 in elongational flow. For the extensional viscosity four characteristic strain rate dependent regimes were found, defined exclusively through the LVE and the molecular topological parameters. At low strain rates, the whole molecule is relaxed, and the extensional viscosity equals the LVE. At medium strain rates and within the limits of the pom-pom model, the extensional viscosity can be predicted using the pom-pom model and the Considère criterion. As theoretically derived in literature and experimental found in this study, the extensional viscosity is increased above the linear-viscoelastic envelope (LVE) by a factor of [q<sup>2</sup>/ln(&Sqrt;3 q)], i.e. the strain hardening depends exclusively on the arm number of the pom-pom molecule. At high strain rates, brittle fracture with a constant fracture stress, which is universal to all PS, is observed.

# Thursday 4:30 LT 17 (Level 7) / Track 2

#### Viscoelastic properties of model linear polymers under elongational flow

Evelyne van Ruymbeke, Alexis André, and Céline Hannecart

IMCN, BSMA, Université catholique de Louvain, Louvain-la-Neuve, Belgium

Tube-based models allow us to quantitatively predict the linear viscoelastic properties of entangled polymers. However, for nonlinear elongational flows, large discrepancies between theoretical predictions based on the tube model and experimental results still persist, the model being unable to describe the qualitative differences observed in extensional flow of entangled polystyrene (PS) melts and solutions characterized by the same number of entanglements. In this work, we investigate the extensional flow behaviour of monodisperse and binary blends of linear polymers, to further understand the role of the molecular environment on the stretch state of a probe chain. For this purpose, we measured the nonlinear extensional responses of different polystyrene (PS) chains, both in the monodisperse and bidisperse state, and systematically vary the concentration and the molar mass of the chains. We also measure the elongational properties of linear chains of different chemistry. This allows us to highlight scaling relationships for describing the steady-state elongational viscosity. Our results show that the steady-state extensional viscosity of bidisperse systems follow a simple mixing rule only if both components are stretching. In relatively short, non-stretching chain matrix, long PS chains exhibit different transient strain hardening properties but the same apparent steady-state elongational viscosity. We further investigate this behaviour for long chains diluted in oligomers, as it is expected that too short chains enhance the strain hardening of the long component. By varying the composition of our solutions, we further quantify this effect. We then compare the elongational properties of the PS solutions to the properties of the linear chains of different chemistry.

# Thursday 4:50 LT 17 (Level 7) / Track 2

# Relations between architecture and rheology of linear and branched polypropylene melts

<u>Aurelio Salerno<sup>1</sup></u>, Aurelio Salerno<sup>2</sup>, Nino Grizzuti<sup>2</sup>, and Salvatore Costanzo<sup>2</sup>

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Polymer blends comprising both linear and branched architectures are widely investigated by both research community and industry aiming to shed light on the correlation between molecular structure, rheology, and processing of such materials [1,2]. The motivation arises from both theoretical (e.g. connection between molecular topology and dynamics) and experimental issues (e.g. predict polymer flow to achieve and optimize polymer melt processing) [1,2]. However, despite great progress, the implementation of polymer blending to predict the molecular structure that develops during synthesis of even most common industrial polymers, such as polyolefins, remains essentially out of reach. The main aim of this work is to combine the knowledge from polymer synthesis and molecular weight distribution (obtained by means of size exclusion chromatography) with the rheological characterization of linear and branched polypropylene blends, and to link these results with the aid of modelling tools capable to predict polymer architecture and branching features [3]. Blends of linear (L-PP) and branched polypropylene (LCB-PP), provided from Borealis, were prepared by solution precipitation process. The chosen L-PP/LCB-PP weight ratios were 0/100, 5/95, 10/90,

30/70, 50/50, 70/30, 90/10, 95/5 and 100/0. Small-amplitude oscillatory shear (SAOS) tests, creep tests and uniaxial extensional measurements were performed to assess the effect of L-PP/LCB-PP blends composition on the linear and extensional viscoelastic responses of the samples. The generalized tube-based branch-on-branch model (BoB) was used to fit the dynamics of the pure polymers and blends, and to find a connection among polymer synthesis, molecular weight distribution, rheology, and molecular architecture.

References: [1] Stange, J. and Münstendt, H. J. Cell. Plastics 2006, 42, 445. [2] Andriano, L. T. et al. J. Rheology 2020, 64, 663. [3] Read, D. J. Et al. Science 2011, 333, 1871. Acknowledgement: This work forms part of the research programme of DPI, project #855

#### Thursday 5:10 LT 17 (Level 7) / Track 2

Segmental dynamics of poly(ethylene oxide) rings in melts slightly contaminated by linear counterparts

<u>Dimitrios G. Tsalikis<sup>1</sup></u>, Dimitrios G. Tsalikis<sup>2</sup>, Harry I. Papargyriou<sup>3</sup>, Vlasis G. Mavrantzas<sup>4</sup>, and Vlasis G. Mavrantzas<sup>5</sup> <sup>1</sup>Department of Mechanical and Process Engineering, ETH Zurich, Zurich, Zurich 8092, Switzerland; <sup>2</sup>Department of Chemical Engineering, University of Patras, Patras, Greece; <sup>3</sup>Department of Chemical Engineering, University of Patras, Patras, Patras, Greece; <sup>4</sup>Department of Mechanical and Process Engineering, ETH Zurich, Zurich, Zurich 8092, Switzerland; <sup>5</sup>Department of Chemical Engineering, University of Patras, Patras, Greece

We have employed molecular simulations with a very accurate forcefield to probe microscopic dynamics in melts of long ring poly(ethylene oxide) (PEO), ranging in molar mass from about 10 kg/mol to about 90 kg/mol, either pure or slightly contaminated with linear analogues. Our main emphasis is on the effect of linear contamination on observables such as segmental dynamics, ring center-of-mass diffusivity, Kratky plots, and normalized single-chain dynamic structure factor that are directly probed experimentally by techniques such as SANS, NSE, and PFG-NMR. To enable a one-to-one comparison with such measured data, the simulations have been performed with PEO melts of exactly the same molar mass as those used in the experimental studies (between 10,000 and 40,000 g/mol). The mass fraction,  $f_L$  of the linear contaminant has been kept very low, below  $f_L$ = 0.1. We have also accessed long simulation times on the order of several microseconds.

Segmental and diffusion dynamics are examined in terms of the mean square displacement of ring segments and entire chains, respectively. We have also computed the dynamic structure factor S(q,t) as well as the rate of decay of several autocorrelation functions probing dynamics at several length scales. The simulations indicate significant changes in all the above-mentioned observables for melts contaminated even by as low as 3 % per mole by linear chains. For example, a linear contamination around  $f_L = 0.05$  seems to depress segmental and diffusion dynamics strongly. We also study how the presence of linear contamination, even at low fractions, can induce dynamic heterogeneity in the melt due to threading of a sub-population of rings by linear chains that significantly slows down their diffusive motion and orientational relaxation.

# Symposium IR

# Industrial Rheology, Sustainability and Additive Manufacturing

Organizers: Esther Garcia Tunon, Claire McIlroy and Sylvie Vervoort

Thursday 2:00 LT 6 (Level 8) / Track 3 **Rheological Optimisation of Battery Electrode Manufacturing** 

Carl D. Reynolds<sup>1</sup> and Carl D. Reynolds<sup>2</sup>

<sup>1</sup>School of Metallurgy and Materials, University of Birmingham, Birmingham, West Midlands B15 2SE, United Kingdom; <sup>2</sup>Faraday Institution, Didcot OX11 0RA, United Kingdom

Battery manufacturing is an increasingly important industry with worldwide requirements for rapid electrification of transport. It is vital that as manufacturing lines are established and expanded, that a through physical understanding of the manufacturing processes is established, to allow rapid adaptation to new technologies and prevent lines from becoming obsolete. Rheology is one of the key tools we can use to probe and optimise the process. Lithium and Sodium ion battery electrodes are currently produced by coating slurry onto a foil current collector. The rheology of this slurry is an early indicator of product quality and is key to determining the final coating microstructure, and the choice of coating process (slot die, blade coating, syringe deposition 3D printing) changes the requirements for the slurry significantly. Here, we discuss the key requirements for coating via these different processes, and use experimental results to show the impact of rheology on the coating manufacture. We then discuss formulation approaches to achieve the target slurry rheology and design principles for slurry and process design. By tailoring the rheology of the slurry, the coating process can be optimised and multi-level structuring from particle to coating can be achieved, improving performance, sustainability and efficiency of manufacturing.

**PF25** 

IR5

# Thursday 2:20 LT 6 (Level 8) / Track 3 IR6 Rheological, electrochemical and machine-learning based evaluation of carbon-based lithium ion battery anode slurry under in-line pipe flow conditions

Chan Hyeok Ahn and Kyung Hyun Ahn

School of Chemical and Biological Engineering, Seoul National University, Seoul, Republic of Korea

&nbspToday, many suspensions are characterized by a high solid content to achieve enhanced functionality, and lithium ion battery electrode slurry is a representative example of high solid content suspension(slurry). Effectively managing slurry with excellent dispersibility in large-scale processes is crucial for ensuring stability and optimal performance. However, the flow properties of the electrode slurries are hard to define clearly due to its complex and unclear rheological and micro-structural properties which are subject to potential changes by compositional and process conditions (variance by flow time, filtration, locally compositional changes). Therefore, comprehensive evaluation through various means, incorporating environmental variables, is necessary.

In this research, we evaluated the carbon-based anode slurry under pipe flow conditions with long time-scale. Bench-scale anode slurries that have different compositions and dispersion properties were made for the evaluation. The rheological evaluation was conducted by slurry sampling during the pipe flow. Also, continuous datasets of pressure, guide-wave velocimetry, and electrochemical impedance spectroscopy had been analyzed. For the massive continuous datasets of pressure, the datasets was normalized, and the machine-learning was applicated for the classification of slurry state with the flow time and compositional changes. It successfully classified slurries with the flow time and even small different composition changes.

#### Thursday 2:40 LT 6 (Level 8) / Track 3

Rheology as a tool to understand solid electrolyte formulation interactions and properties

<u>Nizar Ghazouani</u><sup>1</sup>, Guillaume Ovarlez<sup>2</sup>, Julien Jolly<sup>3</sup>, and Yaocihuatl Medina Gonzalez<sup>2</sup> <sup>1</sup>Laboratoire du Futur, Pessac 33600, France; <sup>2</sup>CNRS, Pessac, France; <sup>3</sup>Syensgo, Pessac, France

One of the key challenges in next generation all solid state batteries (ASSB) is understanding the physicochemical mechanisms impacting the solid electrolyte (SE) performances. A better comprehension of the interactions between the slurry components allows a better control of the final process to obtain the best SE film quality. Rheology is involved during the formulation phase to investigate the particle-solvent-binder interactions. For the binder-solvent rheology, the evolution of the flow considering a good or a theta solvents is in accordance with the polymer spatial configuration proposed in the literature. Koekoekx et al. found that micellious form is obtained for theta solvents while a polymer network with good solvents. The particle-solvent system shows a yield stress behavior with no thixotropy. However, at the flow onset, two critical shear strains have been observed. The largest one is attributed to the network of colloidal interactions and the smallest critical strain is attributed to particle contact. In order to investigate the origin of the small critical strain, the evolution of the torque with the height of immersion of a Vane geometry is studied, which allows attributing these contact forces either to friction or adhesion. Interestingly, adding the binder in this suspension decreased the viscosity and the yield stress associated with the small critical shear stress. This observation supported by sedimentation tests lead to the interpretation that the binder has a lubrication effect. Sedimentation tests were also carried out in different solvents to assess the impact of the solvation power and of the particle-solvent interactions on the slurry rheology. Moreover, the experiments show that the addition of the binder reduces the overall particle size and compactness of the sediment.

# Thursday 3:00 LT 6 (Level 8) / Track 3

Rheological behaviour of nanostructured h-BN water-based drilling fluids under HPHT conditions

Yago Soares<sup>1</sup>, Nathália Fernandes<sup>2</sup>, Lara Berghe<sup>1</sup>, Hélio Ribeiro<sup>2</sup>, Mônica F. Naccache<sup>1</sup>, and <u>Ricardo Andrade<sup>2</sup></u> <sup>1</sup>Mechanical Engineering, PUC-Rio, Rio de Janeiro, Brazil; <sup>2</sup>Engineering School, Mackenzie Presbyterian University, São Paulo, Brazil

An increasing number of industries is applying efforts in the development of smart and ecofriendly fluids, and rheology is an important tool to this end. Specifically in the oil industry, new oilfields are being identified in complex reservoir environments, such as high pressure and high temperature (HPHT) reservoir condition. Currently, extensive research efforts are dedicated to the development of high-quality water-based drilling fluid formulations, aiming to provide an environment and economic alternative to oil-based drilling fluids in high-pressure and high temperature conditions. This research aims to investigate the influence of variation in parameters such as temperature, pressure, and concentration on the rheological behavior of xanthan gum (XG) suspensions with dispersed two-dimensional (2D) oxidized hexagonal boron nitride (hBN-Oxi) nanostructures. Through structural characterization it was possible to prove the successful oxidation of the nanoparticles, which culminated on a better exfoliation and dispersion of hBN-oxi in XG. The main results revealed by rheological studies demonstrated that the effects of temperature and pressure on viscosity are less felt in suspensions with higher concentrations. More specific it was observed a slight decrease in viscosity for the pure XG at low temperature, but as the temperature increases and with the addition of the nanoparticles the pressure effect was suppressed. The experimental results helped to investigate the particle-particle and particle-fluid interactions mechanisms in water-based drilling fluids reinforced with hBN-oxi, and the effects on the rheological behavior.

IR7

IR8

## Thursday 3:50 LT 6 (Level 8) / Track 3 Linking Rheology and Printability in Direct Ink Writing (DIW): advances and opportunities Rishav Agrawal and Garcia-Tunon Esther

School of Engineering, University of Liverpool, Liverpool L69 3GH, United Kingdom

Direct ink writing (DIW) is a three-dimensional printing technique used by researchers working in fields ranging from energy applications to bioprinting. For DIW, the materials should be shear-thinning soft solids, able to flow through narrow nozzles, recovering their structure upon deposition and retaining the predesigned 3D shape. This makes the formulation design and rheology of these soft solids very critical, but these aspects can sometimes be overlooked due to the high specialization required. Linking rheology and printability is a growing area of research amongst the DIW and rheology communities; both are now joining efforts with new reviews being recently published in the field[1]. We will present our most recent work linking rheology and printability through the fundamental understanding of printable and not-printable complex fluids for DIW using Large Amplitude Oscillatory (LAOS), Fourier Transform (FT) rheology[2], the Sequence of Physical Processes and recovery tests. Our methodology enables us to quantify the three rheological stages in the printing process: 1) flow during the extrusion process (or yielding process, flowability); 2) the recovery stage when exiting the nozzle (recoverability); and 3) the ability to retain the predesigned shape once the part is printed for optimum shape fidelity (material strength). We create printability maps using, for the first time, linear and non-linear rheological parameters using three Ashby-type[3] maps, one for each stage in the printing process: flowability, recoverability, and strength. We demonstrate that the three maps must be considered holistically for a fair assessment of printability and that any small change (e.g., solid loading) in a formulation could lead to dramatic changes in one or more rheological parameters, and consequently in the overall formulation's printability.

[1] Rau, D.A., et al., Prog. Mater. Sci., 2023: 101188 4(10) [2] García-Tuñón, E., et al., Phys. Fluids, 2023: 35(1) [3] Corker, A., et al., Soft Matter, 2019. 15(6): 1444-1456

# Thursday 4:10 LT 6 (Level 8) / Track 3

# Printing by yield stress fluid shaping

Philippe Coussot<sup>1</sup> and Anatole Geffrault<sup>2</sup>

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3D printing of yield stress fluids (i.e., materials able to flow like liquids only beyond a critical stress) is opening new doors in the fields of construction, food or medicine. The usual printing technique consists to extrude and then deposit a filament of yield stress fluid onto the previous layer. One major difficulty is that, during the deposition process, some instabilities may appear and have a catastrophic impact on the final 3D structure. Here we show that these instabilities can be predicted as a function of the material properties and/or the printing parameters. More generally, we study the deposit of a filament from systematic tests with a model yield stress fluid, varying the material yield stress value, the distance between the nozzle and the substrate, the extrusion velocity, the nozzle diameter, and the nozzle displacement velocity. We show that a diversity of patterns arises: drops, discontinuous lines, straight lines, meanders, alternating loops or translating coils. In particular, we could describe in detail the process leading to the breakage of a filament, which allows to determine the constitutive equation of the material under extensional flow [1] and to develop a technique for determining the yield stress from the drop mass [2]. Then, we demonstrate that the transitions (frontier curves) between pattern regions can be predicted by theoretical arguments. Finally, we present a generic diagram predicting the patterns observed as a function of dimensionless numbers function of the various characteristics of the system [3]. This provides a reference scheme for optimizing practical processes or devising specific deposit patterns, valid for a wide range of conditions and yield stress fluids.

[1] A. Geffrault, H. Bessaies-Bey, N. Roussel, P. Coussot, Extensional gravity-rheometry (EGR) for yield stress fluids, J. Rheology, 65, 887-901 (2021) [2] A. Geffrault, H. Bessaies-Bey, N. Roussel, P. Coussot, Instant yield stress measurement from falling drop size: The "syringe test", J. Rheology, 67, 305

# Thursday 4:30 LT 6 (Level 8) / Track 3

# 3D printing of electrofluids and its advantage in soft sensor fabrication

Niclas Hautz and Lola González-García

Electrofluids, Leibniz Institute for New Materials, Saarbrücken, Germany

Electrofluids are viscous, liquid composites formed by suspending electrically conductive particles in a non-conductive solvent. Above the percolation threshold concentration, the filler particles form a network, which leads to electrical conductivity of the suspension. The structure of the network determines the mechanical and electrical properties of the composite making each electrofluid unique. Due to the shear thinning behavior, electrofluids facilitate the manufacture of truly soft components. To demonstrate the versatility of a wide variety of electrofluids, we use different filler types ranging from fractals (carbon black) to sheet/flake (graphene) like particles in combination with polar (glycerol) and nonpolar (PDMS) solvents. We show the potential of electrofluids in device integration by using 3D printing to create reproducible and well-defined structures. For that, we generate printability maps based on rheological parameters such flow or yield point and the viscoelastic storage modulus, following the methods discussed in literature [1]. As for 3D printing, the non-linear behavior of the sample plays a crucial role, we measure our electrofluids in the large amplitude oscillatory shear (LAOS) region, by performing the Fourier-Transform (FT) rheology. The calculation of strain stiffening and shear thickening ratios, based on the FT coefficients, provides a deeper understanding of the non-linear behavior of our electrofluids. Making use of the conductivity of the electrofluids, we combine the FT rheology with in-situ electrical resistance measurements. We will discuss the correlation between the mechanical and the electrical network deformations by analyzing the strain stiffening and the shear thickening ratios

IR9

IR10

IR11

with the electrical resistance signal, and how this analysis provides a deeper understanding of the electromechanical properties of the electrofluids for their application as soft sensors. [1] García-Tuñón et al., Physics of Fluids 35.1, 2023

# Thursday 4:50 LT 6 (Level 8) / Track 3

# Particle-induced Instabilities in Uniaxially Elongated Thermoplastic Polymer Blends

Moritz Neukötter, Steffen Jesinghausen, and Hans-Joachim Schmid

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The development and expansion of Selective Laser Sintering (SLS) in additive manufacturing is currently hindered by a limited supply of new materials. The use of additives, fibers, or particles could solve problems such as flame retardancy, or higher stiffness, but the uniform distribution in dry blends is problematic. Therefore, filled materials should be preferred. One approach is the direct incorporation during production in novel manufacturing processes such as Filament Extension Atomization (FEA). In FEA, polymer filaments are continuously stretched at high elongation rates until instabilities form and the filaments decay into droplets. To obtain initial results, we investigated diluted, semi-diluted and concentrated/entangled polymer solutions under uniaxial elongation. The resulting droplet size distribution (DSD) for diluted and semi-diluted solutions is random, rather wide and not applicable for SLS. With increasing polymer concentration, the number of droplets decreased significantly. At this point, particles can be used to create defined instabilities solving two problems at the same time - obtaining a narrow particle size distribution (PSD) while simultaneously creating filled particles. We found in preliminary experiments that the resulting DSD is strongly influenced by the PSD of the added particles and is approximately twice the particle size. Additionally, it seems to be independent of the particle concentration. For polymer melt suspensions, droplet formation time increases with the viscosity. Experiments with polymer melts were performed using several blends to adjust the melt properties and the results will be presented in the talk. The focus of the investigations was the general onset of the instabilities and the droplet sizes as a function of viscosity and particle concentration. The results will be compared to polymer solutions and discussed whether the number and size of the droplets can also be selectively adjusted in polymer melts.

# Thursday 5:10 LT 6 (Level 8) / Track 3

IR13

IR12

# **Viscoelasticity of vinylogous urethane vitrimer particles and selective laser sintering additive manufacturing** Laura Ballester-Bayarri<sup>1</sup>, Nicholas Ballard<sup>2</sup>, Mercedes Fernández<sup>1</sup>, and Robert Aguirresarobe<sup>1</sup>

<sup>1</sup>POLYMAT and Department of Advanced Polymers and Materials, University of the Basque Country, San Sebastián, Gipuzkoa 20018, Spain; <sup>2</sup>POLYMAT, University of the Basque Country, San Sebastián, Gipuzkoa 20018, Spain

Covalent Adaptable networks, and more specifically the so-called Vitrimers, emerged as a more sustainable alternative to conventional thermosets due to their capacity to be reshaped and eventually recycled. However, the difficulties in producing a macroscopic flow in real fabrication conditions limit their manufacturing capacities and this problem is even more important for the fabrication of three-dimensional objects. Thus, additive manufacturing of CANs has been presented as an alternative for expanding the fabrication possibilities of such materials. However, a successful fabrication requires understanding the materials' rheological characteristics. In this work, we focused on the capacities of vinylogous urethane vitrimers synthesized by suspension polymerization as feedstock materials for powder bed fusion additive manufacturing (Ballester-Bayarri et al. 2023). To gain insight into the sintering process, we developed an optical sintering set-up to visualize the evolution of the particle sintering, inspired by the work of Cardinaels et. al. (Hejmady et al. 2019). The neck formation between two particles was correlated with the rheological behavior of vitrimers through a liquid and viscoelastic model. In this context, we discussed the additional dissipations assigned to exchangeable bonds (i.e. Vitrimer Temperature, Tv).

Ballester-Bayarri L, Limousin E, Fernández M, et al (2023) Scalable synthesis of methacrylate-based vitrimer powders by suspension polymerization. Polym Chem 14:1656-1664. https://doi.org/10.1039/d3py00128h. Hejmady P, Van Breemen LCA, Anderson PD, Cardinaels R (2019) Laser sintering of polymer particle pairs studied by in situ visualization. Soft Matter 15:1373-1387. https://doi.org/10.1039/c8sm02081g

# Symposium SG

# **Suspensions and Granular Materials**

Organizers: Erin Koos, Wilson Poon and Olivier Pouliquen

Thursday 2:00 LT 18 (Level 8) / Track 4

SG21

# Influence of suspension rheology on dynamic wetting: Concentrated granular suspensions

<u>Reza Azizmalayeri</u><sup>1</sup>, Peyman Rostami<sup>1</sup>, Thomas Witzmann<sup>1</sup>, Christopher O. Klein<sup>2</sup>, Himanshu P. Patel<sup>1</sup>, and Günter K. Auernhammer<sup>1</sup>

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In concentrated suspensions, particle contacts induce strong contact forces between the particles. In this study, we compare suspensions of silica particles in two distinct dispersion media, each having a refractive-index match to the particles. We observe a significant variation in the rheological response of the suspension, including shear thinning, shear thickening, and a yield stress behaviour. The index-matching is based on
either a complex alcohol solution or a salt solution. These two sample systems are utilized for dynamic wetting experiments in which the contact line dynamics and internal structure of the suspension interact. At moving contact lines, the shear rate increases sharply towards the contact line. This localized shear rate profoundly affects the suspension's non-Newtonian rheological behaviour. At not too high particle concentrations, recent publications have affirmed the applicability of hydrodynamic solutions to granular suspensions. At higher particle concentrations, the impact of particles on the overall flow field and the microstructure of the densely packed granular suspension close to the contact line remains unexplored. In this study, a configuration consisting of a pinned droplet on a moving substrate is utilized. The receding contact line exhibits a dynamic curvature that undergoes a transient phase before reaching a stationary state. Using astigmatism particle tracking velocimetry (APTV), we precisely track the 3D motion of tracer particles within the suspension. Near the advancing contact line, we observe distinct behaviours based on the interaction between particles and the suspension rheology. Concentrated suspension of weakly interacting particles violates the hydrodynamic solution, especially when particle friction becomes a significant factor.

# Thursday 2:20 LT 18 (Level 8) / Track 4

# Signature of structural memory in shear jammed dense suspension

Sachidananda Barik<sup>1</sup>, H a Vinutha<sup>2</sup>, and Sayantan Majumdar<sup>1</sup>

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A wide verity of dense particulate suspensions can show a shear induced transformation from a liquid like state to a solid like jammed state called shear jammed (SJ) state. Such shear jammed system also show very fast relaxation dynamics where the stress in the system relaxes quickly so that the solid like jammed state comes back to the liquid like state when no shear is applied. Here we study the effect of such drastic stress relaxation over the structural relaxation. We observe that under unidirectional step strain perturbation there is an enhancement in the mechanical response whereas under the step strain perturbation in the alternative direction the mechanical response is week. This is suggestive of the structural memory in the system even after the dramatic decrease of the stress in the system and complete change of mechanical properties. We further find that this structural memory is significant only for the volume fractions above the minimum volume fraction required for the shear jamming. Using in-situ boundary imaging, shear reversal technique and numerical simulation we explore the possible microscopic mechanism and propose a model which can explain such structural memory in SJ dense suspensions.

#### Thursday 2:40 LT 18 (Level 8) / Track 4

#### Unravelling electrical and mechanical networks of electrically conductive suspensions

Dominik S. Schmidt<sup>1</sup>, Srishti Arora<sup>1</sup>, Sergio Lago-Garrido<sup>1</sup>, Antonio Núñez-Leyva<sup>2</sup>, and Lola González-García<sup>1</sup>

<sup>1</sup>Electrofluids, INM-Leibniz Institute for New Materials, Saarbrücken, Saarland 66123, Germany; <sup>2</sup>Química y Nanotechnología, Tecnologico de Monterrey, Monterrey, Mexico

Highly concentrated suspensions can be understood as liquid composites. By using electrically conductive particles as fillers, the material turns conductive for concentrations above the so-called percolation threshold. This indicates that a percolating 3D network, in which the particles are electrically connected, has formed. A concomitant effect might arise when such network forms: the formation of a mechanical network, which would lead to a gel-like behaviour of the material. We have found, however, that these two networks, although related, not necessarily form at the same time. We will show how depending on the particle shape, its surface chemistry, and the nature of the solvent used as dielectric matrix, the mechanical and the electrical networks form at the same or at different filler loadings, allowing a wide range of property combinations. Our systems are composed by a conductive filler; we used carbon black (CB), characterized by its fractal structure and multiwalled carbon nanotubes (MWCNTs), unfunctionalized and functionalized by carboxyl groups, and a liquid matrix, for what we used a molecular (Glycerol) and a polymeric (Polydimethylsiloxane (PDMS)) solvent. We studied the electrical percolation by means 4-point probe measurements and the mechanical percolation, expanding the classical equation for electrical percolation, using the storage modulus at the LVE region at a low frequency. Additionally, we measured in-situ electrical resistance of the materials during different rheological tests such as: amplitude sweep, flow curve, creep, and stress relaxation, which give us access to extra information of the mechanical network deformation and the recovery modes. Finally, we will discuss the application of these suspensions as truly soft electronic components such as sensors or stable conductors.

### Thursday 3:00 LT 18 (Level 8) / Track 4

### Extensional rheology of CNTs and cellulose nanofiber in wet spinning process

Gyeong Min Choi and Heon Sang Lee

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The wet-spinning process for producing highly oriented carbon nanotubes (CNTs) and cellulose nanofiber (CNF) is crucial for achieving superior properties. We present a theoretical model (Hyper-Spin-Line (HSL) model) for obtaining well-aligned CNT fibers, which may have superior properties. This theory is an implementation of the mesoscopic Leslie-Ericksen theory, considering the repulsive excluded volume potential acting in the direction opposite to the Frank elastic potential. Our theoretical model predicts the minimum draw ratio ( $D_R^0$ ) at various Deborah numbers (De) necessary for the target orientation of the fibers. The rotational relaxation time is proportional to the reciprocal of excluded volume, particularly for long CNTs with waviness. The existence of a minimum draw ratio is due to the large excluded volume potential of the long CNTs. The stable operation window in the De-D<sub>R</sub> plane is determined through linear stability analysis of draw resonance. Our analysis indicates that the desired orientation factor is achievable only when the draw ratio ( $D_R$ ) surpasses  $D_R^0$  within the stable operation window. The model predictions for the stability window and the effect of  $D_R$  on the properties of the fibers were confirmed by experiment. These Highly oriented fibers can be invaluable in existing and emerging applications, including aerospace vehicles, robotic arms, satellites, and electronic devices.

SG23

**SG22** 

# Symposium CG

# **Colloids and Glasses**

Organizers: Michel Cloitre, Johan Mattsson and George Petekidis

#### Thursday 3:50 LT 18 (Level 8) / Track 4 Central and Non-Central Interaction Forces in Rough Particle Colloidal Gels Florence J. Müller, Shivaprakash N. Ramakrishna, ISa Lucio, and Jan Vermant

Materials Department, ETH Zürich, Zürich, Switzerland

Particle roughness has emerged as a versatile tool for manipulating the properties of colloidal suspensions. Among those, notably, increasing particle roughness enhances the toughness of thermoreversible colloidal gels. In the present work, colloidal probe Atomic Force Microscopy (CP-AFM) measurements are used to quantitatively assess both changes in adhesive and frictional forces between rough thermoresponsive particles, independently adjustable through temperature and topography. The AFM measurements can be linked to changes in macroscopic rheological data on corresponding thermoreversible gels, whose macroscopic properties we then study. Our results underline the significant role of surface roughness in increasing the resistance to changes in the microstructure. Increased surface roughness leads to a significant reduction in the percolation threshold, higher moduli, and apparent yield stress for gels at low volume fractions. However, in densely packed systems, central adhesive forces become more significant due to reduced particle rearrangement. In summary, our study underscores that particles roughness will be able to act significantly when particles can rearrange by rotation.

#### Thursday 4:10 LT 18 (Level 8) / Track 4

#### Shear induced tuning and crystallization of colloidal gels

George Petekidis<sup>1</sup>, Vasiliki Chrisoulaki<sup>1</sup>, Emmanouil Vereroudakis<sup>2</sup>, and Esmaeel Moghimi<sup>3</sup>

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Combined rheology and confocal microscopy is used together with molecular dynamics simulations to probe microstructural changes in attractive colloidal gels under oscillatory shear. The system under study is a depletion gel of (nearly) monodisperse PMMA particles at large volume fraction (44%). We find that when such colloidal gels are sheared dynamically (i.e. applying oscillatory shear) they show a drop in their modulus which depends on the applied strain amplitude (?0), frequency and shearing time, similarly to polydisperse colloidal gels. In both experiments and simulations the drop of the modulus depends non-monotonically on the strain amplitude, with the maximum decrease in the modulus appearing around the yield strain. At larger strain amplitudes, the modulus returns to a similar value as the quiescent gels suggesting that any structural transition responsible for the modulus drop occurring is destroyed for large strain amplitudes. In this range, BD simulations show that the gels undergo a crystallization transition, observed in similar systems before which depends on the shearing time. Small crystallites are formed which later on grow into large crystalline domains, in agreement with confocal microscopy experiments coupled with rheology. The appearance of crystallization depends on both the frequency and the strain amplitude allowing tunability of the colloidal gel's mechanical properties and structure. Furthermore, it suggests that oscillatory shear leads the system towards phase separation, i.e. thermodynamic equilibrium.

#### Thursday 4:30 LT 18 (Level 8) / Track 4

Affine hierarchical compaction of drying beads of colloidal gel

Matteo Milani<sup>1</sup>, Luca Cipelletti<sup>2</sup>, Laurence Ramos<sup>3</sup>, and Christian Ligoure<sup>4</sup>

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The drying of amorphous soft solids comprising a solvent is a ubiquitous process that occurs in many industrial or biological phenomena, such as in coagulation, desiccation of concrete, clay or ceramics vases modelling and food conservation. Drying induces a volume shrinkage that modifies the structure at the microscopic scale, which in turn changes the mechanical properties of the materials. Fractal colloidal gels formed by the aggregation of attractive colloids are an ideal system to investigate the behavior of amorphous material undergoing a drying process. Here, we study the drying of boundary-free millimeter-sized beads of a colloidal gel. We find that, macroscopically, the bead undergoes an irreversible and homogeneous shrinkage, without showing delamination or cracks, which contrasts with findings for gels that adhere to a substrate. By uniaxial compression, we measure a huge stiffening of the beads whose Young modulus increases by several orders of magnitude when they dry. To follow the drying process both at the microscopic scales, we have developed a new dynamic light scattering (DLS) setup adapted to probe microscopic dynamics with spatial and temporal resolution for spherical samples. DLS reveals that stresses imposed by drying on the bead surface propagate homogeneously, inducing rearrangements throughout the bead volume. Moreover, we probe by XRD the structural evolution of the solid network of the gel during drying. Remarkably, we find that drying gels reorganize in a hierarchical manner, restructuring first at large length scales, then at smaller length scales, down to a few particle sizes. In short, by combining several techniques, we provide dynamic and structural insight on the homogeneous shrinkage of beads of fractal colloidal gels.

CG15

**CG14** 

CG16

#### Thursday 4:50 LT 18 (Level 8) / Track 4 Hydrodynamic lubrication in colloidal gels

Kim William Torre<sup>1</sup> and Joost de Graaf<sup>2</sup>

<sup>1</sup>Utrecht University, Utrecht, The Netherlands; <sup>2</sup>Utrecht University, Utrecht, The Netherlands

Colloidal gels are self-assembled networks of solid particles suspended in a liquid medium. They are widely used in everyday products and technologies, but their out-of-equilibrium nature makes it difficult to predict and control their structure. Hydrodynamic interactions --- which are dissipative forces mediated by the liquid solvent --- may affect the structure of non-equilibrium systems, leading to debates on their role in the formation and aging of colloidal gels. The current understanding is that the structural evolution of a gel is slowed down or sped up (depending on the volume fraction) by far-field hydrodynamic interactions. However, the emergent and aging gel network is the same when compared at equal `structural time'. The impact of near-field lubrication interactions remains poorly understood. As the interactions that lead to gelation are typically short ranged, they could compete with short-ranged forces driving formation. Here, we investigate how various approximations to modeling hydrodynamic interactions impact colloidal gels. We considered single-body (Brownian Dynamics), pairwise (Rotne-Prager-Yamakawa), manybody (Stokesian Dynamics), and lubrication-corrected many-body interactions. The dynamics of gel formation is sensitive to the exact nature of the approximation that is used. However, the steady-state structure is relatively unaffected, except when lubrication interactions are included. Counterintuitively, we also find that the effect of lubrication is to accelerate the aging of the gel. Both elements can be explained by lubrication interactions facilitating local rearrangements, which we will explain in detail. Our findings provide a solid foundation for the future study of colloidal gels subjected to external stimuli. We anticipate that despite their computational cost, accounting for lubrication interactions in the study of colloidal gels is crucial to accurately describe their dynamics.

### Thursday 5:10 LT 18 (Level 8) / Track 4

CG18

**BL21** 

# Perfume-induced phase transitions and rheological properties of a cationic vesicle dispersion for sustainable fabric softeners

### Manuela Walteros Leon<sup>1</sup>, Minne P. Lettinga<sup>2</sup>, and Christian Clasen<sup>3</sup>

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Fabric softeners are colloidal dispersions composed of double-tailed cationic surfactants that, when dispersed in water, assemble into vesicles. They are used in the final rinse of the washing cycle to form a protective layer on the fabric. Moreover, thickeners and fragrances are added to adjust flow properties and smell on the dispersion. It has been shown that the type of perfume induces significant changes to the macroscopic properties, mainly in the bulk rheology and viscoelasticity, and in the microstructure namely volume fraction, particle size, and modification in the bilayer structure. Temperature plays an important role on this phenomenon, since it allows the diffusion of the perfume within the bilayer leading to a characterization of thermal and rheological properties. In this paper, a study on the effect of two different type and perfumes concentration on new, sustainable vesicles is performed by using characterization tools as adiabatic and differential scanning calorimetry, thermorheology and broadband dielectric spectroscopy with the aim to link the phase transitions with the structural properties.

# Symposium BL

# **Bio-rheology, Living and Active Matter**

Organizers: Laura Casanellas, Alexander Morozov and Christian Wagner

# Thursday 2:00 LT 23 (Level 8) / Track 5 ON THE EFFECT OF THE ACTIVE STRESSES ON THE STRUCTURAL DYNAMICS AND HEMODYNAMICS IN MICROVESSELS

Antonis Marousis<sup>1</sup>, Konstantinos Giannokostas<sup>2</sup>, <u>Yannis Dimakopoulos</u><sup>3</sup>, and John Tsamopoulos<sup>4</sup> <sup>1</sup>Department of Chemical Engineering, University Of Patras, Patras 26224, Greece; <sup>2</sup>Department of Chemical Engineering, University Of Patras, Patras, Greece; <sup>3</sup>Department of Chemical Engineering, University of Patras, Patras, Greece; <sup>4</sup>Department of Chemical Engineering, University of Patras, Patras, Greece

Vascular tissue responds to local changes in hemodynamics by adjusting its lumen diameter and thickness, thereby regulating blood flow in homeostatic levels. The physiology of vasodilation is a combination of the passive mechanical properties of the constituent layers of the tissue, and the active contractile apparatus of smooth muscle cells generated in the Media. The latter is a complex interaction between mechanical, electrical and biochemical processes that by the intercellular calcium concentration. The abnormal contractility of the vascular wall is associated with many diseases, including hypertension, atherosclerosis and cardiovascular disorders. To better understand the biomechanical behavior of the vascular tissue and the hemodynamics in microvessels, we examine the 1-Dimensional two-phase blood flow, which incorporates the blood cellrich phase in the central core and the blood plasma in the annular part of the arteriole. For the rheological modelling of the blood core, a thixotropic elasto-viscoplastic (TEVP) model is adopted, whereas blood plasma is described as a viscoelastic material following the Phan-Thien and Tanner

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constitutive model. The vascular wall is represented as a two-layer fibre-reinforced hyperelastic material, incorporating the Media and Adventitia, that exhibits both passive and active responses. To model the anisotropic behavior of the tissue, we use the Holzapfel-Gasser-Ogden model. To solve the resulting Fluid-Structure Interaction problem, we adopt the Finite Element Method. A rise in the intercellular calcium intensifies the generated active forces, which in turn diminishes arteriole expansion. In addition, variations in the intercellular calcium have a notable effect on the thickness of the arterial wall, the configuration of the blood core-annular region and the developed wall shear and normal stresses.

# Thursday 2:20 LT 23 (Level 8) / Track 5

**BL22** 

**BL23** 

**BL24** 

#### **Micro-rheological examination of insulin-secreting B-cells in healthy and diabetic-like conditions and beyond** <u>Lukas Woolley<sup>1</sup></u>, Fotios Christakopoulos<sup>2</sup>, Adam Brubidge<sup>3</sup>, and Jan Vermant<sup>1</sup>

<sup>1</sup>Materials Department, ETH Zürich, Zürich, Switzerland; <sup>2</sup>Stanford University, Stanford, CA 94305, United States; <sup>3</sup>Nestlé Research Center, Lausanne, Vaud 1000, Switzerland

Pancreatic ß-cells regulate blood-glucose levels in the human body through glucose-stimulated insulin secretion. Hampered glucose homeostasis and reduced insulin secretion are hallmarks of type-2 diabetes, a disease that touches more than 6% of the world's population. The transport of the insulin vesicles towards the cell membrane prior to exocytosis is expected to be affected by changes in the viscoelastic and transport properties of the cytoplasm<sup>1</sup>. In this work, particle-tracking microrheological measurements we performed to quantify the viscoelastic properties in-situ by utilizing inert tracer particles on a rat insulinoma β-cell line model. To this end, we measured the ensemble average of all trajectories, to calculate the viscoelastic moduli from the mean squared displacement through the Mason method using the generalized Stokes-Einstein equation<sup>2</sup>. Intracellular processes, such as active transport, chemical reactions, and energy consumption, occur in non-equilibrium thermodynamic conditions, leading to a violation of the fluctuation-dissipation theorem. Even though the calculated viscoelastic properties can be characterized as apparent and can therefore slightly differ from the underlying material properties, the comparison between each evaluated condition within the experimental series can be compared.

Thus, it was found that the induction of diabetic-like condition through chronic palmitate treatment<sup>3</sup>, was identified to alter the viscoelasticity of the cytoplasm which diminished its reaction to glucose stimulation, giving rise to a less compliant and significantly slower relaxing cytoplasm.

<sup>1</sup> N. I. Mourad, et al., *American Journal of Physiology*, 299, C389-C398, (**2010**)

<sup>2</sup> G. Mason, *Rheologica Acta*, 39, 371-378, (2000)

<sup>3</sup> M. B.Hoppa, et al., *Cell Metabolism*, 10, 455-465, (2009)

### Thursday 2:40 LT 23 (Level 8) / Track 5

# The role of plasma for the mechanical properties of blood suspensions

Ursula Windberger<sup>1</sup>, Andreas Sparer<sup>2</sup>, and Kareem Elsayad<sup>3</sup>

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Blood plasma cannot only be viewed as a means of transport for cells and solutes. It is part of the entire blood system and ensures cohesion. Plasma components surround the blood cells, and the resulting matrix increases the functional diameter of the red blood cells by 100-500 nm and protects the cells from mechanical damage and thermal input. The plasma coating obviously supports the mechanical coupling between erythrocytes and the flowing part of the plasma. For this reason, linear aggregates align vertically to the direction of flow. Plasma is not always a Newtonian fluid, even when classic steel plates are used. Platelet-depleted porcine plasma is shear-thinning in Couette and capillary flow. Porcine plasma also showed higher longitudinal viscosity than human plasma using Brillouin scattering, a method not affected by surface tension. As a result, hematocrit-adjusted (HCT40%) porcine blood had higher yield stress than (HCT40%) human blood, although erythrocyte aggregation and fibrinogen concentration were similar. To conclude: plasma creates the connection between erythrocytes over distances greater than the distance between two adjacent RBCs in a rouleaux. Such collective behavior occurs if fluids have an elastic threshold, something that we have shown recently for blood plasma by modifying the plates of the rheometer to optimize wetting.

#### Thursday 3:00 LT 23 (Level 8) / Track 5

### Microswimmer propulsion and interactions in thin films

Bohan Zhang, Gerhard Gompper, and Dmitry A. Fedosov

Institute of Biological Information Processing, Research Center Juelich, Juelich 52425, Germany

Bacteria are able to migrate collectively over wet surfaces and form stable and highly motile aggregates. Collective locomotion of bacteria within aggregates is called swarming, and is affected by interactions between bacteria, their shape and the strength of propulsion, and the density of bacteria packing. To better understand the collective behavior of bacteria, numerical simulations of a large number of swimmers are performed. The swimmers are represented by the so-called squirmer model, in which their propulsion is imposed by a prescribed slip velocity field at the surface of the swimmer. This model allows the simulation of swimmers with different propulsion properties, including various motility types (e.g., pusher, puller) and propulsion strengths. We find that local interactions between swimmers mediated by the fluid environment determine their swarming behavior and the formation of clusters. In particular, swarming generally takes place at moderate volume fractions of swimmers, while at high swimmer densities, large non-motile clusters prevail. These results advance our understanding of bacterial aggregate formation and the connection between the collective swarming behavior and the internal properties of individual swimmers.

# Thursday 3:50 LT 23 (Level 8) / Track 5 BL25 T1 rearrangements in epithelial tissue are regulated by the balance of viscoelastic relaxation and cell-cell friction

Jocelyn Étienne

#### LIPHY, CNRS - Univ Grenoble Alpes, Grenoble, France

During development tissues undergo dramatic shape changes to build and reshape organs. In many instances, these tissue-level deformations are driven by the active reorganisation of the constituent cells. This intercalation process involves multiple cell neighbour exchanges, where an interface shared between two cells is removed and a new interface is grown. The key molecular players involved in neighbour exchanges, such as contractile motors proteins and adhesion complexes, are now well-known. However, how their physical properties facilitate the process remains poorly understood. For example, how do cells maintain sufficient adhesive contact while actively uncoupling from one another? Then, how does a new interface grow in a contractile environment? Many existing biophysical models cannot answer such questions, due to representing shared cell interfaces as discrete elements that cannot uncouple.

Here, we develop a model where the dominant element of the cell's cytoskeleton, their *junctional actomyosin cortex*, is modelled as a continuous viscoelastic rope-loop, explicitly representing cortices facing each other at bicellular junctions and the adhesion molecules that couple them. The model parameters relate directly to the properties of the key subcellular players that drive dynamics, providing a multi-scale understanding of cell behaviours.

We show that active cell neighbour exchanges can be driven by purely junctional mechanisms. Active contractility and cortical turnover in a single bicellular junction are sufficient to shrink and remove a junction. Next, a new, orthogonal junction extends passively. Our *Apposed-Cortex Adhesion Model (ACAM)* reveals how the turnover of adhesion molecules regulates tension transmission and junction deformation rates by controlling slippage between apposed cell cortices. Surprisingly, the model predicts that slower turnover of adhesions leads to faster T1 rearrangements thanks to the emerging mechanical balance.

#### Thursday 4:10 LT 23 (Level 8) / Track 5

BL26

# **Controlled mixing patterns in the small intestine lumen via coordination of active finger-like microstructures** <u>Clément de Loubens<sup>1</sup></u>, Rohan Vernekar<sup>1</sup>, Dacil Yanez Martin<sup>1</sup>, and Stéphane Tanguy<sup>2</sup>

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The gastrointestinal tract actively transports complex fluids through various organs, facilitating mixing with secretions and breaking down food particles for absorption while eliminating waste. Understanding flow phenomena in the GI tract is important for human health, as it impacts drug delivery, the spatiotemporal organization of the microbiota, and lead to health problems related to GI motility dysfunction. In this talk, we look at how different flow patterns in the intestinal lumen can be triggered by regulating the coordination of sub-millimetric finger-like structures known as villi. Initially, we present ex-vivo experiments illustrating that the motility of the rat's small intestine transitions from localized fast phasic longitudinal contractions, organized into uncoordinated domains, to propagating and coordinated longitudinal contractions as applied stress increases. This leads to varying degrees of synchronization between the villi. Building on these physiological observations, we develop a numerical model to simulate flow and mixing resulting from the active movement of villi. The numerical method is based on the two-relaxation-time lattice Boltzmann method, incorporating moving boundary conditions and generalized Newtonian fluids. Simulations reveal that the phase lag between villi can either induced transport along the longitudinal direction or radial mixing. By writing scaling laws based on the phase shift between adjacent villi, we demonstrate that these phenomena arise at first order from geometric effects. At second order, inertia effects induce also mixing and longitudinal transport over longer time scales. In conclusion, the small intestine, with a simple mucosal architecture at sub-millimetric scales, can tune flow and mixing patterns, by changing the degree of coordination between these structures. The time scales over which mixing occurs depend significantly on the rheological properties of the digesta.

#### Thursday 4:30 LT 23 (Level 8) / Track 5

**BL27** 

# Interfacial rheology of lung surfactant: experiments and modelling to explore disruption of function by aerosolised compounds

Hugh Barlow<sup>1</sup>, Sreyoshee Sengupta<sup>2</sup>, Maria Teresa Baltazar<sup>1</sup>, and Jorid Sørli<sup>2</sup>

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Lung surfactant is a complex mixture of phospholipids and proteins which resides at the fluid-air interface at the surface of our alveoli. Its function is to modify the surface properties of this interface in order to ease breathing and prevent lung collapse[1]. In particular, it increases the elasticity of the alveolar interface to prevent damage to epithelial tissues during breathing. Incidents of hospitalization have occurred when some commercial compounds are inhaled which disrupt the function of lung surfactant[2]. In this study, we explore this effect by combining experimental measurements and theory to understand the impact of a variety of chemical species on the rheology of model lung surfactant.

We examine the change in the dilational elasticity due to the introduction of a variety of compounds. Our experiments show that several chemicals induce changes in the dilational rheology of the lung surfactant monolayers which could lead to lung damage in vivo. Our findings are concordant with the effects observed in literature such as relative chemical potencies and the dose rate dependency[3]. To understand these effects, we develop a model based on the theory of surfactant adsorption kinetics which effectively reproduces the results seen in the experiments for all compounds studied.

#### References

[1] Veldhuizen et al. Role of pulmonary surfactant components in surface film formation and dynamics, Biochimica et Biophysica Acta (BBA) -Biomembranes, 1467, 2, (2000)

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#### Thursday 4:50 LT 23 (Level 8) / Track 5

# Interfacial rheology of DPPC-Cholesterol monolayers at air-water interface

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**BL28** 

**BL29** 

Pantelis Bourazanis<sup>1</sup>, Ivo Buttinoni<sup>1</sup>, and Laura Alvarez<sup>2</sup>

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In this study, we model pulmonary surfactants using binary lipid monolayers (DPPC-Cholesterol) at air-water interfaces and study their mechanical properties. The phase behavior of the lipid mixture was studied by obtaining the surface pressure isotherms using a Langmuir trough, and by continuing compression with two lateral barriers, we monitor the surface pressure and obtain the surface pressure isotherms, which showed the existence of the liquid expanded (LE) and the transition from this to the liquid condensed (LC) state. Various compositions of DPPC-Cholesterol were prepared in order to study the effect of cholesterol on the properties of the lipid monolayers. Our experimental findings suggest that cholesterol incorporation affects the phase behavior of the monolayer according to the surface pressure isotherms. Adding cholesterol leads to disappearing of the LE-LC coexistence and decreases the collapse surface pressure at ?~51-54 mN/m. The rheology results show that for all compositions, the viscous behavior (G'') dominates over the elastic response (G') especially in the low frequency regime. The surface shear viscosity measurements indicate that both DPPC and the DPPC/Chol mixture present a Newtonian response as it is almost unaffected by the frequency. Adding cholesterol (even at small amounts) generally affects the viscoelastic properties of DPPC: it decreases the surface shear viscosity and the elastic modulus at least 1 order of magnitude. These phenomena can be attributed to the decreased molecular ordering caused by cholesterol. We also investigated the phase behavior and the rheological properties of DPPC-Eicosanol mixtures at different compositions. Fatty alcohols are found in small amounts in biological membranes and they are used from scientists as model systems. Our aim was to investigate experimentally how a fatty alcohol such as eicosanol affects the mechanical properties and the phase behavior of a lipid monolayer when mixed with it.

### Thursday 5:10 LT 23 (Level 8) / Track 5

#### Active nematic-induced dynamics of passive polymers

# Zahra Valei and Tyler N. Shendruk

School of Physics and Astronomy, The University of Edinburgh, Edinburgh, Scotland EH9 3FD, United Kingdom

Rheologically complex materials are ubiquitous in living systems which are often driven out of equilibrium by biochemically fuelled active agents. The nonequilibrium effects then give rise to intriguing responses of polymeric inclusions. While previous studies have focused on active polymers in passive fluids [1] or passive polymers in athermal baths [2], passive polymers in active liquid crystalline solvents have not been modelled but are particularly interesting. This is because of the interplay between the anisotropy of medium, nonequilibrium activity and internal degrees of freedom of the macromolecules. This work reports the first study of a passive polymer in actively flowing nematics. We numerically study the conformation and dynamics of individual flexible polymers coupled to the velocity field of 2D extensile active turbulence. For this purpose, we employ a hybrid Multi-Particle Collision Dynamics [3] and Molecular Dynamics [4] simulation. We find the diffusivity of polymers increases with activity up to the onset of turbulence, after which it saturates. This is due to conformational changes of polymers in response to the active forcing. We see that long polymers are stretched by activity, while short polymers are compressed. This demonstrates how activity represents a pathway by which biological systems can control polymer properties. Future studies should consider macromolecular properties that introduce additional length scales, like stiffness and polymer concentration, to better mimic biological complexity and understand how biological systems self-organise their structure and function.

References: [1] Martín-Gómez and et al. (2019). Soft Matter, vol 15. [2] Harder and et al. (2014). Physical Review E, vol 90. [3] Kozhukhov and Shendruk (2022). Science Advances, vol 8. [4] Slater and et al. (2009). Electrophoresis, vol30.

# Symposium GR

# Geo-rheology

Organizers: Janine Kavanagh, Edward Llewellin and Sandra Piazolo

Thursday 2:00 LT 11 (Level 10) / Track 6

On the accumulation and release of bubbles in volcanic eruptions

Thomas J. Appleford<sup>1</sup>, Pragya Patel<sup>2</sup>, Outi Supponen<sup>2</sup>, and Maziyar Jalaal<sup>1</sup>

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In this study, we employ direct numerical simulations to investigate the effects of vibrations on the dynamics and release of bubbles in a volcanic conduit. We adopt a simple model in which a spherical bubble of compressible gas is immersed in an elasto-viscoplastic medium and subjected to periodic oscillations in the surrounding pressure field. It has been proposed that the response of entrapped bubbles to the vibrations is sufficient to yield the surrounding material and allow the bubbles to rise. In this work, we shed light on this hypothesis and study the influence of non-Newtonian material properties on the process. We also determine critical thresholds for the yielding of surrounding material and the consequent release of entrapped bubbles to occur.

#### 2:20 LT 11 (Level 10) / Track 6 Thursday

#### Rheology of dilute bubble suspensions in unsteady shear flows

Kohei Ohie, Yuji Tasaka, and Yuichi Murai

Faculty of Engineering, Hokkaido University, Sapporo, Japan

The viscoelasticity of dilute bubble suspensions is elucidated based on the constitutive equation proposed by Frankel and Acrivos (J. Fluid Mech. 44(1), 1970), which is originally for a dilute emulsion. Non-dimensionalization of the original tensor equation reveals that the viscoelasticity is systematized by three non-dimensional parameters: volume faction, capillary number Ca, and dynamic capillary number Cd, where Ca and Cd represent deformability of the suspended bubble and unsteadiness of its deformation. The relative viscosity and phase of viscoelasticity were comprehensively investigated regarding these three parameters. As a result, it was revealed that whether the viscosity increase or decrease depends on whether Ca or Cd exceeds a common critical value. In addition, the phase of viscoelasticity takes the minimum value at Ca  $\ll$  1 and Cd = 1, meaning that the bubble suspension has the most prominent viscoelasticity when the suspended bubble keeps a spherical shape and timescale of the shear deformation, that is, reciprocal of the applied oscillation frequency, is as same as the relaxation time of the bubble. Indirect verification of this theoretical work is conducted in a Taylor-Couette system in laminar shear flows, and experimentally measured wall shear stress agreed well with the predicted wall shear stress based on the theoretically derived viscoelasticity.

#### 2:40 LT 11 (Level 10) / Track 6 Keynote Thursday Convective patterns in planetary interiors: a question of complex rheology Anne Davaille

FAST, Orsay 91400, France

The cooling and evolution of a planet strongly depends on the convective regime prevailing in its mantle (e.g. the 2900 km-thick enveloppe of solid rocks under our feet on Earth). Geophysical data show that the planets and rocky satellites of our solar system undergo very different evolutions and present-day dynamics. But the conditions necessary for convection to generate plate tectonics and quasi-continuous resurfacing on Earth, episodic resurfacing on Venus, heat pipes on Io, or no resurfacing on Mars, remain strongly debated. The difficulty comes from the complexity of rocks rheology : ductile at high temperature and on long time-scale, brittle at low temperature and short time-scale. This « soft matter » behaviour can be recovered in the laboratory using hydrogels and aqueous colloidal dispersions, whose rheology can vary from viscous to elasto-visco-plastic to brittle when their temperature, and/or their water or ionic content, vary. We therefore have investigated the characteristics of thermal and solutal convection in those systems. They show a diversity of convective regimes, including the ones encountered in rocky mantles. Their physical understanding provides insights on the dynamics of planetary interiors.

### Thursday 3:50 LT 11 (Level 10) / Track 6

# GR9 Fluid rheology controls coupled solid-fluid processes in flux-driven fractures: Applications to magma transport

in dykes

Caitlin M. Chalk and Janine L. Kavanagh

Earth, Ocean and Ecological Sciences, University of Liverpool, Liverpool L69, United Kingdom

Dykes are thin, vertical, magma-filled fractures that provide the dominant transport mechanism for magma beneath the Earth's surface. Magma molten rock comprising melt, crystals and bubbles - has complex dynamics: melt is Newtonian, yet the presence of crystals and bubbles can cause magma to behave as a non-Newtonian fluid. Magma flow in dykes is also coupled with deformation of the fractured solid rock, making dyke propagation a coupled solid-fluid interaction problem. Understanding the multiphysical processes of dyke propagation is vital for interpreting rock records in the field, and understanding subsurface signals of magma movement in real-time. We conducted a series of scaled analogue experiments

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to investigate the effect of rheology on the fluid and solid mechanics during dyke propagation. Newtonian and non-Newtonian (shear-thinning) magma analogues were injected into gelatine (an elastic rock analogue) to create flux-driven fractures. Seeder particles - which fluoresce in laser light - were suspended either in the fluid or the gelatine in order to track material movements and measure fluid velocities and host rock deformation, respectively. We show that the rheology of the injected fluid controls the flow pattern within the propagating fracture - Newtonian fluids exhibit a central jet and recirculation, whilst non-Newtonian fluids have a uniform, unidirectional velocity profile. In turn, these different flow patterns lead to subtle differences in the elastic solid response. These results provide new insights on the coupled behaviour of magma flow and surrounding host-rock deformation of propagating dykes, and have important implications for interpreting rock records in the field.

#### Thursday 4:10 LT 11 (Level 10) / Track 6

GR10

### Effects of rheology on flow within experimental volcanic plumbing systems

#### Kate Williams, Janine L. Kavanagh, and Caitlin M. Chalk

Department of Earth, Oceans and Ecological Sciences, University of Liverpool, Liverpool, United Kingdom

Magma transport beneath the Earth's surface occurs within volcanic plumbing systems, which are composed of magma-filled fractures called dykes (vertical fractures) and sills (horizontal fractures). Understanding this transport is key to improving models of eruptible magma volumes and understanding the locations and mechanisms for key resource creation. Sills are capable of transporting magma hundreds of kilometres, and yet models for how this transport occurs are incomplete. Magma is often modelled as a Newtonian fluid, although experimental studies have shown that it is a multiphase fluid (silicate melt containing varying proportions of crystals and/or bubbles) with non-Newtonian, shear-thinning properties. Scaled analogue experiments were conducted to explore the evolving dynamics of magma flow in sills. Two fluids (magma analogues) with different rheology (water, a Newtonian fluid, or xanthan gum, a shear-thinning fluid) were injected through the base of a Perspex tank into two gelatine layers (crustal rock analogue) with a weak interface between them to encourage horizontal flux-driven fracture propagation. Particle image velocimetry (PIV) was used to track laser-fluoresced passive tracer particles within the injected fluids to map the fluid flow over time within the dyke-fed sill. The Newtonian fluid flow within the sill spread laterally from the entry point at a consistent velocity across the breadth. A region of focused flow was observed within the shear-thinning magma intrusions (such as basaltic sills). If such focused flow occurs in nature, enhanced by a non-Newtonian magma rheology, this could explain how larger sills develop with magma transport focused through a lower viscosity region localised within a larger volume of potentially eruptible magma.

#### Thursday 4:30 LT 11 (Level 10) / Track 6

GR11

# Understanding how complex rheologies in the magmatic system affect volcanic uplift episodes observed by InSAR and GNSS

Camila Novoa Lizama<sup>1</sup>, Dominique Remy<sup>2</sup>, Andrew Hooper<sup>1</sup>, and Juan Carlos Baez<sup>3</sup>

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In recent decades, the Global Navigation Satellite System (GNSS) and Interferometric Satellite Radar (InSAR) have provided the opportunity to monitor surface displacements at a large number of volcanoes. Uplift episodes have been observed worldwide prior to eruptions and have classically been interpreted as the filling of a magma reservoir at depth, and have therefore been identified as a possible precursor signal for volcanic eruptions. However, several studies have shown that this link between uplift episodes and eruptions does not always work. At present, the processes behind the uplift episodes are unclear and the classical models used to interpret them are being questioned due to their assumption of bedrock elasticity, neglecting the high temperature and presence of fluids expected in a volcanic reservoir, as well as the effects of bedrock failure due to magmatic intrusions. Here, by studying three volcanoes, we show how more realistic mechanical models can help to understand these uplift episodes. First, by analysing 10-year InSAR and GNSS time series and using a 3D finite element model simulating viscoelasticity in the magmatic reservoir, we show how the viscoelastic response of the reservoir to a magmatic intrusion can explain much of the observed uplift in the Laguna el Maule volcanic complex. Secondly, by incorporating elasto-plasticity into the model, we show that the magmatic intrusion associated with the observed uplift episode prior to the 2011 Cordón Caulle eruption was not sufficient to trigger the eruption. Instead, the movement of a nearby fault was able to connect the reservoir to the surface through a dilational plastic pattern triggering it. Finally, by analysing 6-year InSAR and GNSS time series during the Chillan eruption we study how two chambers connected by a conduit can help us to understand the dynamics of the supply process in volcanoes. With these examples, we show that models that take account of complex rheology are essential for advancing our understanding of silicic eruptions.

#### Thursday 4:50 LT 11 (Level 10) / Track 6

Viscous and brittle permeability development in three-phase rhyolite magma

<u>Taylor Witcher<sup>1</sup></u>, Rémi Champallier<sup>2</sup>, Laurent Arbaret<sup>2</sup>, Alexandra R L. Kushnir<sup>3</sup>, Michael J. Heap<sup>4</sup>, Michael J. Heap<sup>5</sup>, and Steffi Burchardt<sup>1</sup>

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The buildup of pore overpressures within magma primes a volcano for explosive behavior. Due to its high viscosity, silica- and/or crystal-rich magma remains coupled with vapor bubbles which nucleate and expand as the magma ascends (degassing). If the vapor cannot escape through a permeable network (outgassing), the stress applied by the growing bubble to the surrounding magma can locally exceed the magma's tensile strength, causing fracture propagation which could trigger a cataclysmic event. Investigating the rheological properties of high-viscosity magma like rhyolite helps us better understand its mechanisms of degassing and outgassing, which leads to better forecasting of dangerous volcanic events. For this study, we focus on systematic porous bands preserved in a shallow microcrystalline rhyolite intrusion located in Eastern Iceland. We coupled detailed microstructural analysis with a suite of torsion experiments run at 850-900°C and 100 MPa, at strain rates between 10<sup>-3</sup>-10<sup>-5</sup> s<sup>-1</sup>. Torsion results show increasing maximum stress (5-14 MPa) sustained by the rhyolite with increasing applied strain rate. The textures of the experimental products show a partially melted rhyolite with islands of microcrysts and deformed bubbles recording shear along melt pathways. Pore geometries within the bands taken from the field grade from sub-rounded, to undulating sub-planar, to planar fractures. We found that these pores likely formed as a result of shear localization during magma movement, eventually nucleating fractures at the tips of the planar pores. Groundmass quartz crystals are anhedral, while quartz inside pores are euhedral. This implies fluid transport within the pores, likely from magma degassing. The extent of the high-porosity bands throughout the laccolith suggests there was some degree of connectivity to the host rock, meaning the magma could outgas and consequently an eruption was avoided. This mechanism of degassing (and potential outgassing) should be considered in all high-viscosity magmatic syste

# Thursday 5:10 LT 11 (Level 10) / Track 6

Thermorheological lava flows: a model study

<u>David J. Fairhurst</u><sup>1</sup>, Wilson C. Poon<sup>1</sup>, Rory O'Neill<sup>1</sup>, Pablo Sandquist<sup>1</sup>, Edward W. Llewellin<sup>2</sup>, and Ceri Allgood<sup>2</sup> <sup>1</sup>School of Physics and Astronomy, The University of Edinburgh, Edinburgh EH9 3FD, United Kingdom; <sup>2</sup>Department of Earth Sciences, Durham University, Durham, United Kingdom

Describing the flow of solidifying melts ranging from lava to chocolate fountains requires understanding the interplay between a temperature dependent viscosity, heat flow onto a substrate and their subsequent effect on the shape of the fluid boundary. The computational fluid dynamics modeling of such systems has lacked experimental data against which to compare their predictions. We provide such data by flowing molten polyethylene glycol (PEG), a common model for lava, down an unheated incline. The extent of the flow, its surface profile and temperature gradient are simultaneously measured to provide data on the shape of the boundary with respect to Graetz number - comparing heat flow and advection speeds - and temperature. Morphological features such as levees, flattened and near-cylindrical profiles and fingering instabilities are mapped against these parameters.

# Symposium EM

# **Experimental Methods and New Rheometric Techniques**

Organizers: Christian Clasen, Dan Curtis and Manlio Tassieri

Thursday 2:00 LT 19 (Level 10) / Track 7

EM7

# Follow-Up of Rheological Properties Under Magnetic Hyperthermia Fast Sintering: Focus on the UHMWPE Explosion Upon Melting Question

Mathieu Salse<sup>1</sup>, Guilhem P. Baeza<sup>2</sup>, Julien Morthomas<sup>3</sup>, and Olivier Lame<sup>4</sup>

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We have developed an innovative rheological setup, coupling a PEEK plate-plate geometry with an induction heating system and an IR camera. The setup is designed to simultaneously and precisely measure the rheological properties and the temperature while applying a high frequency alternative magnetic field (855 kHz, 5 mT) within a rotational rheometer. By using this original method, we aim to unravel the mechanisms at work during the Ultra-High Molecular Weight Polyethylene (**UHMWPE**, M>1 Mg.mol<sup>-1</sup>) processing. The UHMWPE extremely high viscosity in the molten state prevents the utilisation of classic polymer processing methods (extrusion, injection...), obliging manufacturers to use sintering-oriented techniques, i.e. polymer grains interfaces welding through re-entanglement in the molten state ( $T>T_m$ ) and co-crystallization during cooling. Due to an abnormally fast mobility of the polymer huge chains, at the melting of the nascent crystals, the interfaces welding can be

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achieved in several minutes instead of dozens of hours according to classical reptation theory (see P.G. de Gennes' Explosion upon melting 1995). This mechanism remains however scarcely explained. Clarifications thus require in-situ observations during the nascent polymer melting. In this context, our setup allows the fast and homogeneous heating of the UHMWPE matrix (filled with 3 vol.% of Fe nanoparticles), revealing the impact of the melting explosion and subsequent re-entanglement on the dynamic moduli. Coupling these results with *i*) coarse grain molecular dynamic simulation of the nascent polymer, and *ii*) other experimental methods (SEM, SAXS, XPCS...), our work provides unprecedented elements to unravel the explosion upon melting phenomenon.

# Thursday 2:20 LT 19 (Level 10) / Track 7

EM8

EM9

**EM10** 

# **Exploring How Magnetorheological Cell Design Influences Measurement Accuracy in a Rotational Rheometer** <u>Rui Rodrigues</u><sup>1</sup>, Francisco J. Galindo-Rosales<sup>2</sup>, and Laura Campo-Deaño<sup>1</sup>

<sup>1</sup>Dept. Engenharia Mecânica, Faculdade de Engenharia da Universidade do Porto, Porto 4200-465, Portugal; <sup>2</sup>Dept. Engenharia Química, Faculdade de Engenharia da Universidade do Porto, Porto 4200-465, Portugal

Blood rheology is highly dependent on plasma viscosity, haematocrit and erythrocyte mechanical properties. Nevertheless, applying external forces such as magnetic fields to blood flow can affect its rheological behaviour. Thus, rheological characterisation of blood under the action of external fields is key to develop new treatment therapies. To this end, the magnetorheological cell of the rotational rheometer must offer accurate results. However, low and inconsistent apparent-viscosity values were systematically acquired with a rotational rheometer (MCR-302e, Anton Paar) equipped with a magnetorheological cell, without applied magnetic field. A parametric study was conducted experimentally and numerically to evaluate the error's source. Three Newtonian fluids with different viscosities were used: a calibration oil (3.659 mPa·s) and two glycerol and water solutions ( $\approx$  37 and 370 mPa·s); and three rotating upper geometries were tested: two parallel-plate (PP20, PP20 MRD) and one cone-plate (CP20 MRD), combined with an additional non-magnetic lower plate. For the parallel-plate geometries, the gap was varied from 0.05 to 0.35 mm, and for the CP it was kept constant (0.084 mm). The non-magnetic lower plate returned practically constant viscosities near the expected values for all geometries. However, with the magnetic bottom plate, the apparent viscosity decreased with diminishing gap in the case of the PP geometries, whereas the CP slightly improved the results. According to the experimental and numerical results, the complex design of the magnetic bottom plate could be responsible for this behaviour.

This work is supported by national funds through the FCT/MCTES (PIDDAC), under the project PTDC/EME-APL/3805/2021 - MagneticBlood - Magnetorheology of human blood towards the development of new treatment therapies, with DOI 10.54499/PTDC/EME-APL/3805/2021, LA/P/0045/2020, UIDB/00532/2020 and UIDP/00532/2020, and the program Stimulus of Scientific Employment, Individual Support-2020.03203.CEECIND.

# Thursday 2:40 LT 19 (Level 10) / Track 7

#### Optical methods to study structure breakdown in smart materials

Jan Haeberle, Jörg Läuger, and Loredana M. Völker-Pop

### Anton Paar Germany, Ostfildern 73760, Germany

Smart fluids like magneto-rheological fluids have led to development of outstanding applications as vibration control as well as in brakes, clutches, dynamometers and helicopter lag dampers [1]. While rheological testing provides information on the macroscopic behavior of the fluids under the influence of magnetic fields, the microscopic structure formed by these suspensions is what defines the material behavior. Particle alignment, structure forming and breakage are strongly influenced by the type of material and the composition, shape, size and concentration of particles as well as the physical properties of the carrier liquid. In addition to these parameters, the relationship between the applied magnetic fields and the external mechanical load strongly influences the structural changes of the smart fluids.

We study this interplay of macroscopic sample behavior and microscopic structure in two ways. First, the structure is visualized by combining established magneto-rheological devices with optical visualization methods. Second, the response of the smart fluid structure to mechanical oscillations at large strains is measured. The theoretical framework, experimental data and analysis results for large amplitude oscillatory shear (LAOS) on a commercial magnetorheological fluid will be presented.

The development of new measuring techniques aimed at characterizing the microscopic structure is made possible through the knowhow accumulated over the years from close cooperation with R&D departments and universities.

[1] J. S. Kumar, P. S. Paul, G. Raghunathan, and D. G. Alex, "A review of challenges and solutions in the preparation and use of magnetorheological fluids," Int. J. Mech. Mater. Eng., vol. 14, no. 1, p. 13, Nov. 2019, doi: 10.1186/s40712-019-0109-2.

# Thursday 3:00 LT 19 (Level 10) / Track 7

# Introduction of New Rotational Mini-Rheometer for Scarce Sample

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We introduce a miniaturized stress-controlled rheometer, aimed at enhancing rotational rheometry for analysis of scarce sample in material science and product development. This innovative device addresses the critical need for analyzing samples, such as biofilms, nascent drug formulations, and rare metal inks for microelectronics, which are typically limited in quantity due to their high production costs, challenging synthesis, or exclusive origins. Rheological measurements typically require at least 20  $\mu$ l of sample material using commercial rotation rheometer, a requirement significantly reduced by our device. The mini-rheometer, approximately the size of a 50mm cube, utilizes in particular an air bearing rotor made of quartz glass, manufactured via Selective Laser Etching (SLE). This subtractive 3D printing method for glass ensures the precision needed e.g. for air bearing with  $\mu$ m-scale gaps. An outer magnetic field generated by two orthogonal coil pairs drives a centrally-placed magnet. Torque precision is maintained through applied current and the phase angle between inner and outer magnetic fields. The angular displacement is accurately gauged by two Hall effect sensors, that detect the rotational movement of the inner magnet. The mini-rheometer's efficiency and functionality were tested performing a simple shear stress experiment on silicon reference oil. Notably, the employed cone-plate geometry only requires 2  $\mu$ l of sample material, representing a significant advancement in sample conservation. This development extends the benefits of rotational rheometry - such as a wide range of strain rates and homogeneous strain profiles - to experiments where sample availability is limited. Due to the device's miniaturized dimensions common, inverted microscopes could be used to study microstructural properties related to macroscopic flow behavior.

#### Thursday 3:50 LT 19 (Level 10) / Track 7

#### EM11

# Distinguishing Thixotropy, Plasticity and Viscoelasticity in Complex Fluids Using Parallel Superposition Gaborheometry

Joshua D. Rathinaraj<sup>1</sup>, Rishabh More<sup>1</sup>, Marc Graedel<sup>2</sup>, and Gareth H. McKinley<sup>1</sup>

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There has been a resurgence of interest over the past decade in the class of fluids known broadly as Thixotropic Elastoviscoplastic (TEVP) Fluids which exhibit a wide range of rheologically challenging phenomena including thixotropy, elastoplastic creep below yield and an age-dependent yield stress. We illustrate how modern oscillatory rheometric techniques can be used to help distinguish the distinct contributions to what may be referred to broadly as *mutating materials* with rheological properties that are typically both time- and rate-dependent. The relative importance of these effects can be quantified by three distinct dimensionless parameters; a mutation number, a Weissenberg number and a Mnemosyne number that quantifies the importance of thixotropic effects in a fluid. The typical rheologist's approach of using Fourier transforms for analyzing oscillatory data implicitly assumes the signals are time-translation invariant, which constrains the mutation number of a sample to be extremely small. This constraint makes it difficult to quantify shear-induced rheological changes in thixotropic and aging materials. In the present work we use the Gabor transform (a Short Time Fourier Transform (STFT) utilizing a Gaussian window) to provide optimal time-frequency resolution of the local viscoelastic properties of mutating materials. We combine parallel superposition flows with Gaborheometry to arrest the aging of a model thixotropic colloidal systems and simultaneously monitor the temporal evolution in the elastoviscoplastic response. By varying the relative strengths of the steady shearing component that "shear-melts" the microstructure and the superimposed oscillatory probe we show how one can unambiguously distinguish the time-dependent thixotropic aging response from local viscoelastic responses

#### Thursday 4:10 LT 19 (Level 10) / Track 7

A novel double exponential chirp signal for high noise environments

Jarno L. Waeterloos<sup>1</sup>, Gareth H. McKinley<sup>2</sup>, and Christian Clasen<sup>1</sup>

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Fixed frequency sine waves are traditionally used for small amplitude oscillatory shear measurements due to their easy and efficient processing. However, one requirement for these measurement that the material under investigation does not mutate, is generally not satisfied for fast timeevolving systems [1]. In order to follow the evolution of the structure and the related rheological properties of such systems, nature offers more suitable and optimised signals that can reduce the measurement time and increase information density for time resolved mechanical spectroscopy.

The optimally windowed chirp (OWCh) is such an optimised signal where the frequency is increasing with time in an exponential fashion and in a continuous manner [2]. While measurement times can be reduced by orders of magnitude, in high noise environments the required windowing of the OWCh comes with the major disadvantage of generating a noise dominated regime at high frequencies. This noise dominated regime often falls within the traditional measurement range affecting the measurement accuracy and resulting in high frequency artifacts. Changing the tapering parameter of the window delays the onset of the noise dominated region but does not provide a general solution to the problem. In this paper a new chirp signal is put forward with a sigmoidal time-frequency spectrum build up of two exponential functions as an alternative to the single exponential OWCh. This double exponential chirp (DEC) alleviates the high frequency artifacts, maintains the benefits of the windowing and has the added benefit of being versatile due to the free choice of inflection point. The effectiveness of the DEC is illustrated with orthogonal superposition rheometry as a high noise environment. Measurements on Newtonian and viscoelastic standards demonstrate a complete removal of the high frequency artifacts with the novel waveform of the DEC.

Mours, M., Winter, H.H., Rheol Acta 33, 385 (1994).
M. Geri, et al., Physical Review X. 8, 041042 (2018)

**EM12** 

# Thursday 4:30 LT 19 (Level 10) / Track 7

# Optimally Windowed Logistic Chirp (OWL-Chirp) Rheometry

Daniel J. Curtis<sup>1</sup>, Mohua Das<sup>2</sup>, and Gareth H. McKinley<sup>2</sup>

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Probing the rheological evolution of transient and mutating materials requires that their viscoelastic properties be determined as a function of both time and frequency. Undertaking such experiments using conventional rheometric approaches can be challenging. Optimally Windowed Chirp (OWCh) waveforms are ideally suited to such experiments since the acquisition of the frequency-dependent linear viscoelastic properties can be undertaken in a fraction of the time required by discrete frequency sweep methods. In such experiments, the frequency of the waveform evolves exponentially in time between low and high frequency limits,  $\omega_1$  and  $\omega_2$ , respectively. The linear dynamic moduli can then be determined as the ratio of the Discrete Fourier Transforms of the stress and strain (for strain controlled chirps), or stress and strain rate (for stress controlled chirps). However, the use of signals in which the frequency evolves exponentially can generate noise in the moduli at high frequencies due to DFT sampling effects. The situation is compounded by the very high skew rate,  $d\omega/dt$ , at high frequencies and the limited digital sampling rate of the A/D converter. Herein, we consider the use of an alternative chirp in which the frequency evolves between  $\omega_1$  and  $\omega_2$  according to a logistic function and demonstrate that significant improvement in the high frequency data can be achieved using Optimally Windowed Logistic (OWL) Chirps.

#### Thursday 4:50 LT 19 (Level 10) / Track 7

EM14

#### **Effect of Hencky strain on extensional rheology determined from entrance pressure drop measurements** Martin Zatloukal and Jiri Juracka

Department of Polymer Engineering, Faculty of Technology, Tomas Bata University in Zlín, Zlín 760 01, Czech Republic

In this work, new die [1] is proposed for the determination of the extensional viscosity at various Hencky strains from entrance pressure drop measurements using a high-pressure capillary rheometer. The new concept makes it possible to change the Hencky strain independently of the Hencky strain rate. The new die consists of an easily replaceable cylindrical chamber to which free-flow orifice die [2-5] is attached. The Hencky strain is controlled by changing the diameter of the cylindrical chamber while keeping the diameter of the free-flow orifice die the same. The new die was used to determine extensional rheology of various polyolefin melts and the results obtained were compared with corresponding data from the Sentmanat extensional rheometer.

Acknowledgement: The authors wish to acknowledge the Grant Agency of the Czech Republic (Grant No. 24-11442S) for the financial support and NETZSCH Gerätebau GmbH for providing the Rosand RH10-2 capillary rheometer.

References.

- 1. M. Zatloukal, Czech patent 307664 (2018).
- 2. M. Zatloukal, J. Musil, Czech patent 304382 (2014).
- 3. M. Zatloukal, J. Musil, Polymer Testing 28, 843 (2009).
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- 5. M. Zatloukal, Czech patent 309071 (2021).

#### Thursday 5:10 LT 19 (Level 10) / Track 7

**EM15** 

#### **Determination of uniaxial and planar extensional viscosity using a Rosand high-pressure capillary rheometer** <u>Levente Szántó<sup>1</sup> and Martin Zatloukal<sup>2</sup></u>

<sup>1</sup>NETZSCH Gerätebau GmbH, Selb, Bavaria 95100, Germany; <sup>2</sup>Department of Polymer Engineering, Faculty of Technology, Tomas Bata University in Zlín, Zlín 760 01, Czech Republic

Melt processability highly depends on the rheological properties of materials, such as shear-  $(\eta_s)$  and extensional  $(\eta_e)$  viscosity [1].

Using a high-pressure capillary rheometer and employing a long die and a zero-length (orifice) die, crucial data can be gathered. Applying the Cogswell method [2], measuring the entrance pressure drop ( $P_{en}$ ), the  $\eta_e$  can be investigated. However, there is the possibility that material leaving the orifice die sticks to the wall [3,4] leading to overestimated ( $P_{en}$ ) and incorrect ( $\eta_e$ ).

During processing, typically the uniaxial and planar ( $\eta_e$ ) plays a critical role. Generation of pure planar flow is complicated and is considered unsuitable for routine use [5].

Based on the pioneering work of Zatloukal et al. [4,6], current designs of ROSAND dies have been optimized to create a new series of FreeFlow circular and rectangular orifice dies. These dies prevent the possibility of overestimating the ( $P_{en}$ ), allowing precise determination of uniaxial and planar ( $\eta_e$ ). In addition, FreeFlow dies enables a new way of evaluating extrudate swell, which is frequently missed or overestimated using conventional orifice dies.

Measurements using the ROSAND high-pressure capillary rheometer can now provide in-depth studies of extrudate swell, uniaxial and planar  $(\eta_e)$ .

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[5] Dealy J. M., Read D. J., Larson R. G., Structure and Rheology of Molten Polymers, 2nd Edition, 2018, pp. 443
[6] Zatloukal M., Polymer 104 (2016) 258-267.

# Symposium NF

# Non-Newtonian Fluid Mechanics and Flow Instabilities

Organizers: Marco Ellero, Stylianos Varchanis and Helen Wilson

Thursday 2:00 LT 24 (Level 10) / Track 8

# A Darcy-type law for yield-stress fluid flows through porous media Emad Chaparian

#### Mechanical & Aerospace Engineering, University of Strathclyde, Glasgow G1 1XJ, United Kingdom

Yield-stress fluid flows through porous media are inherent to many industries including filtration, oil & gas and mining and also biomedical applications. The challenge in finding a Darcy-type law for bulk transport properties of yield-stress fluids is twofold: firstly, due to the yield stress of this type of fluids, a finite pressure gradient should be applied to initiate the flow, otherwise, the fluid remains quiescent and the flow rate is zero. Moreover, the flow rate is not a linear function of the imposed pressure gradient in the flowing regime mainly due to the non-linear rheological behaviour of this type of fluids [1]. In this study, we investigate the onset of the flow when yield-stress fluids percolate inside porous media. A mathematical model is presented that can successfully predict the critical pressure gradient which reduces to purely geometrical features of the media [2]. We validate our model with our previously published data [3] where the porous media are mimicked with mono-dispersed circular obstacles [3]. We validate our mathematical model further with more exhaustive simulations, i.e. bi-dispersed cases and also various shapes of obstacles. Moreover, far from the yield limit or the critical pressure gradient, we propose a universal Darcy-type law that can predict the bulk transport properties of yield-stress fluid flows in porous media. Again, exhaustive simulations validate the proposed universal general Darcy law.

[1] E. Chaparian & O. Tammisola, J. Fluid Mech. 911 (2021) A17. [2] E. Chaparian, (2023) arXiv:2308.08913. [3] D. Fraggedakis, E. Chaparian & O. Tammisola, J. Fluid Mech. 911 (2021) A58.

#### Thursday 2:20 LT 24 (Level 10) / Track 8

#### Rising bubble through viscoplastic - Newtonian interface

Marjan Zare<sup>1</sup>, Ian Frigaard<sup>2</sup>, and Gregory Lawrence<sup>3</sup>

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The passage of single air bubbles through the horizontal interface between miscible viscoplastic and Newtonian fluids, considering various combinations of densities and viscosities for the fluid layers, is studied computationally [1]. The primary focus is on the quantity of liquid transferred from the lower layer (Viscoplastic fluid) to the upper layer (Newtonian fluid) as a result of the bubble's ascent, a factor with significant implications for the turbidity of methane-emitting lakes and water bodies. The results show that at Bo>1 and moderate Ar, prolate-shaped bubbles crossing the interface undergo elongation in the direction of their poles. This elongation is further accentuated when the viscosity of the upper layer is less than the plastic viscosity of the lower layer. The bubble is found to break up when leaving the lower layer, of a critical capillary number,  $Ca_c \approx 5$ . The results show a significant reduction in the volume of entrainment compared to the Newtonian counterpart. This suggests disturbances caused by the rising bubble at the interface dissipate over a smaller region. Four distinct entrainment regimes are identified, mainly indicating the height to which the entrained fluid can be transported away from the interface. In contrast to Newtonian fluids, the volume of entrainment increases by decreasing the viscosity of the upper layer. Interestingly, the heavy yield stress fluid that has been dragged up into the light Newtonian fluid does not recede down by time.

[1] M. Zare, I. Frigaard, and G. Lawrence. Bubble-induced entrainment at viscoplastic-Newtonian interfaces. arXiv:2310.17791, (2023) https://doi.org/10.48550/arXiv.2310.17791

#### Thursday 2:40 LT 24 (Level 10) / Track 8

#### Squeeze cementing: managing uncertainty in viscosplastic Hele-Shaw flows

Ian Frigaard<sup>1</sup>, Mahdi Izadi<sup>1</sup>, and Elizabeth Trudel<sup>2</sup>

<sup>1</sup>University Of British Columbia, Vancouver, Bc V6T 1Z4, Canada; <sup>2</sup>University Of British Columbia, Kelowna, Bc V1V 1V7, Canada

Approximately 40% of Methane emissions in Canada come from fugitive emissions in the oil and gas sector. This constitutes a variety of source, one of which is leakage from wells. Repair of such leakage involves a process called squeeze cementing in which a cement slurry (typically of microfine particles) is pumped under pressure through holes (perforations) in the side of the well, forced into the microannular gaps that are believed to be the main pathways for gas leakage to surface. From the rheological perspective, this constitutes a filling/displacement flow of a yiscoplastic suspension into a channel. The complexity comes from the extreme uncertainty of the channel dimensions, which are known to vary

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along the well. Here we present our first attempts both at modelling the invasion/sealing process and at handling the uncertainty of the leakage pathways.

<sup>1</sup>Mechanical, Industrial and Aerospace Engineering, Concordia University, Montreal, Quebec, Canada; <sup>2</sup>Mechanical, Industrial

Viscoplastic fluids (VPFs) are non-Newtonian fluids that exhibit solid-like behavior below a critical shear stress threshold, termed yield stress ( $\tau_y$ ), transitioning to fluid-like behavior beyond this threshold. Mixing VPFs is pivotal in diverse industrial applications, including food processing, pharmaceuticals, and waste management. Mixing of VPF is particularly challenging because of their typically high viscosity and the solid-like behaviour associated with the yield stress. More specifically, the rapid variation of viscosity with the strain rate is known to lead to the localization

We consider a simple stirring strategy in a circular flow domain to explore the influence of the yield stress on flow development and mixing dynamics. Mixing is promoted by moving a circular stirrer, with a constant speed, on a circular path. The diameter of the mixing vessel is sufficiently large to ensure wall effects are negligible. Half of the fluid filling the tank is marked with a passive dye. Simulations are conducted using a modified solver in OpenFOAM. We highlight the impact of yield stress, represented by the Bingham number (Bn), on fluid flow evolution. By comparing the mixing of a Newtonian fluid with VPFs of different Bn values, we identify distinct stages in flow development. The influence of Bn on these stages is explained through changes in vortical structures. In contrast to Newtonian fluids, where vortices move unboundadly, the presence of yield stress constrains the advection, thus significantly altering the mixing dynamics. We measure the radius of the well-mixed region

# Thursday 3:00 LT 24 (Level 10) / Track 8

Mohammad Reza Daneshvar Garmroodi<sup>1</sup> and Ida Karimfazli<sup>2</sup>

3:50 LT 24 (Level 10) / Track 8

and Aerospace Engineering, Concordia University, Montreal, Ouebec, Canada

Scalar mixing in viscoplastic fluids

James Shemilt<sup>1</sup>, Alex Horsley<sup>2</sup>, Oliver Jensen<sup>1</sup>, Alice Thompson<sup>1</sup>, and Carl Whitfield<sup>1</sup> <sup>1</sup>Department of Mathematics, University of Manchester, Manchester, United Kingdom; <sup>2</sup>Division of Respiratory Medicine,

Interplay of surfactant and viscoplasticity in the Rayleigh-Plateau instability of a film coating a tube

at different Bn values and show that as Bn increases, the radius of the well-mixed region decreases proportional to 1/Bn.

of the flow and mixing. Overcoming these challenges requires detailed understanding on the flow development in the VPFs.

Department of Mathematics, University of Manchester, Manchester, United Kingdom; "Division of Respiratory Medicin University of Manchester, Manchester, United Kingdom

Airway closure in the lungs can be caused by a capillary instability of the mucus layer that lines the airway wall. To investigate the combined effects of surfactant and mucus yield stress in a simple airway closure model, we study the Rayleigh-Plateau instability of a film of Bingham liquid coating the interior of a rigid tube with insoluble surfactant at the air-liquid interface. We use thin-film and long-wave approximations to derive equations describing the evolution of the liquid layer. Using numerical solutions and asymptotic analyses, we quantify how surfactant amplifies the stabilising effects of the yield stress. For thin films, increasing the surfactant strength increases the size of perturbation to the free surface required to trigger instability, slows growth of the instability and reduces the total amount of deformation caused to the layer. The dynamics of a thin film with very strong surfactant coincide with those of a surfactant-free film but with time slowed by a factor of four and the capillary Bingham number, which measures yield stress relative to capillary stresses, exactly doubled. By solving the long-wave equations numerically, we detail how the critical layer thickness required for a liquid plug to form in the tube is increased by adding strong surfactant when the capillary Bingham number is sufficiently large. This contrasts with the case of a Newtonian layer, in which adding surfactant only acts to delay plug formation. We discuss the implications of our results for understanding the impact of surfactant deficiency and increased mucus yield stress in obstructive lung diseases such as cystic fibrosis.

Thursday 4:10 LT 24 (Level 10) / Track 8

# The effect of fluid elasticity and plasticity on the flow within wavy channels

Mohamed S. Abdelgawad<sup>1</sup>, Simon J. Haward<sup>2</sup>, Amy Shen<sup>2</sup>, and Marco E. Rosti<sup>1</sup>

<sup>1</sup>Complex Fluids and Flows Unit, Okinawa Institute of Science and Technology, Onna son, Okinawa 904-0495, Japan; <sup>2</sup>Micro Bio Nanofluidics Unit, Okinawa Institute of Science and Technology Graduate Univers, Okinawa, Japan

Elastoviscoplastic (EVP) fluids are a distinct category of materials characterized by their ability to demonstrate both solid-like and liquid-like behaviors in response to applied stress. This dual nature makes them particularly relevant in industrial processes involving complex geometries, such as porous media and wavy channels. We use numerical simulations, validated by microfluidic experiments, to explore the influence of wavy channel design (e.g., amplitude and wavelength) and flow characteristics on the development of yielded and unyielded regions, flow regimes (steady or unsteady), and pressure drop in wavy channels. Our findings demonstrate that the plasticity of EVP fluids plays a critical role in such geometries, promoting a transition from steady to time-dependent behavior when compared to viscoelastic fluid under the same flow condition. This plastic-induced instability is attributed to the formation of the unyielded regions within the flow, which perturb the flow field and, in turn, affect the stress distribution. A phase diagram is introduced, outlining the emergence of this instability in relation to the Bingham number (a measure of plasticity) and the Weissenberg number (a measure of elasticity).

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# Thursday 4:30 LT 24 (Level 10) / Track 8 Interplay between complex fluid rheology and wall compliance affects the hydrodynamic resistance of deformable configurations

#### Evgeniy Boyko

#### Faculty of Mechanical Engineering, Technion - Israel Institute of Technology, Haifa, Israel

Viscous flows through configurations manufactured from soft materials apply pressure and shear stress at the solid-liquid interface, leading to deformation. The resulting fluid-structure interaction affects the relationship between the pressure drop and the flow rate or the hydrodynamic resistance, which is the ratio between the two. While the hydrodynamic resistance in deformable configurations has been extensively studied for Newtonian fluids, it remains largely unexplored for non-Newtonian fluids even at low Reynolds numbers. In this work, we use the Lorentz reciprocal theorem to derive a closed-form expression for the hydrodynamic resistance of complex fluids in deformable axisymmetric channels. For small deformations, our theory applies to a wide class of shear-thinning and viscoelastic constitutive models in the weakly non-Newtonian limit and provides the leading-order effect of the interplay between complex fluid rheology and wall compliance on the hydrodynamic resistance, bypassing the detailed calculations of the non-Newtonian fluid-structure-interaction problem. We illustrate our approach for a viscoelastic Oldroyd-B fluid and a shear-thinning Carreau fluid, highlighting the simplicity and elegance of applying the reciprocal theorem to obtain integrated quantities in multi-physics problems.

#### Thursday 4:50 LT 24 (Level 10) / Track 8

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# Complex fluid flows in grooved superhydrophobic channels

Hossein Rahmani and Seyed Mohammad Taghavi

Department of Chemical Engineering, Laval University, Quebec, QC GIV 0A6, Canada

The Poiseuille flow of a viscoplastic Bingham material in channels with a groovy superhydrophobic (SH) wall is studied via numerical simulations. The grooves of the superhydrophobic wall can show a different orientation with respect to the applied pressure gradient, i.e. making an angel ? from 0 to 90°. In the limit of thin channels, the groove period is much larger than the half-channel height. Assuming trapped air pockets inside the grooves and flat interfaces between the Bingham flow and the air pockets, we implement the Navier slip law to model the dynamics of the formed liquid/air interface. The Bingham plastic rheology is modeled via implementation of the Papanastasiou regularization model. For ?=90° (transverse grooves), the inertial effects are also taken into consideration. Therefore, considering wide value ranges for the flow parameters, i.e. Reynolds number (R), Bingham number (B), slip number (b), groove periodicity length (l), slip area fraction (f) and the groove orientation angle (?), the main flow features are comprehensively studied; such features include the slip and axial velocity profiles, the effective slip length, frictions factor, flow mixing index, as well as the unyielded plug dynamics. We find a decrease in the slip velocity with an increase in ? and R. For 0<?<90°, a secondary flow is generated that enhances the flow mixing. In addition, we explore the effective slip length tensor for the Bingham material over the SH surface, finding asymmetric shear component profiles with respect to ?=45° due to the yield stress effects. An increase in R also causes an asymmetry in the velocity profiles, while hindering the growth of SH wall-induced perturbations, leading to a non-monotonic effect on the center plug yielding and breakage. We also find a non-monotonic effect of ? on the center plug yielding, which is associated to the simultaneous effects of a decrease in the longitudinal component (W) and an increase in the vertical component (V) of the velocity field as ? increases.

#### Thursday 5:10 LT 24 (Level 10) / Track 8

#### Flow of shear-banding viscoelastic fluids over long cavities

<u>Fabian Hillebrand</u><sup>1</sup>, Stylianos Varchanis<sup>1</sup>, Stylianos Varchanis<sup>2</sup>, Cameron Hopkins<sup>1</sup>, Simon J. Haward<sup>1</sup>, and Amy Shen<sup>1</sup> <sup>1</sup>Micro Bio Nanofluidics Unit, Okinawa Institute of Science and Technology Graduate Univers, Okinawa, Japan; <sup>2</sup>Center for Computational Biology, Flatiron Institute, New York, NY 10010, United States

We perform a detailed numerical investigation of the flow of a shear-banding, viscoelastic wormlike micellar (WLM) solution over cavities with various depths (D) and lengths (L). The fluid is modelled using the diffusive Giesekus model, with parameters quantitatively describing the rheology of a 100:60 mM cetylpyridinium chloride : sodium salicylate aqueous WLM solution used for experimental validation. Both experimental and numerical velocity fields show the formation of lip vortices upstream of the cavity entrance corner after a first critical Weissenberg number (Wi) and an onset of time-dependence after a second critical Wi. Furthermore, we observe a transition from "cavity flow" to "one-sided expansion-contraction flow" as L is increased. This transition is manifested by a change of vortical structures from large-scale recirculations within the cavity to separated vortices in the salient corners of the expansion-contraction. We construct state diagrams in Wi-L space for various cavity depths D, characterising this transition and describing the transformation of the vortical structures with fluid elasticity.

# **Friday Morning**

# Symposium EF

# **Emulsions, Foams and Interfacial Rheology**

# Organizers: Simon Cox, Thibaut Divoux and Nick Jaensson

#### Friday 8:50 LT 1 (Level 7) / Track 1 Flowing foams and varicose veins Simon Cox

# Department of Mathematics, Aberystwyth University, Aberystwyth, United Kingdom

In varicose vein **sclerotherapy**, a liquid foam containing sclerosant is injected into the vein to displace blood and to trigger vein collapse. The choice of foam parameters such as liquid fraction and bubble size are critical in ensuring that the blood is displaced and that the foam then disperses quickly.

Foams are used in the process because of their **yield stress**. We model foams as Bingham fluids to predict optimal parameter values for sclerotherapy. To allow investigation of more realistic vein geometries, such as uniformly curved and sinusoidal channel, we use two-dimensional **FEM calculations**.

To extend this approach we would like to have **bubble-scale information**. For example, does the discrete nature of the bubbles in a foam suppress or enhance dead zones, affecting the success of the displacement process? A two-dimensional **kinetic model**, for foams of arbitrary liquid fraction, allows us to predict bubble motion, bubble shape, and local stresses in channels and past obstacles.

This is joint work with Tirion Roberts (Bangor) and Denny Vitasari (Surakarta).

### Friday 9:10 LT 1 (Level 7) / Track 1

# Understanding the foamability and mechanical properties of foamed polypropylene blends by shear and extensional rheology

Ester Laguna-Gutierrez<sup>1</sup>, Miguel Rodriguez-Perez<sup>1</sup>, and <u>Paula Moldenaers</u><sup>2</sup> <sup>1</sup>University of Valladolid, Valladolid, Spain; <sup>2</sup>KU Leuven, Leuven, Belgium

In this work the influence of the rheological behaviour of miscible blends of a linear and a high melt strength, branched, polypropylene (HMS PP), on the cellular structure and mechanical properties of cellular materials fabricated with these polypropylenes (PPs) has been investigated. The rheological properties of the polypropylene melts were investigated in steady as well as oscillatory shear flow and in uniaxial elongation in order to determine the strain hardening coefficient. While the PP blends display very similar behaviours in shear, there are substantial differences in the elongational behaviour. Cellular materials with a fixed relative density of 0.2 have been fabricated by Improved Compression Moulding (ICM), a technique that allows controlling the foam density by mechanical means. The open cell content has been quantified and a detailed characterization of the cellular structure has been performed. In addition, mechanical properties in compression have been measured. While the linear PP does not exhibit strain hardening, the blends of the linear and the HMS PP show pronounced strain hardening, increasing with the concentration of HMS PP. A relation has been observed between the strain hardening coefficient and the open cell content: more strain hardening leads to a lower open cell content. Related with cellular structure, in general, the cell size and the width of the cell size distribution, increase with the amount of linear PP in the blends. These results indicate that the cell walls are less stable when large amounts of linear PP are used. Also mechanical properties are conditioned by the extensional rheological behaviour of PP blends. Cellular materials with the best mechanical properties are those that have been fabricated using large amounts of HMS PP. The results demonstrate the importance of the extensional rheological behaviour of the polymers for a better understanding and steering of the cellular structure and properties of the cellular materials.

# Friday 9:30 LT 1 (Level 7) / Track 1

#### Elastoviscoplastic interfaces and their relevance for the stability of multiphase materials

#### Alexandra Alicke<sup>1</sup> and Jan Vermant<sup>2</sup>

<sup>1</sup>Eindhoven University of Technology, Eindhoven, The Netherlands; <sup>2</sup>Materials Department, ETH Zürich, Zürich, Switzerland

Particle-laden interfaces have been extensively used due to their superb capabilities of imparting stability in multiphase materials in what is called Pickering-Ramsden stability. Typically, they are used to fully cover the surface of bubbles and droplets, although it has been shown that the existence of an interfacial yield stress at sub-monolayer coverages suffices to resist bubble dissolution (and the main driving force for Ostwaldripening). Analogously to bulk materials, the elastoviscoplastic nature can be achieved by different material design routes. Here, we discuss which handles are available to tune these interactions, and consequently the microstructure and rheological behavior of these interfaces. Moreover, we

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**EF14** 

combine a suite of different experimental techniques to relate the measured interfacial rheological properties with microscale stability in microfluidic experiments that mimic the main driving forces for coarsening mechanisms.

### Friday 9:50 LT 1 (Level 7) / Track 1

Design and production of ultrastable micrometer-sized foams

Mariana Rodríguez-Hakim<sup>1</sup>, Luka Oblak<sup>2</sup>, and Jan Vermant<sup>3</sup>

<sup>1</sup>Departamento de Física Fundamental, UNED, Madrid, Spain; <sup>2</sup>SIKA, Zurich, Switzerland; <sup>3</sup>Materials Department, ETH Zürich, Zürich, Switzerland

Stable foams that can resist disproportionation for extended periods of time are important in a wide range of technological and consumer materials. Environmentally damaging surfactants have often been used to stabilize these types of multiphase materials, since traditional thermodynamicsbased processing routes are not efficient enough for creating eco-friendly (particle stabilized) Pickering-Ramsden foams. This work describes an innovative foaming technology that can produce ultrastable nanosilica-stabilized foams, with bubbles of micrometer-sized dimensions. A nanosilica suspension is foamed at subatmospheric pressures, allowing for adsorption of the particles onto large bubbles. This is followed by an increase back to atmospheric pressure, which induces bubble shrinkage and compresses the adsorbed particle interface, forming a strong elastoplastic network that provides mechanical resistance against disproportionation. We quantify the foam's interfacial mechanical properties to predict the range of processing conditions needed to produce permanently stable foams, and a general stability criterion is derived by considering the interfacial rheological properties under slow, unidirectional compression. The applicability of our stability criterion can be extended to other types of rheologically complex interfaces. Foams that are stable against disproportionation are characterized by interfaces whose mechanical resistance to compressive deformations can withstand their tendency to minimize the interfacial stress by reducing their surface area. Our ultrastable nanosilica foams are tested in real-life applications by introducing them into concrete. In comparison to other commercial air entrainers, our foam improves concrete's freeze-thaw resistance while supplying higher material strength, providing an economically attractive, industrially scalable, and durable alternative for use in real-life applications involving cementitious materials.

# Friday 10:10 LT 1 (Level 7) / Track 1 RHEOLOGY OF A PARTICLE-LADEN SOAP FILM

Jonathan Lalieu, Antoine Seguin, and Georges Gauthier

#### FAST, Universite Paris Saclay, Orsay, France

Particle laden interfaces are ubiquitous in industry (e.g. oil recovery, filtration processes, armored drops used as microreactors, or concrete foams) for which interface stabilization effect have to be controlled. Recent studies [1,2] have shown that mechanical properties of particle-laden films differ from those of single particle laden interface, though the origin of the differences remains unclear. We study experimentally the rheology of a macroscopic particle-laden soap film constituted of polystyrene spheres trapped in a single film made of a tetradecyl trimethyl ammonium bromide (TTAB) and glycerol aqueous mixture of the same density as the particles. The particles are larger than the typical film thickness, they cross both interfaces and capillarity gives rise to attractive interactions between the particles. To study the strain-stress relation of such particle laden film, it is stretched in an annular rheometer cell and sheared at imposed velocity, measuring the stress. The strain-stress results are compared with local shear rate measured through image correlation. We show that, at dense particle volume fraction, the granular film exhibits a complex visco-plastic behavior which is influenced by interfacial parameters such as surface tension and surface viscosity of the carrying fluid. To account for the non-local rheology of the particle laden soap film, we confront the particle velocity fields to kinetic theory extended to dense granular media [3].

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#### Friday 11:00 LT 1 (Level 7) / Track 1

#### Effects of Surface-Active Agents on the Interface Dynamics of an Oil Drop in Water Dimitrios Papavassiliou, Sepideh Razavi, and Thao X. Nguyen

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The oil-water interfacial dynamics can be affected by the presence of surfactant molecules or nanoparticles. In this work, Dissipative Particle Dynamics (DPD) computations are employed [1-2] to investigate the effect of nanoparticle wettability on the stability of the interface between two immiscible fluids, dodecane (oil) and water. The nanoparticles are spherical, and are either homogenous (hydrophobic or hydrophilic), or they are anisotropic Janus particles with amphiphilic behavior. The level of amphiphilicity is controlled during the computations so that the range is from zero degrees to 490 for Janus particles. The surfactant is octaethylene glycol monododecyl ether, which is often used in oil recovery applications. The computations indicate that an oil droplet attached to a channel wall can migrate the farthest with flow when only surfactants are present at its interface. The drop migrates the least when only particles are present, irrespective of their wetting characteristics The re-arrangement of the surfactant molecules and the nanoparticles at the interface, generating weak Marangoni stresses, plays a role in the motion of the drop. For a drop that extends across the channel, a meniscus is formed. Under flow, the meniscus becomes unstable, leading to fingering phenomena, and finally to the detachment of the oil drop from the channel wall. The appearance of fingers depends on the presence of particles or surfactants, with the particles leading to longer fingers at the same fluid velocity. There appears to be a critical contact angle for the detachment of the oil drop from the channel wall. The appearance of fingers depends on the presence of particles or surfactants, with the particles leading to longer fingers at the same fluid velocity. There appears to be a critical contact angle for the detachment of the oil drop from the channel walls, the particles are present or not. When the oil drop detaches fully for the channel walls, the particles and the surfactant molecules reorient on the interface modi

**EF17** 

**EF18** 

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#### Friday 11:20 LT 1 (Level 7) / Track 1

Pore-scale visualization of emulsion flow in porous media

Clarice de Amorim, Amanda C. Pessoa, Ranena V. Ponce, and <u>Marcio S. Carvalho</u> Department of Mechanical Engineering, PUC-Rio, Rio de Janeiro, RJ 22451900, Brazil

Pore-blocking efficiency is a key factor when dilute and stable emulsions are used as flow diversion agent in oil recovery, remediation of contaminated sites, gas storage and acidification processes. Emulsion plugging occurs when droplets of the dispersed phase are trapped in the pore throats as they flow through porous media. The droplet capture phenomenon is highly dependent on the drop-to-pore size ratio, the dispersed phase concentration, and capillary number. Thus, understanding the transport of droplets and the physical mechanisms of pore-blocking at the micro-scale is fundamental for a proper design of emulsion flooding for different applications. Oil-in-water emulsion systems with two distinct drop size distributions were formulated to evaluate mobility change as a function of drop-to-pore throat diameter ratio and capillary number. The tests were performed by recording the injection pressure response during the sequential flow of an aqueous phase, followed by emulsion (oil drops dispersed in the aqueous phase) and then by a second slug of aqueous phase. The association of microfluidic devices and imaging techniques provided a robust methodology, combining accurate pressure measurements and pore-scale visualization of the droplets' capture phenomenon during emulsion flooding. Detailed visualization of the flow was achieved by high-speed image acquisition at different stages of the test to identify the droplets' capture mechanisms and the capture/release dynamic of the droplets in the pore throats. At low capillary numbers, a larger number of drops were captured in the pore throats because of the stronger capillary forces. As the capillary number was increased above a threshold value, the viscous force was large enough to overcome the capillary resistance and the drops were able to deform and re-enter the flow stream. This behavior was quantitatively demonstrated by comparing the mobility of the emulsion to that of the aqueous phase.

#### Friday 11:40 LT 1 (Level 7) / Track 1

### Stabilization of O/W emulsions with sodium alginate and Pluronic

Pietro Renato Avallone, Nadia Russo, Rossana Pasquino, and Nino Grizzuti

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Emulsions are heterogeneous systems composed of at least two immiscible phases, where droplets of one liquid are dispersed into the other. They are thermodynamically unstable, and they need a third component, typically an emulsifying agent or stabilizer. This work seeks to explore the effects of varying concentrations of Pluronic, on one side, and sodium alginate (Alg), on the other side, on the properties of oil-in-water emulsions. Pluronics are used as amphiphilic molecules to stabilize the oil droplet interface, the Alg is instead used to increase the viscosity of the suspending medium in a way to slow down the droplet coalescence. Although the purpose is the same, i.e. increasing the stability of the o/w emulsions, the approach in the two cases is, of course, different. Rheology and microphotography were employed to assess the characteristics of the resulting emulsions. Ultrasound and high shear mixing have been exploited for reducing the size of the droplets and increase the emulsions stability in the Alg case. Ultrasound induces biopolymer degradation, making its application on emulsions counterproductive. High shear mixing, instead, decreases the droplet sizes and narrows its distribution as the Alg concentration increased. This led to a substantial improvement in emulsion stability, demonstrating a concentration-dependent effect and achieving stability up to 9 days at a concentration of 10 wt%. The results highlight the potential of Alg as an environmentally friendly stabilizer for oil-in-water emulsions. Concerning with Pluronic, the use of L64 as an alternative amphiphilic polymer to stabilize the o/w interface has been exploited. The concentration is changed in the range 0.1-10 wt%, exploring both the concentration range in which pluronic molecules are only at the o/w interface, and the range in which pluronic self-assembly needs to be considered. Stability depends of course on the concentration regime, but phase separation occurs in any case within 24 hours.

#### Friday 12:00 LT 1 (Level 7) / Track 1

#### Interfacial rheology effects in crude oil production

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Crude oil extraction involves retrieving oil from beneath the ocean's seabed. Within this process lies the concept of flow assurance, which pertains to engineering practices aimed at ensuring the uninterrupted flow of reservoir fluids from their origin in the reservoir to the refining stage. The primary issues linked to halting line production are the formation of hydrates, emulsions, waxes, and asphaltenes. Predicting the occurrence of each of these poses significant challenges, despite numerous numerical and experimental studies. These problems are primarily studied separately, often disregarding the real-time presence of all elements, which is closer to reality in this case. Few publications primarily focused on bulk analysis conducted under low-temperature and high-pressure conditions, mirroring those found in the reservoir. In this study, we aim to comprehensively analyze the interfaces within this system by employing various techniques simultaneously. We delve into the interfaces of water in waxy crude oil emulsions and synthetic hydrates (using cyclopentane), comparing different crude oils. Moreover, we elucidate how these findings correspond to the distinctive chemical characteristics of each oil. Our investigation involves measuring shear and dilatation oscillatory modes using a rotational rheometer and a pendant drop tensiometer. We focus on examining several crucial parameters, including how cooling rates impact wax formation, the inhibitory effects of salt presence on hydrate formation, and the role of cyclopentane in both hydrate formation and inhibiting wax buildup.

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# Friday 12:20 LT 1 (Level 7) / Track 1

Exploring the mechanisms of emulsion separation via millimetric droplet coalescence

Andrei Zlobin<sup>1</sup>, Enric Santanach-Carreras<sup>1</sup>, Laurence Talini<sup>2</sup>, François Lequeux<sup>3</sup>, and Pascal Panizza<sup>4</sup>

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During petroleum production, emulsification phenomena occur and the metastable phase of W/O emulsion (*a dense-packed zone*) forms due to the presence of the surfactant species in the oil. Recent studies [1] have described the separation kinetics of such an emulsion with the micrometric droplet size distribution (below 50  $\mu$ m) at the bottle-test conditions. However, the real separator unit is a dynamic system with inlet and outlet flows where the droplet size distribution varies from micrometres (at the separator inlet) to millimetres (at the water-DPZ interface).

In this research, we use a simple millifluidics setup to generate monodisperse droplets from 0.2 mm to 2 mm one-by-one and stock them in the recipient. The experiment has two stages:

the emulsion accumulation while the droplet generation and coalescence are concurrent,

the emulsion separation with no more droplets arriving into the DPZ.

Emulsion interfaces are traced in time with spatiotemporal images to obtain the data on the separation speed.

The first significant difference with [1] is the linear separation rate instead of the formation of the metastable phase for the same system ("water in dodecane + 800 ppm Span 80"). This can be explained with a complete droplet deformation depending on the emulsion height and the droplet size. Another major difference is the coalescence only at the water-emulsion interface (contrary to the model [1]). Visually the droplets from below have a polyhedral "foam-like" shape, while ones on top are spherical with a thick oil layer around them.

The presented results and perspective of millifluidics application in the emulsion generation will be useful in the study of the different factors of coalescence.

#### References

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#### Friday 12:40 LT 1 (Level 7) / Track 1

#### Air-water interface properties and foam stabilization by mildly extractedlentil protein

Penghui Shen<sup>1</sup>, Jinfeng Peng<sup>2</sup>, Leonard Sagis<sup>1</sup>, and Jasper Landman<sup>1</sup>

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Lentil proteins have shown potential as substitutes for animal proteins in foam-based food products, although the mechanisms by which the proteins stabilize the air-water interface are largely unexplored. Here, we systematically studied the air-water interfacial behavior (including adsorption kinetics, and interfacial dilatational and shear rheology), interfacial structures and foaming properties of mildly extracted lentil protein isolate (LPI). We compared these properties of LPI with two animal-based proteins commonly used in food industry, i.e., dairy-derived whey protein isolate (WPI) and egg-derived egg white protein isolate (EPI). The mildly extracted LPI had high protein content (85.2%), high protein solubility (91%) and high nativity, with a globulin-to-albumin ratio of 81:19. LPI adsorbed fast at the air-water interface, showing a foam overrun of 285% that is comparable with WPI and EPI. Although LPI formed interfaces with lower stiffness than EPI, it showed comparable foam half-life time (115 min) with EPI, due to the higher thickness of the interface formed by LPI, which is dominated by lentil globular protein particles with a size around 11 nm. The lentil globular proteins appear to have weaker in-plane interactions than smaller WPI molecules at the air-water interface, resulting in a pronouncedly lower interfacial stiffness and foam half-life time of LPI compared to WPI (295 min). However, the LPI-formed interface is more stretchable than the WPI-formed interface and shows less disruption in both large dilatational and shear deformations. Thus, LPI might have comparable or better performance than WPI in stabilizing foam under processing conditions. Overall, lentil proteins have good foaming properties and could be applied as an animal-based protein replacer in producing foam-based food products.

# Symposium PF

# **Polymeric Fluids**

Organizers: Francesco Del Giudice, Richard Graham and Evelyne van Ruymbeke

#### Friday 8:50 LT 17 (Level 7) / Track 2

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**Mesoscopic simulations of the linear viscoelasticity of semidilute unentangled wormlike micellar solutions** Avishek Kumar<sup>1</sup>, P Sunthar<sup>2</sup>, and <u>J. Ravi Prakash<sup>3</sup></u>

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We investigate the structure and dynamics of unentangled wormlike micellar solutions (WLMs) in the semidilute concentration regime by mesoscopic simulations. These solutions exhibit remarkable properties since wormlike micelles constantly break and reform. While they are viscoelastic like polymer solutions, their 'living' character leads to a rich diversity of behaviour which is currently being exploited in a wide range of industrial applications. However, the connection between the microstructural transitions and the rheological properties of these solutions is still poorly understood. We present simulation predictions of equilibrium properties and the linear viscosity of WLM solutions across a range of concentrations ranging from the dilute to the semidilute regime as a function of model parameters. WLM dynamics have been simulated using Brownian dynamics coupled with the reversible scission/association of so-called "persistent worms". A persistent worm in the model represents the smallest possible length of a wormlike micelle and is represented by a bead-spring chain with sticky beads at the ends. The persistence length of a WLM is captured with a bending potential between the springs internal to a persistent worm and between springs across persistent worms that are stuck to each other. Hydrodynamic interactions have been included in the model for the first-time using Jim Swans' algorithm for the efficient computation of HI in colloids within the framework of HOOMD-blue. This unique model is capable of predicting the influence of sticker strength and concentration on storage and loss moduli and enables the distinction between the Rouse and Zimm dynamics of WLM solutions.

#### Friday 9:10 LT 17 (Level 7) / Track 2

#### Exploring drag reduction for polymers with varying molecular weight distribution

Lukas Brandfellner<sup>1</sup>, Lukas Brandfellner<sup>2</sup>, Alexander Bismarck<sup>1</sup>, Alexander Bismarck<sup>2</sup>, and Hans Werner Müller<sup>2</sup> <sup>1</sup>Doctoral College Advanced Functional Materials, University of Vienna, Vienna, Austria; <sup>2</sup>Institute of Materials Chemistry & Research, University of Vienna, Vienna 1090, Austria

Turbulent flow of liquids is strongly influenced by addition of small amounts of high molecular weight polymers (>10<sup>6</sup> g/mol). The dissolved polymers change the flow profile of turbulent pipe flow and friction is decreased. This effect known as polymeric drag reduction (DR) is of great interest for liquid transport applications as well as for improving the understanding of the interplay of molecular dynamics and turbulent flow. With increasing Reynolds number (Re) different DR regimes can be distinguished. At low and high Re DR becomes independent of molecular properties and either the friction factor *f* coincides with *f* of a Newtonian fluid (low Re) or follows a universal maximum drag reduction (MDR) asymptote (high Re). The onset of DR, the approach of the MDR, and the transition regime in between are governed by concentration and molecular weight of the polymer additive. These relations are key parameters for understanding the DR mechanism. Investigations of the influence of the molecular weight are challenged by the polydispersity of the polymer and polymer degradation in turbulent flow. We explored drag reduction of polyacrylamide with different initial molecular weight distributions in a pilot scale flow facility. We monitored drag reduction of degrading polyacrylamide by measuring the pressure drop along a straight pipe and cycling the same solution multiple times. The flow experiments were accompanied by gel permeation chromatography, which provided the development of the molecular weight distribution of the applied polymers. Re scans at selected flow distances allowed us to describe *f* as function of degradation and Re and to extrapolate *f* along these two independent variables. The extrapolation provided DR in the whole transition regime for each flow distance and for each measured molecular weight distribution.

### Friday 9:30 LT 17 (Level 7) / Track 2

#### A possible route to measure meaningful polymer drag reduction in Taylor Couette flow

Hans Werner Müller<sup>1</sup>, Lukas Brandfellner<sup>1</sup>, Lukas Brandfellner<sup>2</sup>, Alexander Bismarck<sup>1</sup>, and Alexander Bismarck<sup>2</sup> <sup>1</sup>Institute of Materials Chemistry & Research, University of Vienna, Vienna 1090, Austria; <sup>2</sup>Doctoral College Advanced Functional Materials, University of Vienna, Vienna, Austria

Drag reduction (DR) by polymers as described for the first time by Toms [1] is utilised in many applications as e.g. pipelines, fracking, and sewage systems. There is still demand for new DR agents showing improved resistance against degradation in turbulent flow or offering switchable activity. Testing new DR agents in a rotational rheometer has numerous advantages over characterisation in pipe flow. The rheometer allows for smaller sample volume, reduced parasitic perturbations, and continuous flow, which cannot be achieved in pipe flows avoiding pumps, which cause extra degradation of the polymers.

For pipe flow it is well known that DR only occurs in turbulent flow, not in the laminar regime. Taylor Couette (TC) flow as it occurs in rotational rheometers shows a large variety of different flow regimes with increasing Reynolds number (Re). The flow regime can be expected to affect the polymer's DR capability. In literature this aspect is mentioned [2] but not thoroughly examined. We worked on a more complete picture for DR

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in TC flow and followed the idea of Eckhardt et al. [3] to define a pseudo Nusselt number, which was used to identify the flow regime and to determine the friction factor [4]. Since aqueous polymer solutions for DR show low viscosities (~1 mPa.s) we characterised them in double gap geometry where a bob between two static cylinders rotates. The inner gap remained in the laminar Couette regime for a large range in Re and we derived a pseudo Nusselt number related to the outer gap flow only. A comparison to measurements in Taylor-Couette geometry was performed. We will present our first results on DR investigated by means of a rheometer with double gap geometry in dependence of the flow regime.

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#### Friday 9:50 LT 17 (Level 7) / Track 2

#### Biopolymer/ionic liquid networks controlled by water

Roshan A.M. Yunus<sup>1</sup>, Marcus Koch<sup>2</sup>, Philippe Dieudonné-George<sup>3</sup>, Domenico Truzzolillo<sup>3</sup>, Ralph H. Colby<sup>4</sup>, and <u>Daniele Parisi<sup>1</sup></u> <sup>1</sup>Chemical Engineering, University of Groningen, Groningen, Groningen 9747 AG, The Netherlands; <sup>2</sup>INM. Leibniz Institute for New Materials, Saarbrücken 66123, Germany; <sup>3</sup>Laboratoire Charles Coulomb L2C, UMR 5221 CNRS Université de Montpellier, Montpellier, France; <sup>4</sup>Materials Science and Engineering, The Pennsylvania State University, State College, PA, United States

Native cellulose is the most abundant biopolymer on Earth. However, the strong intra and interchain hydrogen bonds, make its industrial use very limited, as it is not melt-processable, and it dissolves at the molecular level only in a few solvents. 1-ethyl-3-methylimidazolium acetate is an ionic liquid (IL) that dissolves native cellulose without derivatizing it. It is already known that water can catalyze the sol-gel transition in cellulose/IL solutions. However, the link between polymer concentration, water content and gel strength is still missing, yet pivotal to design gels whose rheology can be tailored via water or environmental humidity. In the present work, the relationship between polymer (Cp) and water (Wc) concentration, and shear modulus (Gp) has been established. The formation of a gel is contingent on the presence of entanglements within the system, and its strength is elegantly regulated by the content of introduced water. As a matter of fact, water can be used as an effective crosslink density (rho). At low water content, entanglements govern the gel elasticity, while at higher water concentrations, the modulus scales as Gp ~ Cp^2\*Wc^(3.0\pm0.2). Note that "traditional" synthetic polymer networks exhibit Gp ~ Cp^2\*rho setting apart native cellulose from simple flexible linear polymer chains. It is conjectured that the stiffening of the chains, and the formation of polymer bundles/aggregates catalyzed by the presence of water is at the origin of this strong dependence of the modulus on Wc. Our work provides new insights on gel networks constituted by complex polymers, such as natural polysaccharides. A rheological toolkit and scaling laws are provided to tailor the rheological properties of biogels at wish. In view of these results, we trust that our work could push forward the knowledge of (bio)polymer gels, where water is used as an "unusual" crosslinking agent.

#### Friday 10:10 LT 17 (Level 7) / Track 2

#### Acid-induced gelation of carboxymethylcellulose solutions

Gauthier Legrand<sup>1</sup>, Guilhem P. Baeza<sup>2</sup>, Matteo Peyla<sup>1</sup>, Lionel Porcar<sup>3</sup>, Carlos Fernandez-de-Alba<sup>2</sup>, Sébastien Manneville<sup>1</sup>, and <u>Thibaut Divoux<sup>1</sup></u>

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The sodium salt of carboxymethylcellulose (NaCMC) is a water-soluble derivative of cellulose, which is broadly used for industrial applications such as food, pharmaceuticals, paints, etc., and serves as a thickener and water retention. The properties of NaCMC can be tuned via the degree of substitution (DS), which corresponds to the average number of carboxymethyl groups per repeating glucose unit and varies between 0 and 3. Highly substituted polymers, i.e., for DS >1, are hydrophilic and disperse easily in water, yielding rheological features typical of polyelectrolyte solutions. In contrast, weakly substituted polymers, i.e., for DS<0.9, contain hydrophobic regions, which favor interchain aggregation and the formation of so-called ``fringed micelles" yielding thixotropic and even gel-like properties at high enough concentrations. In practice, the gelation of NaCMC solution can be induced by lowering the pH, which decreases the charge density along the CMC chain and promotes the formation of multichain aggregates.

This presentation will offer a comprehensive description of the acid-induced gelation of carboxymethylcellulose (CMC). Linear viscoelastic properties measured at various pH and CMC contents allow us to build a sol-gel phase diagram, and show that CMC gels exhibit broad power-law viscoelastic spectra that can be rescaled onto a master curve following a time-composition superposition principle. These results demonstrate the microstructural self-similarity of CMC gels, and inspire a mean-field model based on hydrophobic inter-chain association that accounts for the sol-gel boundary over the entire range of CMC content under study. Neutron scattering experiments further confirm this picture and suggest that CMC gels comprise a fibrous network crosslinked by aggregates. Finally, low-field NMR measurements offer an original signature of acid-induced gelation from the solvent perspective. Altogether, these results open avenues for precise manipulation and control of CMC-based hydrogels.

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PF30

### Friday 11:00 LT 17 (Level 7) / Track 2

Rheological Behavior of Amin-Functionalized Liquid Polybutadiene and Butadiene-Styrene Copolymer

Amir Malmir, Saeed Ataie, Sabrina S. Scott, Laurel L. Schafer, and Savvas G. Hatzikiriakos

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The rheological characteristics of amine-functionalized liquid polybutadiene and random copolymer of butadiene-styrene is investigated to understand the influence of intermolecular association on the dynamics of these polymers. Through the hydroaminoalkylation method (Scott, J. Am. Chem. Soc. 2023, 145, 42, 22871-22877), varying quantities of hydrogen-bonding stickers (amines) are introduced to each chain in the unentangled high vinyl content polybutadiene and butadiene-styrene copolymer. This process significantly affects both the glassy and rubbery dynamics, as well as the stretch-ability of these polymers. The amination process results in a delay within the transition region from the glassy to rubbery state, attributed to the lower mobility of stickers in comparison to the backbone segments. As the density of stickers increases, a sol-togel transition is observed. This transition is accompanied with the emergence of a plateau rubbery modulus owing to formation of hydrogen bonding between amine functional groups. Moreover, the time scale of hydrogen bonding dissociation significantly increases with a higher degree of gelation, indicating strong intermolecular associations. Aminated copolymers exhibit improved stretchability compared to aminated polybutadiene, with an optimized density of stickers contributing to this improved property.

# Friday 11:20 LT 17 (Level 7) / Track 2

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# Viscoelastic properties of metallo-supramolecular networks: relationship between stickers dynamics and terminal relaxation

<u>Pierrot de Wergifosse</u>, Maxime Dalne, Charles-André Fustin, and Evelyne van Ruymbeke IMCN, BSMA, Université catholique de Louvain, Louvain-la-Neuve, Belgium

Metallo-supramolecular networks, among other dynamic networks, are particularly interesting due to the wide range of binding energies possibly exhibited by metal-ligand coordination complexes. Depending on the amount or the nature of metal ions, these networks can either be labile or stable. Our work aims to relate the composition of various metallo-supramolecular networks to their viscoelastic properties.

We investigate the viscoelastic properties of poly(n-butyl acrylate) (PnBA) supramolecular networks based on bis-complexes formed by terpyridine ligands and bivalent metal ions, in the melt state. These networks are composed of telechelic stars[1,2] with various molar masses (entangled or not). The metal ions are of different nature (Zn(II) and Cu(II) ions, blended or not) and density (stoichiometric amount or excess, stickers or entanglements dominated).

We systematically compare the terminal relaxation and the stickers lifetime for the different networks. We also compare the activation energies obtained from these two characteristic lifetimes, evidencing that the one obtained from the stickers lifetime should be "universal" for a specific type of coordination complex, no matter the molar mass of the building blocks and ions density. These results are rationalized by including the stickers dynamics into appropriate Rouse model or tube based models (Time Marching Algorithm).

Then, the non-linear response of the supramolecular networks under shear deformation is inves- tigated. In particular, the shear hardening mechanism induced by the presence of metal-ligand bonds is highlighted. By comparing the non-linear response of networks based on different building blocks (some networks exhibiting similar linear viscoelastic responses), we investigate the influence of the shear flow on the lifetime of the supramolecular junctions.

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[2] Pyromali, et al., Journal of Rheology, 66, 1223 (2022)

#### Friday 11:40 LT 17 (Level 7) / Track 2

#### Highly stretchable room-temperature self-healing Vitrimers

Jiaxin Zhao<sup>1</sup>, Richard Mandle<sup>2</sup>, Nicholas Warren<sup>3</sup>, Peter Hine<sup>4</sup>, Andrew Wilson<sup>5</sup>, and Johan Mattsson<sup>6</sup>

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Vitrimers are a relatively new class of covalent adaptable networks characterised by cross-links that are based on associative dynamic exchange reactions1. Vitrimers combine advantages from both thermosets and thermoplastics, yet avoid some of their problems. By introduction of reversible cross-links with tunable association kinetics, they combine mechanical rigidity and chemical resistance with malleability. Here, we present a new vitrimer system based on poly(methyl acrylate) (PMA). Polyacrylates such as PMA and poly(butyl acrylate) are viscous fluids at room temperature and are often used in coatings and adhesives, as liquid crystal elastomer backbone components, or as components in block copolymers to improve material flexibility2, but are rarely used on their own as commercial materials3. The PMA-based vitrimers in this work contain dynamic cross-links at nominal concentrations ranging from 1 to 8 mol%. We characterised their rheology using tensile testing (stress vs strain), small amplitude oscillatory shear rheology (SAOS), and dynamic mechanical analysis (DMA). We also determined the structure using small and wide angle X-ray scattering (SAXS/WAXS) and the relaxation dynamics using broadband dielectric spectroscopy (BDS) and differential scanning calorimetry (DSC). Our vitrimers demonstrate self-healing properties and a very significant degree of uniaxial stretchability ~8000%

(10 mm/min) at room temperature. They have significant toughness (40 MJ/m3) and a mechanical loss factor higher than 2, which suggest that these materials might be interesting as shock absorbers. Moreover, the vitrimers can be re-processed at high temperatures (120 °C).

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#### 12:00 LT 17 (Level 7) / Track 2 Friday

#### Frequency-hydrophobicity superposition in associative polymers with comb-like architecture Michel Cloitre and Arnaud Chaub

Molecular, Macromolecular Chemistry and Materials, ESPCI-PSL, Paris, France

Associative gels are temporary networks made of polymer chains connected by short-lived associations. They can deform and flow when submitted to external mechanical solicitations. These properties make them valuable materials in applications such as 3D printing, injectable gels for tissue reconstruction, or rheology additives for paints and coatings.

Many types of associative polymers are now available, encompassing a variety of architectures and interactions. Here we focus on comb-like polymers comprising a polyelectrolyte backbone and lateral pending chains made of a hydrophilic spacer end-capped by a hydrophobic alkyl group. The hydrophobic groups associate into micelles that connect the polymer chains. The strength of the associations is tuned at will by changing the length of the alkyl groups, leading to a huge variety of materials suitable for applications. So far, a general framework connecting the viscoelastic properties of solutions to the interactions is still missing.

Here we take on this challenge using associative polymers with the same backbone, which are functionalized with a series of alkyl groups with an increasing number of carbon atoms (n). When  $\leq 20$ , solutions are viscoelastic liquids; when n = 32, gelation occurs at a well-defined polymer concentration. We collapse the viscoelastic spectra of this family of associative polymers over a wide range of frequencies and concentrations by introducing a concept of frequency-hydrophobicity superposition. The master curves, which cover several decades of frequencies, are well described by the sticky Rouse model. Finally, we discuss and justify the validity of the frequency-hydrophobicity superposition principle at the molecular level.

#### Friday 12:20 LT 17 (Level 7) / Track 2

Resolving the molecular mechanisms controlling the rheology in a supramolecular telechelic polymer

Johan Mattsson<sup>1</sup>, Guanghui Cui<sup>2</sup>, Simon Connell<sup>3</sup>, Matthew Reynolds<sup>4</sup>, Victor Boudara<sup>5</sup>, Daniel J. Read<sup>6</sup>, and Andrew Wilson<sup>7</sup> <sup>1</sup>School of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, United Kingdom; <sup>2</sup>School of Physics and Astronomy, University of Leeds, Leeds, United Kingdom; <sup>3</sup>School of Physics and Astronomy, University of Leeds, Leeds, West Yorkshire LS2 9JT, United Kingdom; <sup>4</sup>School of Physics and Astronomy, University of Leeds, Leeds, United Kingdom; <sup>5</sup>School of Mathematics, University of Leeds, Leeds, United Kingdom; <sup>6</sup>School of Mathematics, University of Leeds, Leeds LS29JT, United Kingdom; <sup>7</sup>School of Chemistry, University of Birmingham, Birmingham, United Kingdom

Supramolecular polymers are important for applications including printing, adhesives, coatings and nano-fabrication, and the ability to tune their properties by control of the supramolecular associations makes these materials both versatile and powerful. Here, we present a detailed experimental investigation of a telechelic supramolecular polymer UPyPPG made from poly(propylene glycol) end-functionalised with hydrogen bond associating pyrimidinone groups. This particular polymer exhibits fascinating dynamics, combining very slow flow with relatively fast segmental and chain dynamics, resulting in self-healing properties. We demonstrate the origin of these properties at the molecular level through a combination of rheology, AFM, broadband dielectric spectroscopy (BDS) and X-ray scattering (SAXS/WAXS). Using small amplitude oscillatory shear (SAOS) rheology, we compare the response of UPyPPG to pure PPG, demonstrating a dramatic slowing down in the terminal rheological response which is linked to the Arrhenius behaviour of the local chain dynamics (normal mode response probed by BDS). We identify the effects to be due to both association-driven chain-extension and to the presence of nano-scale fibre-like aggregates; the actual molecular structure is revealed through a combination of atomic force microscopy (AFM) and small and wide angle X-ray scattering (SAXS/WAXS). We track the molecular relaxation dynamics in detail with high-speed AFM to directly characterise the motions of the fibre-like aggregates down to nanometre lateral resolution and time steps of seconds, providing detailed information about the material behaviour at the nano-scale. For the first time, we apply a differential dynamic microscopy (DDM) approach to quantify the AFM nanoscale relaxation movies, providing a link between the nanoscale motions and the bulk rheology, thereby providing a fundamental insight into the molecular mechanisms controlling the behaviour of this polymer system.

#### Friday 12:40 LT 17 (Level 7) / Track 2

#### **PF36**

# Diffusion Mechanisms in Star-Shaped Associating Polymers: Insights from Molecular Dynamics and Monte **Carlo Simulations**

Javier Oller<sup>1</sup> and Jorge Ramírez<sup>2</sup>

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Recent studies have advanced the understanding of the complex self-diffusive behaviour in unentangled associating polymers. This research particularly focuses on the diffusion of star-shaped associating polymers, studying diverse diffusion mechanisms such as hopping and walking

**PF35** 

**PF34** 

across various spatial and temporal scales. Previous models explaining this anomalous diffusion behaviour often relied on mean-field approaches, which might oversimplify intermolecular associations. To enhance this understanding, Molecular Dynamics simulations have been employed, utilizing the Kremer-Grest model with an explicit solvent. This methodology comprehensively represents network fluctuations. Additionally, a Monte Carlo algorithm has been integrated to depict sticker association more accurately. This combination of techniques offers a nuanced view of polymer behaviour, linking chemical details and network topology to empirical measures like relaxation modulus and self-diffusion coefficients. The research spans a broad concentration range, from dilute to concentrated solutions, enabling an examination of different network defects. These defects provide insights into the mechanisms underlying star-shaped polymer diffusion, revealing their behaviour across varying time and space scales. This in-depth exploration sheds new light on the multifaceted nature of polymer diffusion, enhancing the understanding of the physical processes governing the movement of star-shaped associating polymers.

# Symposium FR

# Food Rheology

Organizers: Jan Engmann, Peter Fischer and Brent Murray

#### Friday 8:50 LT 6 (Level 8) / Track 3

Recent advances in the engineering of concentrated soft microgel dispersions for food applications

Deniz Gunes<sup>1</sup>, Gabriele D'Oria<sup>2</sup>, Christophe Hartmann<sup>3</sup>, Hans-Joerg Limbach<sup>4</sup>, and Lilia Ahrne<sup>5</sup> <sup>1</sup>CIT & M2S, KU Leuven, Leuven 3000, Belgium; <sup>2</sup>Department of Food Science, Faculty of Science, University of Copenhagen, Copenhagen 1958, Denmark; <sup>3</sup>Nestle Research, Lausanne 1000, Switzerland; <sup>4</sup>Nestle Research, Lausanne 1000, Switzerland; <sup>5</sup>Department of Food Science, Faculty of Science, University of Copenhagen, Copenhagen 1958, Denmark

Concentrated suspensions of soft microgel particles are yield stress fluids, also called fluid gels in the context of food applications, e.g. for increasing in-mouth lubrication. Despite they are thought for a long time to possess the remarkable property of low controllable yield stress [1], the mechanisms of their measurable aging, and the question of their reversibility were still to be fully established. Such insights should allow for their application for a variety of applications, via fine control of their reversibility were still to be fully established. Such insights should allow for their application for a variety of applications, via fine control of their rheology including their long-term behavior. Our studies were mainly based on an anionic (gellan) polysaccharide and their comparison with non-ionic fluid gels, whereby we found evidence for the following: a) fluid gels display physical aging properties like colloidal glasses do, which can be fully reversed by flow [1]; b) particle shapes become significantly elongated at high flow rate, influencing their rheology [2]; c) the influence of the effective particle volume fraction is similar to that of core-shell particles; d) there is significant influence of the continuous phase salt environment, which is larger for fluid gels from ionic polysaccharides. Furthermore, we demonstrated a new approach based on a simple process, which allows for particle encapsulation in a shell of gel material, in other words of coating particles with a thin gel layer of the fluid gel base material. We applied the technique to coat (e.g. gritty and astringent perceived fiber particles) i.e. to increase their palatability. Our data show significant reduction of the astringency and grittiness perception of the particles thanks to the coating [3], which shows we have a promising the approach towards an increased palatability (in view of liquid food applications).

1. D'Oria, Gunes, Lequeux, Hartmann, Limbach, Ahrné. Food Hydrocoll. 137, 108401 (2023) 2. D'Oria, Zeng, Limbach, Hartmann, Ahrne´, Gunes. Food Hydrocoll. 149, 109614 (2024) 3. D'Oria, Bredie, Limbach, Hartmann, Gunes, Ahrne. Abtracts of EFFoST 2023, Valencia (2023)

#### Friday 9:10 LT 6 (Level 8) / Track 3

# Linking the yield stress functionality of polyglycerol polyricinoleate in a highly filled suspension to its molecular properties

<u>Ruth Price</u><sup>1</sup>, David A. Gray<sup>1</sup>, Nicolas Watson<sup>2</sup>, Josélio Vieira<sup>3</sup>, and Bettina Wolf<sup>4</sup> <sup>1</sup>Division of Food Science, Nutrition and Dietetics, University of Nottingham, Leicester, Nottinghamshire LE12 5RD, United Kingdom; <sup>2</sup>Division of Food Science and Nutrition, University of Leeds, Leeds LS2 9JT, United Kingdom; <sup>3</sup>Nestlé Product

Technology Centre, York YO91 1XY, United Kingdom; <sup>4</sup>Chemical engineering department, The University of Birmingham, Birmingham B15 2TT, United Kingdom

Consumers increasingly insist on only natural ingredients in their food, so understanding how synthetic ingredients work at a fundamental level is essential in our pursuit for natural alternatives. This research focuses on polyglycerol polyricinoleate (PGPR), a food-grade emulsifier known for its unique ability to reduce yield stress in fat-based suspensions. Although its general structure is well known, the method of PGPR production follows an established protocol, finishing when the optimal performance range is achieved rather than a set chemical profile.

Five commercial PGPR samples were obtained with different efficacies empirically noted during use in food manufacture. PGPR was added at 0.3% to a chocolate model: a concentrated suspension of finely ground sugar in sunflower oil (pre-treated to remove naturally occurring surface-active molecules). The evaluation of the yield stress reducing capacity was conducted using a rheometer equipped with serrated concentric cylinder geometries. A difference in the yield stress was observed; the calculated Herschel-Bulkley yield stress varied between  $0.90 \pm 0.06$  Pa and  $1.90 \pm 0.18$  Pa, compared to  $57.6 \pm 15.8$  Pa in the absence of PGPR. The ranked order of efficiency of the PGPRs reflected the empirical observations and was supported by data obtained using interfacial tension analysis.

FR5

FR6

The molecular weight range of the molecules within these PGPR samples was analysed using size exclusion chromatography, electrospray ionisation-mass spectrometry and nuclear magnetic resonance. The results showed very little difference in the molecular weight range, which inferred that the molecular size of PGPR does not affect its efficacy, as initially hypothesised. The analysis of the terminal fatty acid on the polyricinoleate chain suggested that the presence of a hydroxyl group on the fatty acid group was linked to inferior yield stress reducing power. We hypothesised that this hydrophilic character at the end of the hydrophobic chain is the reason for the reduced efficacy.

### Friday 9:30 LT 6 (Level 8) / Track 3

#### Bolus rheology of texture-modified food for swallowing disorder management

FR7

Mats Stading<sup>1</sup>, Ana Miljkovic<sup>2</sup>, Johanna Andersson<sup>2</sup>, and Koichiro Matsuo<sup>3</sup>

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Texture-modified foods are used to manage swallowing disorders, with the degree of modification depending on the severity of the condition. Rheological and physiology-related properties of boluses were determined for a set of texture-modified foods, for a group of younger subjects and compared to a group of older subjects.

The softest class in the set for texture-modified foods was gel food, then a smooth timbale which both were compared to the corresponding regular, unmodified food. The foods investigated were bread, cheese, tomato and the combination as a sandwich. The subjects chewed until ready to swallow and the expectorated bolus was immediately measured for  $G^*$  and ?, and moisture and saliva content was determined.

As the degree of texture modification increased, the viscosity ? and modulus  $|G^*|$  of the bolus decreased. Saliva content and chews-to-swallow also decreased with increased modification. The combination of foods (sandwich) showed lower saliva content compared to individual components. The older group had lower age-related properties such as one-legged standing, salivary flow, and bolus saliva content as expected, but were otherwise rather healthy and fit. Surprisingly, they needed fewer chews-until-swallow and consequently the bolus ? and  $|G^*|$  were higher in the older group compared to the younger group.

Bolus rheology plays a critical role in determining the safety of swallowing. Overall, the intended texture modification was reflected in bolus rheological and physiological-related properties. Bolus modulus, viscosity, saliva content and chews-until-swallowed all decreased from regular food to timbale food to gel food. Age-related changes were also observed, with older individuals exhibiting higher bolus modulus and viscosity compared to younger individuals. Overall, when comparing the older group with the younger group, it is interesting to note that there are differences with age, but as the older group was healthy and fit, the differences were relatively small.

#### Friday 9:50 LT 6 (Level 8) / Track 3

# Rheo-mechanics of mixtures of K-Carrageenan and other polysaccarides. A critical study on potential gelatin alternatives

#### Simona Russo Spena, Rossana Pasquino, and Nino Grizzuti University of Naples Federico II, Naples, Naples 80040, Italy

The basic texture of a wide variety of formulated food depends on the gelling capabilities of polysaccharides and proteins, two polymer families that are increasingly used as a texture modifier. Among commercial hydrocolloids used in the food industry, gelatin (an animal protein) is remarkably known for its unique gel forming ability. Creating a perfect, possibly green substitute for animal gelatin is extremely difficult if not impossible, because this versatile hydrocolloid offers many special properties that are not easily imitated by other, similar systems. Numerous attempts have been made to mimic the texture of gelatine gels by combining different gelling agents. Combination of more than one type of hydrocolloids is commonly used in food too impart novel organoleptic (mouthfeel) to food product, modify rheological characteristics, and satisfy processing requirements in the industry. In this work we study the rheology and the texture of water mixtures of k-Carrageenan (k-C) and other polysaccharides (PS). By fixing different KC concentration (1, 1.5, 2% by weight) and varying the k-C/PS ratio, we explore a wide range of interesting and potentially usefully texture. These hydrocolloids interact synergistically forming a thermo-reversible gel. The results obtained for the green systems have been compared to animal gelatin formulations. The latter has a fixed concentration of 6.67%, which is generally used as a reference for gelatin properties characterization. The objective of this study is twofold. On the one hand, we deeply study the rheological behavior of the composed vegetable system. On the other hand, we try to analyze the results from a critical point of view to explain how different rheological tests and Texture Profile Analysis (TPA) analysis can provide information on the mechanical behavior, for food applications, of a vegetable gel system and comparison with a reference animal gelatin.

#### Friday 10:10 LT 6 (Level 8) / Track 3

FR9

FR8

# To which extent can food texture perception be explained in terms of food rheological and tribological properties?

<u>Marco Ramaioli</u><sup>1</sup>, Yurixy Bugarin-Castillo<sup>1</sup>, Alejandro Avila-Sierra<sup>1</sup>, Anne Saint-Eve<sup>2</sup>, Anais Lavoisier<sup>3</sup>, Vincent Mathieu<sup>1</sup>, and Vincent Mathieu<sup>4</sup>

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Recent results providing new insights on the link between the rheological and tribological properties of different food products and their perceived texture will be reviewed in this contribution. Different biomimetic instrumental approaches developed at INRAE to study in vitro some key aspects

of food oral processing will be introduced, each with a different focus: tongue-palate shearing, fragmentation, hydration by artificial salivary fluids and swallowing. The importance of considering boundary conditions relevant for oral conditions will be discussed. The shear and extensional rheology of different food products have been characterized at different strain rates. Elastic liquids with low shear viscosity are excellent lubricants and induce low fiction between tongue and palate in vitro and are perceived as highly lubricious in vivo. Specific sensory protocols were developed to evaluate perceived lubrication, adhesiveness and other textural attributes. Elasticity coupled to low shear viscosity can be leveraged when formulating natural salivary substitutes and can lead to a compact bolus facilitating swallowing and the administration of solid oral dosage forms. The enzymatic activity of saliva can alter the rheological properties of some semi-solid products on a time-scale comparable to the oral transit time and can have a strong impact on the dynamics of swallowing and on the control of the bolus on the tongue. Similarly, ?-amylase can also affect the properties of the bolus resulting from the mastication of solid food, such as bread. Current limitations of the in vitro biomimetic experiments to study food oral processing will be discussed as well as the perspectives for future research to further understand the link between rheological and tribological properties and perceived food texture.

Y.Bugarin-Castillo et al., On the rheological and sensory properties of a novel natural salivary substitute, 2023, subm. A.Lavoisier et al., 2022, A novel soft robotic pediatric in vitro swallowing device, Int.J.Pharm.

#### 11:00 LT 6 (Level 8) / Track 3 Friday

#### Structural and contact-driven ageing in enzymatic casein gels

Julien Bauland<sup>1</sup>, Thibaut Divoux<sup>1</sup>, Ramya A. Koduvayur<sup>2</sup>, Christopher J. Garvey<sup>2</sup>, and Thomas Gibaud<sup>1</sup> <sup>1</sup>Laboratoire de physique, ENSL, Lyon 69007, France; <sup>2</sup>Heinz Maier-Leibnitz Zentrum, Technische Universität München, Garching, Germany

In the limit of strong particle interactions and low volume fraction, colloidal suspensions form gels, whose elasticity arises from the stress-bearing capacity of the particle network. In many "real-life" gel systems, the interparticle interaction energy is of the order of the thermal energy and bonds have a finite lifetime. Consequently, weak colloidal gels show time-dependent properties, referred to as "aging", and attributed to structural rearrangements as the structure explores deeper energy minima. Recently, it was shown that a change of interparticle interactions with time could also lead to aging without structural changes. It demonstrates that the time-dependent properties of colloidal gels can have different origins and require systematic characterization to draw a general picture. Enzymatic milk gels are natural colloidal gels formed through the destabilization of "casein micelles", a colloidal structure composed of proteins and minerals. In this study, we use time-resolved mechanical spectroscopy to investigate the time-evolving viscoelastic properties of enzymatic milk gels. We first show that, by applying a time-superposition principle, the mechanical spectra acquired at various intervals following the gel point can be rescaled onto a single master curve. This master curve shows that aging consists of a continuous transition from a critical gel characterized by a power-law viscoelastic response with an exponent  $\alpha = 0.6$  to a mature gel with a power exponent  $\beta = 0.3$ . These power exponents can be consistently related to the evolution of the gel structure probed by SAXS and confocal microscopy. When the mesoscopic structure of the mature gel is set, aging proceeds by contact aging solely, as evidenced by the scaling of the yield stress with the square root of the elastic modulus. These results offer a comprehensive understanding of the complex aging scenario in enzymatic casein gels and provide a benchmark set of data for future theoretical studies.

#### Friday 11:20 LT 6 (Level 8) / Track 3

**FR11** 

**FR10** 

#### Viscoelastic properties of lactoferrin/ β-lactoglobulin coacervates studied by rheology and microrheology Ghazi BEN MESSAOUD, Rima SOUSSI HACHFI, Florence ROUSSEAU, Pascaline Hamon, Marie Hélène Famelart, and Saïd **BOUHALLAB**

#### STLO, INRAE, Institut Agro, Rennes 35042, France

Heteroprotein complex coacervates (HPCC) has great potential in many food applications. However, understanding the rheology-coacervate structure relationship, as well as their sensitivity to slight changes in the physicochemical environment, is still an active research topic. Herein, HPCC between two oppositely charged proteins, lactoferrin (LF) and β-lactoglobulin (βLG) was investigated. The influence of ionic strength and temperature on the rheological properties of LF/BLG coacervates was examined using oscillatory shear rheology and microrheology from dynamic light scattering. LF/ $\beta$ LG HPCC exhibited a liquid-like character with G'( $\omega$ ) < G"( $\omega$ ) and an increase of both moduli with decreased temperature but a softening effect with increased ionic strength. The dependency of G' and G'' on angular frequency ( $\omega$ ) demonstrated a scaling of G''  $\sim \omega^1$  and a lack of terminal behavior with G'~ $\omega$ <sup>1.4</sup>. The application of time-temperature superposition (TTS) and time-salt superposition (TSS) principles allowed the prediction of the rheological properties over a wide range of timescales and temperatures below the denaturation temperature of  $\beta$ LG and LF. The two principles suggested that increasing temperature or ionic strength accelerates coacervates dynamics but does not affect largerscale physics. Microrheology experiments using polystyrene-coated microspheres as tracers, allowed access to a frequency range up to ( $\omega \sim 10^6$ rad/s) and revealed a variable scaling of G'= G'' ~  $\omega^{3/4}$  at the high-frequency terminal regime and approaching theoretical predictions of Rouse regime and Worm-Like Chain model for polymers. In the present case, it reflected a freely draining system where hydrodynamic interactions are neglected at these timescales. This study offers new insights into the microstructure of HPCC, relevant to various applications, as function of ionic strength and temperature.

# Friday 11:40 LT 6 (Level 8) / Track 3

# Thermo-rheological properties of a gelatin analogue mixed gel of Low acyl gellan gum and tamarind seed xyloglucan as a dysphagia meal

Thidarat Makmoon<sup>1</sup>, Tom Mills<sup>2</sup>, and Bettina Wolf<sup>3</sup>

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There is renewed interest in developing gelatin analogue gels due to the rise in consumers following a vegan diet. With this dietary trend, the number of vegan patients presenting eating disorders, such as dysphagia, who benefit from melt-in-the mouth meals to avoid medical complications is on the rise too. Building on a recently introduced mixed gel system of relevant melting properties (Fenton et al, 2021), synergistic combinations of low acyl gellan gum (LAG) and tamarind seed xyloglucan (TSX), with up to 2.5 wt% total biopolymer concentration, were studied, using rheology and thermal analysis methods. LAG is a microbial polysaccharide with ability to form brittle clear gels. TSX is a plant polysaccharide with weak gelation property. The formulation target was a melting temperature close to in-mouth temperature, and a soft gel texture. Successful formulations and a microstructure model for LAG-TSX interactions will be discussed. Data acquired so far suggest that LAG forms the dominant network with pore spaces occupied by TSX and some degree of molecular interaction between the two biopolymers. Future work will consider the incorporation of plant protein and an oil dispersed phase into the gelatin analogue gel, to enhance the nutritional properties of this potential dysphagia meal.

#### Friday 12:00 LT 6 (Level 8) / Track 3

#### Characterisation of chemo-mechanical properties of starch gels

Zisheng Liao<sup>1</sup>, Alexandros Makrypidis<sup>1</sup>, Aaron M. Lett<sup>2</sup>, Aygul Dagbasi<sup>2</sup>, Gary Frost<sup>2</sup>, Maria M. Papathanasiou<sup>3</sup>, and Maria N. Charalambides<sup>1</sup>

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The gastric digestion of food involves both the mechanical breakdown via peristaltic waves of the stomach wall and the chemical breakdown facilitated by the diffusion and reaction with the gastric liquids. This study investigates the mechanical and rheological properties of starch gels under chemical loads in in-vitro breakdown conditions, in an effort to replicate in-vivo digestion environments. Compression tests and shear rheology tests are performed across different levels of water diffusion, swelling, and starch hydrolysis catalysed by  $\alpha$ -amylase. A pseudoelastic-viscoelastic constitutive model based on continuum mechanics and thermodynamics is proposed to describe the chemo-mechanical behaviour of starch gels. The material properties are dependent on the extent of water absorption and hydrolysis. The model is calibrated using the experimental data and is implemented in the finite element software to simulate the compression and shear tests for validation. The results show the efficacy of the material model in capturing the chemo-mechanical responses of starch gels. The material law and numerical simulation technique developed in this study provide tools for predicting the in-vivo gastric digestion process of foods.

#### Friday 12:20 LT 6 (Level 8) / Track 3

# Clarifying the deformation behaviour of meat and meat analogues

Elle I. Wilhelm<sup>1</sup>, José A. Rodríguez Agudo<sup>2</sup>, David O. Schmelzeisen<sup>3</sup>, and Anja M. Wagemans<sup>4</sup>

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Plant-based meat analogs serve as a sustainable alternative to meat, yet current attempts to mimic the visual anisotropic structure on a macroscale fall short of fully reproducing the sensory experience of meat products. A significant challenge lies in understanding the mechanism of failure, in particular the irreversible structural deformation of meat that occurs during the initial bite, which is different for meat and meat analogs. Consequently, this study aims to elucidate the mechanism of failure in both meat and meat analogs, employing a multi-scale approach to describe the structural deformation and textural properties during the initial compression. Firstly, the macroscale mechanical anisotropy was characterized using texture profile analysis by measuring samples parallel and perpendicular in the fiber direction. Secondly, the irreversible structural deformation and anisotropy were determined by rheological measurements in and out of the linear viscoelastic regime in two different deformation modes, shear (G', G'') and compression (E', E''). Thirdly, the mesoscale structural changes were characterized by scanning electron microscopy and by coupling polarized light microscopy with rheological shear measurements. Meat analogs exhibited macroscale anisotropy, as indicated by the directional dependence of springiness. In contrast, meat exhibited macroscale isotropy with no directional dependence. Meat analogs exhibited a longer linear viscoelastic range, which was associated with their high springiness. The linear viscoelastic range was found to correlate with the degree of mesoscale structural deformation. These results contribute to a deeper understanding of the textural properties of meat.

FR13

FR14

**FR15** 

# Friday 12:40 LT 6 (Level 8) / Track 3 Challenges in obtaining the steady-state shear viscosity of protein-water mixtures under extrusion-like conditions

Felix Ellwanger<sup>1</sup>, Heike P. Karbstein<sup>1</sup>, and Azad M. Emin<sup>2</sup>

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Meat substitutes made from plant proteins have recently established themselves on the refrigerated shelves of supermarkets and mimic classic meat products in terms of taste and texture. The meat substitutes are mainly produced using the twin-screw extruder, with functionalization of the proteins taking place in the screw section and structuring taking place in the die section. While the die section of the extruder can be characterized and represented well, the screw section is challenging, which is why it is often regarded as a black box. Numerical flow simulations offer the opportunity to investigate the screw section of the extruder. In addition to the numerical model, the success of the simulation is largely dependent on the material properties, in particular the steady-state shear viscosity of the material used. Determining the steady-state shear viscosity under extrusion-like conditions is a challenge for protein-water mixtures, as water loss and reactions taking place can influence the measurement. Closed-cavity rheometers (CCR) offer a possible solution, but so far they have hardly been used to investigate the steady-state shear viscosity. In this conference contribution, we shed light on the challenge of determining the steady-state shear viscosity of highly concentrated protein-water mixtures. The ramp test, a method for obtaining the steady-state shear viscosity of a CCR, will be presented. First, to verify the method and the accuracy of the CCR data, three polyolefin polymers, a low density and a linear low density polyethylene and a polybutadiene, were used as model systems. Measurements of the magnitude of the complex viscosity were compared with the steady-state shear viscosity data obtained by capillary rheometer and CCR. Finally, the established method is used to analyze the protein-water mixtures. It is presented under which conditions the measurements are successful and what information can be obtained.

# Symposium SG

# **Suspensions and Granular Materials**

Organizers: Erin Koos, Wilson Poon and Olivier Pouliquen

#### Friday 8:50 LT 18 (Level 8) / Track 4

SG25

**Rheological characterization of primary sewage sludge for ecological and economic exploitation** <u>Ana Chitanu</u><sup>1</sup>, Laurent Jossic<sup>1</sup>, Albert Magnin<sup>1</sup>, André Pauss<sup>2</sup>, Thierry Ribeiro<sup>3</sup>, and Marcos Oliveira<sup>4</sup> <sup>1</sup>Laboratoire Rhéologie et Procédés, CNRS, Grenoble-INP, Université Grenoble Alpes, Grenoble, Isère 3800, France; <sup>2</sup>Laboratoire Transformations Intégrées de la Matière Renouve, Université de Technologie de Compiègne, Compiègne, Oise 60200, France; <sup>3</sup>Institut Polytechnique UniLaSalle, Université d'Artois, Beauvais, Oise 60026, France; <sup>4</sup>Direction Innovation, SIAAP, Colombes, Hauts-de-Seine 92700, France

Wastewater treatment plants (WWTP) generate large quantities of sewage sludge, the management of which is a regulated phase of fight against pollution. Nowadays, sludge management is considered as a parallel step to wastewater treatment, presenting relative costs rising up to 50% of the total operating costs of a WWTP [1]. Thus, for an economic and ecological reason, the development of a valorisation strategy by exploiting sewage sludge's value as a fertiliser for soil, in construction for earthworks and as renewable energy by producing biogas is of great importance.

Sewage sludge is a complex heterogeneous mixture of flocs, organic and inorganic colloids, fibres, microorganisms, extracellular polymers, and ions in water, whose consistency varies greatly during treatment operations. They go from primary sludge with or without a low yield stress to dehydrated sludge with a very high yield stress. Thus, the study of the rheological behaviour becomes crucial to properly determine and optimise the designing parameters in transporting, pumping, storing, and heat transfer operations.

In this study, the developed methods apply to primary sludge, containing a low dry matter content, that are likely to sediment during rheological measurements. This phenomenon makes the material heterogeneous under shear and leads to distorted experimental results. To remedy this, measurement protocols have been developed in 3D printed transparent and rough co-axial cylinders, to control sedimentation through visualizations and slip at the wall. This proposed methodology allows to evaluate the influence of the pre-shear rate on the resuspension of the particles. Moreover, the effect of the shear rate on the sedimentation rate was evaluated to measure the steady state flow curve of a homogeneous material.

[1] L. Appels, J. Baeyens, J. Degrève, and R. Dewil, "Principles and potential of the anaerobic digestion of waste-activated sludge," Prog. Energy Combust. Sci., vol. 34, no. 6, pp. 755-781, 2008

# Friday9:10LT 18 (Level 8) / Track 4Numerical simulations of the sedimentation of soft particles in confined liquidsMassimiliano M. Villoneand Pier Luca Maffettone

University of Naples Federico II, Naples, Italy

The gravitational sedimentation of solid particles in liquids is important in several applications, many of which involve soft particles (e.g., microplastics, microgels, and biological cells). However, there is still little knowledge on the sedimentation of deformable beads. We employ fully three-dimensional finite-element numerical simulations to investigate the sedimentation of an initially spherical elastic bead in a Newtonian liquid contained inside a square-cross-section vessel. In particular, we carry out a wide parametric analysis to understand the effects of inertia, particle deformability, and geometrical confinement on the particle sedimentation dynamics and settling velocity.

# Friday 9:30 LT 18 (Level 8) / Track 4

# Unravelling the influence of alternated oscillatory and continuous shear on particle alignment in viscoelastic liquids

Sebastian Gassenmeier<sup>1</sup>, Christophe De Graaf<sup>2</sup>, Anja Vananroye<sup>1</sup>, Ruth Cardinaels<sup>1</sup>, Erin Koos<sup>1</sup>, and Deniz Gunes<sup>3</sup>

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The formation of particle strings under shear in viscoelastic media is an elegant method to induce anisotropy in suspensions, with potential implications for structuring of meat analogues and synthesis of other multiparticle structures. Shear thinning and elastic stresses, quantified by the Weissenberg (Wi) number, arise in the medium under flow and drive the particles to arrange in pearl-necklace like string structures [1,2].

We use an optical shear sell to set the shear protocol, simultaneously capturing micrographs of the suspended particles. A model system of noncolloidal PS-spheres and a density matched PEO-solution allowed for variation of Wi-numbers. Leveraging computer vision based on a machine learning model, we obtained time-resolved data of string formation and properties.

Surprisingly, we found that strings form at lower Weissenberg numbers in oscillatory flow, and that alignment is more pronounced than for steady shear. Furthermore, we demonstrated this difference by alternating between the two shear modes. We observed a Wi-dependent switch from rapid re-dispersion to slower de-aggregation of strings when switching from oscillatory to steady, and track the remaining order in the suspension by quantifying the degree of the cumulative effect in alignment from one cycle to another. Finally, we connect the found differences between shear protocols to the elastic forces driven by shear-induced alignment phenomena in the suspending medium.

[1] Michele, R. Pätzold, and R. Donis. Alignment and aggregation effects in suspensions of spheres in non-Newtonian media. *Rheologica Acta*, 16:317-321, 1977

[2] N. O. Jaensson, M. A. Hulsen, and P. D. Anderson. Direct numerical simulation of particle alignment in viscoelastic fluids. *Journal of Non-Newtonian Fluid Mechanics*, 235:125-142, 9 2016.

#### Friday 9:50 LT 18 (Level 8) / Track 4

# Layer formation in magnetorheological fluids

Jose R. Morillas<sup>1</sup>, Óscar Martínez-Cano<sup>1</sup>, Jeffrey F. Morris<sup>2</sup>, and Juan de Vicente<sup>1</sup>

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Under the application of a magnetic field and a progressively growing shear flow, the inner structure of magnetorheological fluids evolves from a collection of upright chains to a striped pattern consisting of particle layers contained in the flow-gradient plane. This picture totally disagrees with the scenario conceived by the traditional chain model [1] that predicts the progressive destruction of the chains till a point where only individual particles are homogeneously distributed in the sample.

Previous works in the literature have concluded that the onset for layer formation is controlled only by the Mason number (Mn, ratio between shear and magnetic forces). Namely, it happens at the Mn responsible for the breakage of the shortest (two-particle) chains in the sample [2]. However, the majority of those works studied the layer pattern once the flow had ceased or did not use samples of interest in current applications.

In this work, the chain-layer transition and pattern formation are reported as Mn is increased during rheograms thanks to the use of a video camera fully synchronized with a rheometer in a plate-plate configuration. Both the sample rheological signature and inner structure (pattern morphological properties) are analysed and correlated with each other. It is found that the layered pattern is strongly affected not only by Mn but also by the gap between plates. In particular, the layer onset happens at smaller Mn as the gap increases while the layer period and width increase linearly with the gap. Finally, it is proposed a numerical model able to explain the previous experimental findings and reconcile them with the widely-accepted chain model.

#### Acknowledgements:

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References:

[1] Martin, J. E., Anderson, R. A. (1996). J Chem Phys, 104, 4814.

[2] Volkova, O., Cutillas, S., Bossis, G. (1999). PRL, 82, 233.

SG27

SG29

#### Friday 10:10 LT 18 (Level 8) / Track 4

Study the concentration of nanofillers on curing behavior of silicone elastomer

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Silicones have advantages like transparency, biocompatibility, chemical resistance, thermal stability, and flexibility. Curing time is a crucial parameter to consider when dealing with large-scale processing. The potential of silicone materials for further exploration is highlighted by the prospect of enhancing these properties through different fillers. However, the addition of fillers changes not only the flow behavior but also the curing time of the materials. This study investigates the influence of fumed silica nanoparticles (NPs) on the flow behavior and curing time of silicone elastomer materials. Three quantities of fillers (5, 30, 60 v/v %) are utilized to explore the flow behavior of the composites through frequency sweep (FS) analysis conducted before and after curing. Samples containing 30% and 60% NP exhibit heightened viscosity and distinct thixotropy at lower frequency regions. In addition, the Time-Resolved Mechanical Spectroscopy (TRMS) method is applied to determine curing times at 70°C. Results indicate an increase in the curing time of the sample with the addition of 5% and 30% NP, followed by a substantial decrease in samples with 60% NP. However, the Differential Scanning Calorimetry (DSC) measurements reveal that curing peaks shift to higher times with an increasing concentration of NP. This phenomenon can be attributed to the relationship between the chemical reaction's kinetics and the fillers' physical hindrance. DSC measurements confirm that nanoparticles prolong the chemical curing time. This is why 5% and 30% of the samples showed longer curing times in TRMS. However, the highly filled nanoparticles (e.g., 60 % sample) increased the spatial hindrance of the system, leading the material close to the gel point. As a result, the curing time of the 60% sample was shorter than the other samples

#### Friday 11:00 LT 18 (Level 8) / Track 4

# Particle Redistribution in Complex Flow of Complex Fluids

#### Mahdi Davoodi<sup>1</sup> and Andrew Clarke<sup>2</sup>

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SLB is a leading oilfield services company that has provided sub-surface measurement, well-construction and production services to the oil and gas industry worldwide for the last 100 years. A major part of well-construction is drilling the well which, despite being a 100yr old industry, is a process that still experiences significant technical challenges. These industry challenges continue to be highly relevant as slb extends its, already significant, activity in adjacent energy transition technologies that also require deep drilling, that is geothermal, carbon capture & sequestration and sustainable lithium production. Here I introduce the company, drilling fluids and the essential parts of a well-construction process. I focus on the ability of drilling fluids to carry rock cuttings to the surface. Within the industry this particle-carriage problem is, in general, modelled using a generalized Newtonian fluid. Yet there are repeated industry reports comparing fluids where the flow curves are closely similar but where cuttings transport is demonstrably different. To probe this, experimentally we combine a Xanthan solution as a proxy for a water-based drilling fluid together with an aqueous Bentonite dispersion that at low concentrations in the Xanthan solution is representative of shale contamination of a water-based drilling fluid. The aqueous Bentonite solution alone has rheological properties similar to those of an oil-based drilling fluid. The concentrations of each of the two fluids, Xanthan and Bentonite, are chosen so that their generalized Newtonian properties are closely similar. However, their extended rheological properties, in particular normal stress differences, differ markedly. We examine dense particle motion in a horizontal Couette in the presence of Taylor vortices for each fluid using both experimental and numerical approaches. We find significant effects dependent on fluid type which, through comparison with our model, we speculatively assign as primarily due to normal forces generated by

#### Friday 11:20 LT 18 (Level 8) / Track 4

#### On the linear viscoelastic behaviour of semidilute polydisperse bubble suspensions in Newtonian media

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In this work, we investigated the linear viscoelasticity of semidilute polydisperse bubble suspensions via small ampliptude oscillatory shear (SAOS) tests performed in a rheo-optical set-up. For all tested suspensions, the measured viscoelastic moduli (G', G") aligned with the theoretical predictions of the Jeffreys model for average dynamic capillary numbers (<Cd>) greater than unity. But at lower <Cd> values, experimental G' values exceeded theoretical predictions.To investigate this, we considered the effects of suspension polydispersity, bubble rise, clustering, alignment, and coalescence. Polydispersity could not cause the observed deviation, as the trends of G' and G" deviate from those of classic single-mode relaxation only for bimodal bubble size distributions with equal volume fractions of very small and very large bubbles; in any other case, the polydisperse suspension behaves as monodisperse with a bubble radius equal to the volume-weighted average radius. Furthermore, bubble rise proved to play a minor role, while SAOS rheo-optical experiments revealed negligible microstructural changes owing to bubble clustering, alignment and coalescence induced by the rheological tests. The G' deviation at low <Cd> values was linked to bubble fluid dynamic interactions induced by the bubble spatial distribution. Image analysis showed that at low bubble volume fractions, stronger and prolonged pre-shearing reduces these interactions by increasing the average inter-bubble distance. But this effect is negligible in denser suspensions, which show similar G' trends for any applied pre-shearing. Finally, a multi-mode Jeffreys model fitted to the experimental G', G" values showed that bubble interactions complicate the relaxation process, leading to multiple relaxation modes.

SG30

# Friday 11:40 LT 18 (Level 8) / Track 4

Effects of salinity on flows of dense colloidal suspensions

Marc Lagoin, Rémy Fulcrand, Agnès Piednoir, and Antoine Bérut

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We experimentally study the effects of salt concentration on the flowing dynamics of dense suspensions of micrometer-sized silica particles in microfluidic drums. In pure water, the particles are fully sedimented under their own weight, but do not touch each others due to their negative surface charges, resulting in a "frictionless" dense colloidal suspension: when the pile is in inclined above a critical angle  $?c \sim 5^{\circ}$  a fast avalanche occurs, similar to what is expected for classical athermal granular media ; when inclined below this angle, the pile slowly creeps until it becomes flat. The addition of ions in solution screens the repulsive forces between particles, and the flowing properties of the suspension are modified. We observe significant changes in the fast avalanche regime: a time delay appears before the onset of the avalanche and increases with the salt concentration, the whole dynamics becomes slower, and the critical angle ?c increases from ~ 5° to ~ 20°. In contrast, the slow creep regime does not seem to be heavily modified. These behaviors can be explained by considering an increase in both the initial packing fraction of the suspension, and the effective friction between the particles. These observations are confirmed by confocal microscopy measurements to estimate the initial packing fraction of the solution.

#### Friday 12:00 LT 18 (Level 8) / Track 4

SG33

**Fiber orientation-induced strain hardening in uniaxial extensional flow of fiber-filled polymer composites** Thijs R. Egelmeers<sup>1</sup>, Ruth Cardinaels<sup>2</sup>, Patrick D. Anderson<sup>1</sup>, and Nick O. Jaensson<sup>1</sup>

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Fiber-filled polymer composites are important materials for the high-tech industry because they combine important material properties such as increased strength, low weight, and low cost of manufacturing. One big challenge for fiber-filled systems is the prediction of the fiber orientation during processing as this influences various mechanical properties. Understanding the rheology during processing is as important, because the rheology influences the local flow fields, which in turn influence the fiber orientation. In this work, strain-hardening induced by fiber orientation in uniaxial extension of fiber-filled polymer composites is investigated using numerical direct FEM simulations as well as constitutive modelling. The considered fiber-filled systems have a fiber aspect ratio, L/D, up to 10 and a fiber volume fraction up to 10%. The numerical simulations show that the transient uniaxial extensional viscosity is a function of fiber aspect ratio, fiber volume fraction and fiber orientation, which could be accurately described using a constitutive model having only 2 parameters. At higher strains, for increasing fiber volume fractions, the numerical direct FEM simulations show additional strain hardening, which we attribute to a local change in microstructure.

# Friday 12:20 LT 18 (Level 8) / Track 4

Coupling slip and thixotropy to model the transient rheological behaviour of kaolinite suspensions

Jourdain H. Piette, Xiaohan Jia, and Savvas G. Hatzikiriakos

The University Of British Columbia, Vancouver, British Columbia V6T1Z3, Canada

The transient rheological behavior of semi-dilute kaolinite clay suspensions is investigated. Specifically, the flow curve hysteresis and step shear rate tests are used to investigate the shear behaviour of kaolinite suspensions. We find that there is a coupling effect between thixotropy and slip that dominates this transient rheological behaviour. It appears that the onset of solid-like slip (below the yield stress) in the system is a function of wall roughness and interfacial phenomena between the suspension and the wall. The coupled phenomena of slip and thixotropy are investigated using the application of sandpaper, varying the gap, and using different geometries. To illustrate the importance of slip in this system, we propose a model that couples a thixotropic structure parameter model (SPM) to an existing slip model developed in our group.

#### Friday 12:40 LT 18 (Level 8) / Track 4

**Influence of controlled aging on the onset of desiccation cracks in aqueous clay suspensions** <u>Vaibhav Raj S. Parmar<sup>1</sup> and Ranjini Bandyopadhyay<sup>2</sup></u>

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Accumulation of drying-induced stresses in a film of colloidal suspension causes the formation of desiccation cracks. When clay, a constituent of natural soil, is mixed with water, it transforms spontaneously from a viscoelastic liquid to a viscoelastic solid over time. This property, popularly referred to as physical aging, occurs due to the spontaneous and gradual change in electrostatic interparticle interactions during the swelling and breakup of clay particle stacks. In this work, we report the influence of physical aging on the onset of desiccation cracks, i.e., the time corresponding to the appearance of the first crack in a drying aqueous suspension of Laponite clay in a controlled environment. We observe that increasing the interparticle attractive strength results in early crack formation. Additionally, crack onset time is seen to scale with the inverse of the root of the suspension elasticity of the partially dried sample, which is measured using AFM nanoindentation. Finally, we show that our experimental results are supported by a poroelastic model combined with the Griffith criterion for crack formation.

SG32

SG35

**IR14** 

# Symposium IR

# Industrial Rheology, Sustainability and Additive Manufacturing

Organizers: Esther Garcia Tunon, Claire McIlroy and Sylvie Vervoort

#### Friday 8:50 LT 23 (Level 8) / Track 5

Tuneable conductive foamed microstrands through additive manufacturing

Daniele Tammaro<sup>1</sup>, Dimaggio Elisabetta<sup>2</sup>, and Pier Luca Maffettone<sup>1</sup>

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Electrically conductive polymer composites have sparked considerable interest in the research community due to their unique advantages that come from combining regular polymers with the electronic properties of metals or semiconductors in a synergistic manner. Additive manufacturing (AM) offers promising prospects in the realm of conductive polymer composites by allowing for greater design flexibility, more complicated shapes, and rapid manufacturing. Recently, the development of additive manufacturing foams (AMF) offers the possibility to tune locally the electrical percolation for the creation of electronic microchips. Potential applications of such foams by AM in the field of electrically and thermally conductive materials, as well as electromagnetic interference (EMI) shielding materials can be developed by incorporating conductive nanofillers such as carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs) within the porous matrix prior to foaming. In the current work, a small, cost-effective and simple modification of a commercial 3D printer (called, 3D RheoPrinter) was used to measure inline viscosity, extrudate swell and melt fracture of a bio-based and biodegradable electrically conductive polymers (such as polylactic acid (PLA) filled with carbon nanotubes, CN). Using a capillary rheometer approach, the measured pressure and the imposed volume flow rate were used to calculate the shear viscosity through the correction of Weissenberg-Rabinowitsch equation. Moreover, a camera placed at nozzle exit allowed the measurement of extrudate swell and melt fracture that are important for achieving a good resolution in additive manufacturing. The results of the nonlinear rheological characterization for PLA at 180 °C with different amount of CN will be presented. Based on a good knowledge of the rheological nonlinear behavior of the conductive polymers melts, we are able to control the properties of foamed microstrands by tuning the position and orientation of bubbles and the solid conductive particles.

#### Friday 9:10 LT 23 (Level 8) / Track 5

IR15

# New advances in the rheological characterization of 3D printing controlled in-situ microfibrillation of PP/PET composites

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For several decades, blending of polymers has been considered an advantageous and cost-effective method of developing high-performance products. Morphology control of immiscible systems results of great importance to tailor the physical and mechanical properties of the blend. Thus, the well-dispersed and fine spheres in the matrix can increase the impact strength of materials, while the sheet-like dispersed morphology has the ability to enhance the barrier properties of a film, and the micro-fibrils in the blends can increase the uniaxial strength significantly. Recently, the latter, commonly called microfibrillar composites (MFC) has attracted special interest as they permit obtaining in-situ fibrillated morphologies, providing reinforced products with enhanced mechanical performance due to the high level of isotropy. A common technique to produce these MFCs is based on preparing a polymer blend of two semicrystalline materials, which is drawn, either in melted stage (hot drawing) or already solidified (cold drawing). This process will transform the spherical domains into highly oriented micro-fibrils with an increased slenderness. Nevertheless, in recent years, innovative technologies based on Additive Manufacturing, commonly called 3D-printing, have provide the opportunity to obtain MFCs with an absolute control of the fibril morphology. In this work, the potential of 3D printing for obtaining in-situ morphologies achieved. The addition of an appropriate compatibiliser has contributed to the modification phase morphology of the system. An exhaustive conventional and advanced rheological study has made it possible to gain more insight into the MFCs formation, magnitude and evolution of fibre isotropisation.

#### Friday 9:30 LT 23 (Level 8) / Track 5

# Rheological investigation and modeling of healing properties during extrusion-based 3D printing of poly(lactic-acid)

Khalid Lamnawar<sup>1</sup>, Jixiang Li<sup>2</sup>, Xavier Lacambra<sup>3</sup>, and Abderrahim Maazouz<sup>4</sup>

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The focus of the present paper is the rheological study of poly(D,L-lactic-acid) (PDLLA) towards a modeling of their healing properties during 3D direct pellet printing extrusion (DPPE). The viscoelastic properties of PDLLA and the filament temperature during deposition are first characterized. The influence of DPPE processing conditions is investigated in terms of temperature, time, and printing speed. For this, we propose a modeling of the process-induced interphase thickness between two deposited layers considering the non-isothermal polymer relaxation and accounting for the contribution of entanglement rate through the Convective constraint release model. Hence, taking into account the induced chain orientation and mobility coming from filament deposition, this model quantifies the degree of healing between 3D-printed layers. Eventually, the proposed model is validated by comparing the theoretically calculated degree of healing with experimental tensile properties and lap shear results.

### Friday 9:50 LT 23 (Level 8) / Track 5

#### **Rheo-kinetic study of PET chain extension**

Ilaria Cusano<sup>1</sup>, Salvatore Costanzo<sup>1</sup>, Marco Aurilia<sup>2</sup>, and Nino Grizzuti<sup>1</sup>

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The chain extension reaction between polyethylene terephthalate (PET), a polymer from the polyester family, and pyromellitic dianhydride (PMDA), a small multifunctional molecule, was investigated through rheology. The reaction mechanism lies in the joining of the hydroxyl terminal group of the linear PET chain and the anhydride functional group of PMDA, so that a branched 4-arm star, or possibly more complex, multibranched structures are built. Through this procedure, virgin and recycled PET can be successfully adapted to undergo processes such as foaming and film blowing. We prepared different dry mixtures of the two components, with weight percentages of PMDA over PET ranging from 0.15 to 1%. The powder samples thus obtained were loaded between the rheometers' parallel plates and oscillatory transient tests were carried out: the reaction was conducted in situ, in order to evaluate its evolution in terms of complex viscosity over time. The time increase of the viscoelastic properties was a clear indication of the ongoing branching reaction. A fitting procedure was implemented to model the transient results and allowed to estimate also the earliest reaction times, not available in the measurement due to the inevitable high temperature loading operations. This made it possible to evaluate all the key kinetic parameters, such as the characteristic time of the reaction. Finally, such methods resulted in the formulation of an equation for the overall reaction rate of PET-PMDA interaction to describe the branching kinetics.

# Friday 10:10 LT 23 (Level 8) / Track 5

Vitrimers as a potential solution to improve the viscoelastic and mechanical properties of recycled polymers

<u>Alexandru Tudor Boborodea</u><sup>1</sup>, Evelyne van Ruymbeke<sup>1</sup>, Renaud Nicolaÿ<sup>2</sup>, Amandine Guérinot<sup>2</sup>, and Ibrahim Göde<sup>2</sup> <sup>1</sup>IMCN, BSMA, Université catholique de Louvain, Louvain-la-Neuve, Belgium; <sup>2</sup>Chimie Moléculaire, Macromoléculaire, Matériaux, ESPCI Paris, Paris, France

Nowadays, the majority of the objects that surround us are built from virgin polymers, which are ingeniously designed for specific applications. However, once these objects reach their end-of-life, they hold little commercial value and ultimately contribute to the growing problem of plastic waste accumulation. One potential solution to address this pressing environmental issue is mechanical recycling. However, a major drawback of this process is the gradual deterioration of the viscoelastic and mechanical properties of the resulting material. This deterioration occurs for example due to contamination from other polymers or the occurrence of chain scission with each processing cycle. It is therefore imperative to develop innovative approaches that maintain adequate viscoelastic properties throughout the recycling processes.

In this project, the possibility of using vitrimers as additives to enhance the viscoelastic properties of an incompatible polymer blend is explored. Vitrimers are polymeric networks that incorporate dynamic covalent bonds, which can undergo exchange reactions, allowing the network to rearrange itself.

As a first step, we investigate the viscoelastic properties of poly(styrene) (PS) and poly(ethyl hexyl methacrylate) (PEHMA) vitrimer samples of various molar mass and containing different amounts of dioxaborolane cross-linkers. In particular, we use the Time-Temperature-Superposition principle to investigate the influence of temperature on these samples and extract the activation energy of the exchange bond reactions, in function of their composition.

Then, to simulate the contamination that occurs during the recycling of polymers, the properties of PS samples containing low concentrations (up to 20 wt.%) of (PEHMA) are investigated. Due to their immiscibility, these blends display poor mechanical properties. We therefore study the possibility to improve the cohesion of the polymers at the interface by addition of vitrimers, to enhance the mechanical properties of the blends.

IR17

IR16

IR18

# Symposium BL

# **Bio-rheology, Living and Active Matter**

Organizers: Laura Casanellas, Alexander Morozov and Christian Wagner

#### Friday 11:00 LT 23 (Level 8) / Track 5

BL30

# Evaluation of disposable PDMS rheometer geometries for growth and bulk rheology measurements on P. fluorescens biofilms

Vince Engelborghs<sup>1</sup>, Jan Vermant<sup>2</sup>, Steffen Geisel<sup>2</sup>, and Christian Clasen<sup>1</sup>

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Biofilms are bacterial communities of cells enclosed in a matrix of self-produced extracellular polymeric substances. While unwanted in many domains such as wound healing, food production, medical treatments and industrial fouling, biofilms are also growing in importance as a positive tool in biotechnological applications including bioremediation, biofertilization and energy production. Biofilms are viscoelastic in nature, allowing them to adapt to a changing environment, and significantly complicating their mechanical removal.

Understanding the mechanical properties of biofilms is thus a crucial aspect to many applications. However, their rheological characterization is difficult due to the inherent change of the heterogeneous film structure when transferring and loading onto conventional measurement tools. In this work we demonstrate how to adapt conventional rheometry to the study of biofilms. More precisely, the evaluation and use of disposable PDMS geometries where the biofilm can be grown under relevant conditions (flow of nutrients, temperature) is shown [1]. For the geometries, custom-made 3D-printed molds are used to prepare biocompatible and oxygen-permeable PDMS plates. The setup developed here allows to grow biofilm in situ, directly on the PDMS geometry, eliminating the need to scrape and transfer the biofilm. Furthermore, the setup allows to feed the biofilm with nutrients via a channel structure integrated into the PDMS geometries, allowing to obtain long-term, time-resolved information of the rheological properties and the role of nutrient conditions.

[1] Geisel, S., Secchi, E., & Vermant, J. (2022). Experimental challenges in determining the rheological properties of bacterial biofilms. *Interface Focus*, *12*(6), 20220032.

#### Friday 11:20 LT 23 (Level 8) / Track 5

Control of bacteria turbulence through surfaces

Benjamin Perez-Estay<sup>1</sup>, Anke Lindner<sup>1</sup>, Eric Clement<sup>1</sup>, Alexander Morozov<sup>2</sup>, Wilson C. Poon<sup>2</sup>, Vincent Martinez<sup>2</sup>, Jochen Arlt<sup>2</sup>, and Carine Douarche<sup>3</sup>

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Hydrodynamic instabilities appear in E. coli suspensions at high enough concentrations. Controlling such instabilities could, for example, allow extracting energy at the micron scale. We achieved control of the collective motion length scale in a sample confined between two parallel solid surfaces at a distance of H. By measuring the velocity correlation function in the center of the sample, we determined that the decay length scales increase linearly with H up to 800  $\mu$ m. At low confinements and densities, we have also observed a single vortex state spanning an entire centimeter. This vortex is stable for some minutes, so we can observe it by scanning the whole sample. These results show that controlling the size of the collective motion is possible even at larger scales, revealing the importance of surface effects in the properties of the bulk in the active suspension.

#### Friday 11:40 LT 23 (Level 8) / Track 5

#### Anomalous bacterial transport in confined geometries

Peixin Zhang<sup>1</sup>, Malihe Ghodrat<sup>2</sup>, Ignacio Pagonabarraga<sup>3</sup>, Eric Clement<sup>4</sup>, and Anke Lindner<sup>5</sup>

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Bacteria are known to respond to shear flows and display enhanced upstream swimming by the presence of edges. Statistically, the combination of hydrodynamic interactions and flow-induced orientation leads to a strong density increase near the surface. In particular, a previous study reported a non-homogeneous density distribution of E. coli in the presence of a constriction, where they accumulate downstream under flow.

To elucidate the combined role of flow and the presence of surfaces, here we developed experimental devices suited to observe individual trajectories and assess the emerging dispersion processes with single and periodic constrictions using motile E.coli bacteria. Moreover, a Brownian dynamics model for ellipsoidal self-propelling particles is introduced in the simulation, clarifying the mechanisms underlying the experimental observations. By meticulously regulating a low and stable flow field while reducing the roughness of the confining edges, we found in the single constriction system:(1) an extremely strong bacteria accumulation downstream occurring in a range of shear rates,(2) such accumulation effect

BL31

BL32

persisting within the surface layer but disappearing in bulk,(3) a very large spatial extend of such effect reaching equilibrium after a given time. In the multi-constriction system, E.coli still move upstream along the edges and detach downstream of the constriction where they are then retained displaying butterfly-like trajectories. However, pronounced bacterial densification occurs only at the last constriction after a short period. It might indicate that bacterial transport approaching the constriction from downstream takes place over very long distances and is interrupted by the periodic constriction array. Additionally, reaching an equilibrium state might require a longer time.

This work aims to identify the optimal conditions for regulating and amplifying the anomalous transport phenomenon and to understand the interactions of E.coli with geometrically complex surfaces under flow.

### Friday 12:00 LT 23 (Level 8) / Track 5

#### A hybrid computational approach for swimming in shear-thinning and viscoelastic fluids Cara V. Neal<sup>1</sup>, Rachel N. Bearon<sup>1</sup>, and Rachel N. Bearon<sup>2</sup>

<sup>1</sup>University of Liverpool, Liverpool, United Kingdom; <sup>2</sup>King's College London, London, United Kingdom

The locomotion of microswimmers in non-Newtonian fluids is of crucial importance in many biological processes including infection, fertilisation, and biofilm formation. The behaviour of microswimmers in these media remains an area with many conflicting results, with swimmers displaying varying responses depending on their precise morphology, propulsive mechanisms, and elastic properties, as well as the complex characteristics of the surrounding fluid. We numerically investigate the effect of multiple non-Newtonian fluid properties on microswimmer propulsion. Firstly, we explore the influence of shear-thinning rheology on planar sperm-like swimmers. This is achieved through a novel hybrid computational approach that utilises known Newtonian solution techniques (The Method of Regularised Stokeslets, Cortez 2001) to approximate the rapidly varying flow surrounding a swimmer, with a non-Newtonian correction term obtained through solving using the finite element method. Modelling also the elasticity of the flagellum, we demonstrate that shear-thinning rheology can either enhance or hinder propulsion of sperm cells, depending on a balance of elastic and viscous forces as well as fluid properties. Following this, we outline how the hybrid approach can further be used to solve swimming problems in fluids exhibiting both shear-thinning and viscoelastic properties. We investigate how both fluid effects influence the swimming speed and efficiency of a simple conceptual three-sphere swimmer and discuss extensions of this model for problems involving helical bacterial locomotion.

#### Friday 12:20 LT 23 (Level 8) / Track 5

# Enhanced upstream swimming of bacteria in non-Newtonian fluids

#### Arnold J. Mathijssen

#### University of Pennsylvania, Philadelphia, PA 19104, United States

Microbial contamination and infection dynamics are aggravated by the ability of bacteria to swim upstream. While most microorganisms live in polymer solutions including mucus or biofilm biofilm precursors, this motility against flows has not been studied in such macromolecular suspensions until now. Here, using 3D bacterial tracking experiments and hydrodynamic simulations, we find that bacterial upstream swimming is enhanced in complex fluids compared to Newtonian media. These results are explained by a rheotactic torque due to shear-thinning and a flagellar lift force due to viscoelasticity, which together increase the alignment of cells against the flow direction. Yet, upstream motion can be inhibited at large polymer concentrations or by increasing the viscosity with Newtonian polymers. Besides their importance for controlling the spread of diseases, these findings also shed light on the efficacy of spermatozoa for fertility, and the upstream transport of cargo by microrobots in complex flow environments.

#### Friday 12:40 LT 23 (Level 8) / Track 5

#### Active Fluidification of Entangled Polymers by Loop Extrusion

Filippo Conforto, Yair Augusto Gutierrez Fosado, and Davide Michieletto

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Loop extrusion is one of the main processes shaping chromosome organisation and folding across the cell cycle. Its role in regulating DNA entanglement and effective nucleoplasm viscoelasticty remains broadly overlooked. Here we simulate entangled solutions of linear polymers under the action of generic Loop Extruding Factors (LEF) with a model that fully accounts for topological constraints and LEF-DNA uncrossability. We discover that LEFs drive the formation of bottle-brush-like structures which significantly lower the entanglement and effective viscosity of the system. Interestingly, this active fluidification displays a weak optimum at one LEF every 300-3000 basepairs. Finally, in marked contrast with the usual cubic scaling of linear chains, we observe a linear scaling of the effective viscosity with polymer length yielding up to 1000-fold fluidification. Despite this, the scaling of the two-point mean squared displacement often measured in experiments appears insensitive on the presence of an active extrusion process. We argue that our results illuminate how loop extrusion contributes to actively modulate genome entanglement and viscoelasticity in vivo and may guide the interpretation of future experiments.

BL33

BL35

**BL34**
#### Symposium SV

#### Soft Solids and Viscoplastic Fluids

Organizers: Maria Charalambides, Maziyar Jalaal and Ian Wilson

#### Friday 8:50 LT 11 (Level 10) / Track 6

SV20

#### Highly deformable magnetic semi-interpenetrating hydrogels based on acrylamide and natural biopolymers

<u>Alberto Leon-Cecilla</u><sup>1</sup>, Cristina Gila-Vilchez<sup>1</sup>, Francisco J. Vazquez-Perez<sup>1</sup>, Luis F. Capitan-Valley<sup>2</sup>, Vanesa Martos<sup>3</sup>, Maria D. Fernandez-Ramos<sup>2</sup>, Luis Álvarez de Cienfuegos<sup>4</sup>, Antonio L. Medina-Castillo<sup>2</sup>, and Modesto T. Lopez-Lopez<sup>1</sup>

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New materials that can respond to different stimuli and have a high toughness and reliability has been demanded by the growing field of soft robots and actuators. The materials that meet these requirements are hydrogels, due to their soft consistency, natural response to certain stimuli and the possibility of incorporating different inclusions in their polymeric matrix. Among the wide range of hydrogels, interpenetrating and semiinterpenetrating polymer networks stand out due to their multi-responsiveness and high toughness. Therefore, different magnetic semiinterpenetrating hydrogels have been developed in this work. These materials were based on acrylamide (chemically cross-linked) and a natural biopolymer, such as alginate or cellulose. The addition of iron microparticles made the hydrogels responsive to an external magnetic field, but it affected the physical properties of the original polymeric matrix. These magnetic semi-interpenetrating hydrogels were studied under shear, tensile and compressive stresses to mechanically characterize their behavior regarding the concentration of magnetic particles and the used biopolymer. In these studies, the hydrogels showed low values of elastic moduli, while they maintained a high deformability (stretchability up to 220 % and compressibility up to 98%). Furthermore, they showed a good magnetic response that translated into a desired actuation under an external magnetic field. Thanks to their mechanical and magnetic properties, these hydrogels were applied as possible valves for liquid or gaseous systems as well as an oxygen sensor. The possible applications of these materials are numerous thanks to their multi-responsiveness, high toughness and adaptability to different operational circumstances.

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#### Friday 9:10 LT 11 (Level 10) / Track 6

### Exploring the complex dynamics of buoyant jets: from Newtonian to viscoplastic fluids

Hossein Hassanzadeh and Seyed Mohammad Taghavi

Department of Chemical Engineering, Laval University, Quebec, QC G1V 0A6, Canada

We report on the flow dynamics observed in a miscible buoyant jet entering a stagnant ambient of either a Newtonian or a viscoplastic fluid. We employ a combination of different non-intrusive experimental techniques, i.e., time-resolved tomographic particle image velocimetry (TR-Tomo PIV), planar laser-induced fluorescence (PLIF), ultrasound Doppler velocimetry (UDV), and high-speed imaging, to comprehensively analyze the effects of the injection velocity, the density difference, the viscosity ratio, and the rheological features (particularly the yield stress) on the jet flow behaviour. To quantify our buoyant jet behaviour, we define various jet characteristics, including the laminar length, the jet radius, the fluctuating kinetic energy, and the flow intermittency. Based on our findings, increasing the injection velocity results in a decrease in the laminar length. Besides, the density difference contributes to increasing the laminar length. In addition, an increase in viscosity ratio generally causes a decrease in the jet radius. The yield stress of the ambient fluid enhances the flow intermittency at the jet centerline and is also able to completely change the fluctuating kinetic energy behaviour. Finally, we succeed in classifying different jet flow regimes versus the governing dimensionless numbers.

#### Friday 9:30 LT 11 (Level 10) / Track 6

Viscoplastic flows over topographies: an experimental – numerical cross-comparison

Colin Ginot<sup>1</sup>, <u>Guillaume Chambon</u><sup>1</sup>, Maxime Wallon<sup>2</sup>, Paul Vigneaux<sup>2</sup>, and Pierre Philippe<sup>3</sup> <sup>1</sup>Univ. Grenoble Alpes, INRAE, IGE, Grenoble, France; <sup>2</sup>Univ. Picardie Jules Verne, CNRS, LAMFA, Amiens, France; <sup>3</sup>Aix-Marseille Univ., INRAE, RECOVER, Aix-en-Provence, France

Numerous industrial and geophysical applications involve free-surface flows of viscoplastic materials over topographies. The interplay between inertia, rheological effects and topographical features can give rise to particular flow patterns, such as the formation of possibly unyielded accumulation zones ahead of obstacles, the formation of dry zones downstream of obstacles, etc. We report on laboratory experiments specifically designed to study these complex processes. Finite volumes of a viscoplastic material (Carbopol) are released from a rectangular reservoir onto 3D-printed topographies characterized by different configurations of ridges and mounds. The evolution of flow thickness over time is monitored with a temporal resolution of 250 Hz and a typical accuracy of 0.5 mm through a Moiré projection technique. The influence of release position, release volume, and fluid rheology on flow dynamics are investigated. In all case, an abrupt transition is observed between an inertia-dominated regime at short times, and a regime mainly controlled by plasticity and rheological effects at longer times. Interactions with the topography affect this transition by either promoting or delaying the deceleration of the flow. Experimental results are then compared to numerical simulations based

**SV21** 

**SV22** 

on depth-averaged shallow-flow models. In this approach, which is widely used for practical applications, the vertical structure of the flow is not resolved, such that properly representing complex rheological effects can be challenging. Different formulations proposed in the literature for accounting for the internal shearing of the flow and the friction with the bed surface have been tested. Systematic cross-comparisons with the experimental data provides a unique benchmark to assess the predictive capabilities of these models. Once validated, the models can then be used to further explore the dynamical properties of the flows in a wider range of parameters.

#### Friday 9:50 LT 11 (Level 10) / Track 6

#### Non-axisymmetric patterns in floating viscoplastic films

<u>Neil J. Balmforth<sup>1</sup> and Thomasina V. Ball<sup>2</sup></u>

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Expanding cylinders of shear-thinning fluids can become non-axisymmetric due to a linear instability driven by non-Newtonian hoop stresses acting at the advancing fluid edge (Sayag & Worster, 2019). This instability carries over to radially spreading films of the same material when that fluid floats on a deeper bath of less viscous, but denser, Newtonian fluid. In this talk, I will present experiments exploring the instability, in which spreading films of aqueous suspensions of Xanthan gum and Carbopol are floated on a bath of perfluorinated oil. The experimental observations are compared against theoretical predictions from a shallow film model in which viscoplastic rheology is captured by the Herschel-Bulkley constitutive law (Ball & Balmforth 2021). The original instability of Sayag & Worster is modified in the geometry of a thinning expanding film, with shear-thinning no longer driving a loss of axisymmetry at early times (when the degree of expansion is small). Instead, at later times when the film has expanded in radius by a factor of two or so, shear thinning hoop stresses do come into play to drive non-axisymmetric instabilities. Unstable modes possess relatively low angular wavenumber, and the loss of symmetry is not particularly dramatic. When the oil in the bath is replaced by salt solution, however, the experiments are completely different, with dramatic non-axisymmetric patterns appearing. These patterns have their origin in interfacial effects, rather than Sayag & Worster's instability, with the expanding film appearing to fracture under extensional stresses at the fluid edge, prompted by a reduction in fracture toughness in the presence of water (the film's solvent).

#### Friday 10:10 LT 11 (Level 10) / Track 6

#### Formulation of Yield-Stress Fluids for Direct Ink Writing

Alexie Mattei<sup>1</sup>, Jean-Noël Tourvieille<sup>2</sup>, and Jacques Leng<sup>1</sup>

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Direct Ink Writing (DIW) is a 3D-printing technique in which inks based on soft matter are extruded to form materials that may offer a hierarchy of length scales and numerous functionalities. These two characteristics are made possible by the formulation of inks that incorporate polymers, nanomaterials, micromaterials, etc. (Truby & Lewis, 2016)

Here, we focus on inks dedicated to the fabrication of composite polymers whose conductivity can be controlled by adding fillers. We target yieldstress inks that benefit from shear thinning to facilitate flow and yield stress to overcome gravity after deposition. The formulations incorporate a minimal set of ingredients: a thermoplastic polymer powder (e.g., PA, PMMA), a filler that can modify some properties of the composite - the end product of thermal annealing of the 3D printed architecture -, a Carbopol microgel that both confers mechanical strength to the ink and prevents powder settling, and possible other additives such as surfactants. The formulation has to be optimized so that it can be 3D printed *with high fidelity* (M'Barki, 2017) upon extrusion, while also allowing thermal annealing of the material with minimal impact on the degradation of additives during heating.

In this work, we first focus on the rheological properties of the microgel by systematically studying the impact of its grade and of the preparation procedure (Dinkgreve, 2018). We then describe how the addition of powder and additives changes the rheology of the ink and identify at least one grade of Carbopol that is resilient to formulation and can accommodate powders, fillers and additives. Finally, we construct a map of the rheological properties of the blends and relate them to the printability defined in terms of yield stress, surface forces, weight, etc. of printed reference structures.

#### Friday 11:00 LT 11 (Level 10) / Track 6

#### Merging filaments of elastoviscoplastic fluids

Hugo L. França and Maziyar Jalaal

#### Van Der Waals-Zeeman Institute, University Of Amsterdam, Amsterdam, The Netherlands

In this work we study the coalescence dynamics and final shape of two elastoviscoplastic (EVP) filaments. These non-Newtonian materials flow like a viscoelastic liquid above the yield stress and viscoelastic solid below it. Upon touching each other, these filaments flow under surface tension forces with a bridge of increasing height being formed between them. Eventually, the bridged filaments reach a final equilibrium shape when the internal stresses are below the resisting rheological stresses. Through numerical simulations, we systematically study this process by using the Saramito model to represent the EVP behaviour of this material and present a preliminary comparison with experiments. The process studied here finds applications in coating and 3D printing, where complex fluids such as paints, thermoplastic filaments, or bio-inks are deposited side-by-side onto surfaces.

SV23

**SV24** 

SV25

Friday 11:20 LT 11 (Level 10) / Track 6

Spreading of yield stress fluids under vertical vibration

<u>Alice Woodbridge<sup>1</sup></u>, Claudio Pereira da Fonte<sup>2</sup>, and Anne Juel<sup>1</sup>

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Yield stress fluids are ubiquitous in our daily lives. Although model fluids can be accurately characterised in a rheometer, industries which process yield stress fluids, including personal care, construction, and foods, would benefit from low-cost rheological tests which can handle industrial fluids accurately. In this work, we aim to determine which rheological information can be extracted from a vibrated drop of yield stress fluid beyond its fluidisation threshold. Initial experiments are performed using Carbopol as a model yield stress fluid. We deposit a sessile drop of Carbopol on a layer of the same fluid and allow it to relax under gravity to its yield threshold. When the substrate supporting the drop is set into vertical oscillatory motion, one would expect the droplet to spread rapidly upon forcing. Instead, we observe a 'creep' regime where the droplet only reduces its height by <10% up to a threshold acceleration which is proportional to the yield stress [1]. Beyond this threshold the spreading increases significantly. We then compare the behaviour of our Carbopol microgel to that of an emulsion and a suspension. We employ a combination of flow visualisation techniques and simulations to elucidate the mechanism of fluidisation and predict the fluidisation threshold as a function of the method used to deposit the droplet.

[1] Garg, A., Bergemann, N., Smith, B., Heil, M., and Juel, A. (2021). "Fluidisation of yield stress fluids under vibration". In: Journal of Non-Newtonian Fluids Mechanics 294, p. 104595.

#### Friday 11:40 LT 11 (Level 10) / Track 6

#### Interaction of two viscous drops sedimenting in elastoviscoplastic materials

Giancarlo Esposito, Yannis Dimakopoulos, and John Tsamopoulos

Department of Chemical Engineering, University of Patras, Patras, Greece

Multiphase systems composed of a liquid and another immiscible intrusion are found in several formulation industries, ranging from the food sector to the pharmaceutical and construction ones. The interaction between the two phases plays a critical role in the stability of emulsions and significantly impacts on the mechanical properties of the final product. In this work, we perform a computational study to investigate the buoyancy driven motion of a tandem of viscous drops sedimenting in a material which exhibits elasto-viscoplasticity. We model the continuous phase via the Saramito Herschel-Bulkley constitutive equation (Saramito 2009). To solve the mass and momentum balances, we employ a Finite Volume formulation coupled with a Volume of Fluid method to capture the interface between the two phases. We validate our numerical setup via comparison with previous experimental studies (Holenberg, Lavrenteva, and Nir 2011), finding satisfactory agreement in terms of terminal shapes and velocities. We are able to capture the existence of a critical initial distance below which the pair coalesces and above which it separates, as previously reported in experiments. In the case of unequal drops, our results show that the pair might reach a steady separation depending on the initial distance, through an interplay of viscoelasticity, buoyancy and shear thinning. This effect has been previously reported for spheres sedimenting in viscoelastic solutions (Bot, Hulsen, and Van Den Brule 1998) and bubbles rising in elastoviscoplastic materials (Kordalis et al. 2023). We propose a physical mechanism that elucidate such dynamics and present a quantitative relation that correlates the critical distance with the geometrical properties.

Acknowledgement: This work has been financially supported by the European Union's Horizon 2020 research and innovation program under the Marie Sklodowska-Curie Grant Agreement N°955605.

### Friday 12:00 LT 11 (Level 10) / Track 6

#### Stability of bubbles in a yield-stress fluid: from individual to multiple bubbles

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In this work, we study the mechanics of bubbles in a yield-stress fluid. This setting can be found in a wide range of industrial and natural processes including ponds, flooded soils, terrestrial sediments, and other practical fluid/paste storages. In almost all cases, Carbon-dioxide and Methane gas bubbles are emerged due to the microbial degradation of an organic matter. Studying the gas dynamics (e.g. entrapment and release scenarios) within these matrices helps engineering the system advantageously to mitigate the GHG emissions. In these applications, the fluid entrapping the bubbles behaves like yield stress fluids with time-dependent rheology. This fact raises questions regarding the stability of bubbles that are trapped in a yield-stress fluid: here we try to address these questions through a series of experiments and numerical simulations. A vacuum chamber system equipped with a visualization unit was used to control the size of the bubbles trapped in the fluid and determine their sizes and shapes at the onset of motion. Numerical simulations using adaptive finite element method based on augmented Lagrangian scheme was also used to study the yielding mechanism around the bubbles. First, we focus on the stability of a single bubble in a yield-stress fluid that can be considered as the simplest idealization of the entrapment/release scenario. Yield number of the bubbles at the onset of motion with the rheology of the material. Next, we extend our study by looking at more practical scenarios, i.e. multiple bubbles at different orientations and separation distances. Our objective is to understand how the stress fields of neighbouring bubbles interfere with each other and how this interaction affects their onset of motion.

**SV27** 

SV28

#### Friday 12:20 LT 11 (Level 10) / Track 6

#### Dynamics of bubbles evading stasis in elastic yield stress materials

Athanasios Kordalis<sup>1</sup>, Yannis Dimakopoulos<sup>2</sup>, and John Tsamopoulos<sup>3</sup>

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Yield stress (YS) materials have the innate property of both solid and fluid behavior, entrapping bodies whose buoyancy doesn't outbalance the yield force. Bubbles entrapped in YS materials concern the industry since they affect the quality of the final product. Up to now, researchers have modeled the surrounding medium as viscoplastic fluid to study bubble entrapment. However, YS fluids often exhibit finite elasticity, i.e. the unyielded material deforms prior to yielding. In this study we investigate numerically the transient onset of motion of an already trapped bubble in an elastic YS material by ramping down the ambient pressure. The assumed isothermal gas expands, hence buoyancy increases, and beyond a critical volume it starts rising in the medium. The matrix is modelled using the Saramito/Herschel-Bulkley constitutive equation. We solve the governing equations assuming axial symmetry with the stabilized finite element formulation by Varchanis et al. PEGAFEM-V [1]. We have a fine agreement of the predicted shapes and the onset of motion with respective experimental results [2]. The cavity expansion yields the surrounding material initially around the poles as the bubble tries to move [3]. The front and back yielded areas gradually grow and eventually merge initiating the bubble rise. The plastic behavior prevails and prolate bubble shapes appear in accordance with the experiment, similar to results in viscoplastic fluids. Finally, we subject the system to additional pressure protocols and assess the response based on the rheological characteristics of the material.

ACKNOWLEDGEMENT This work has been supported by the Hellenic Foundation of Research and Innovation under the grant HFRI-FM17-2309 MOFLOWMAT and by the European Union's Horizon 2020 research and innovation program under the Marie Sklodowska-Curie Grant Agreement N°955605.

[1] Varchanis et al., J. NonNewton. Fluid Mech. 284, 104365 (2020) [2] Daneshi and Frigaard, J. Fluid Mech. 957, A16 (2023) [3] Kordalis et al., Phys. Rev. Fluids 8, 083301 (2023)

### Friday 12:40 LT 11 (Level 10) / Track 6 SV30

#### Flow cessation during the yielding transition of soft jammed materials

H a Vinutha and Emanuela Del Gado

Department of Physics, Georgetown University, Washington, DC, United States

Dense soft solids under shear deformation lose their rigidity and begin to exhibit plastic flow beyond a threshold strain called yield strain. The yield strain value depends on the shear rate and sample preparation protocol. These soft solids can also experience a stress overshoot that is associated with the formation of shear bands before establishing a steady-flowing state. Here, we investigate the evolution of a model soft solid using flow cessation simulations as it undergoes the yielding transition. The flow cessation tests give access to the stress fluctuations due to the non-affine particle motion and jammed microstructure of the sheared state. Preliminary findings suggest that the stress relaxation timescale and residual stress exhibit non-monotonic behavior around the yield strain, upon flow cessation. Through microscopic analysis of particle stresses, dynamics, and structure, we aim to identify the underlying stress relaxation mechanisms and precursors to the yielding transition. Our work has a potentially significant impact on the manipulation and processing of soft jammed materials in various industries, such as 3D printing.

### Symposium EM

#### **Experimental Methods and New Rheometric Techniques**

Organizers: Christian Clasen, Dan Curtis and Manlio Tassieri

Friday 8:50 LT 19 (Level 10) / Track 7

EM16

## Simple way to measure extensional stress growth coefficient up to a Hencky Strain of e= 8, introducing 2D-UXF method

Masood Khabazian Esfahani<sup>1</sup>, Max G. Schussmann<sup>1</sup>, José A. Rodríguez Agudo<sup>2</sup>, Jan Haeberle<sup>2</sup>, Joerg Laeuger<sup>2</sup>, Christos Georgantopoulos<sup>1</sup>, and Manfred Wilhelm<sup>1</sup>

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Recently, the maximum reachable Hencky strain for the Sentmanat viscosity fixture (SER) [1] is improved by changing the sample placement angle from horizontal positioning (Max. Hencky strain of e=4, equal to linear strain of 55) to tilted positioning (Hencky strain of around e=6, equal to linear strain of 403) [2]. Although this method improves the measurable Hencky strain, it has the same problem as before: The sample eventually overlaps.

A new 2D-UXF method is introduced, in which the upper drum of the universal extensional fixture (UXF) installed on an Anton Paar MCR 702e TwinDrive, moves up in a constant speed. Depending on the sample width, the drum vertical speed is set in a way that the stretching material

doesn't touch itself during the drum rotation. This way, either the material breaks or it stretches to the point at which the sample torque falls below the instrument sensitivity. For a comb-PS (PS290k-60-44, Abbasi 2017 Macromolecules), a maximum Hencky strain of  $\varepsilon$ =8 (equal to linear strain of 2981) is achieved at strain rate of 0.03 s<sup>-1</sup>, (T = 180 °C).

Extensional stress growth coefficient of 2D-UXF method is compared with the data from extensional viscosity fixture (EVF, ARES G2) in horizontal and tilted sample positioning and with data measured on a filament stretching rheometer [3]. The result of 2D-UXF method show a very good agreement with before mentioned methods at low Hencky strain and extend the measuring data to higher Hencky strain.

[1] M.L. Sentmanat, Rheologica Acta (2004), 43(6), 657-669.

[2] D. Parisi, S. Coppola, S. Righi, G. Gagliardi, F. S. Grasso and F. Bacchelli, Rubber Chem. Technol. (2022), 95(2), 241-276.

[3] A. Bach, H.K. Rasmussen and O. Hassager, Journal of Rheology (2003), 47(2), 429-441.

#### Friday 9:10 LT 19 (Level 10) / Track 7

EM17

#### **In-Situ Alignment Measurements during Extensional Flow Using a CaBER-SANS Sample Environment** Matthew A. Wade<sup>1</sup>, Aurel Radulescu<sup>1</sup>, Christian Clasen<sup>2</sup>, and Minne P. Lettinga<sup>3</sup>

<sup>1</sup>JCNS-4, Forschungszentrum Jülich GmbH, Garching, Bravaria 85748, Germany; <sup>2</sup>Chemical Engineering Department, K.U. Leuven, Leuven, Brabant 3001, Belgium; <sup>3</sup>Biomacromolecular Systems and Processes, IBI-4, Forschungszentrum Jülich GmbH, Jülich, North Rhine-Westphalia 52428, Germany

Capillary break-up extensional rheometery (CaBER) has emerged as one of the key techniques to characterize polymer solutions experiencing uniaxial extensional flow<sup>1</sup>. CaBER enables the calculation of apparent extensional viscosities and relaxation times based on capillarity driven thinning or pinching rate of a liquid filament obtained from monitoring the filament shape and diameter evolution in time<sup>1,2</sup>. However, resolving the structural contributions of the polymers that give rise to these macroscopic properties has remained as an outstanding challenge<sup>3</sup>. Based on a novel CaBER device<sup>4</sup>, a CaBER sample environment has been prepared to conduct simultaneous CaBER-small angle neutron/X-ray scattering (SAN/XS) experiments. The instrument is configured such that the beamline, linear motor, and high-speed camera are perpendicular to one another. An essential feature of the setup is a novel CaBER geometry to allow for repeated testing of a single polymer solution sample by limiting effects such as solvent evaporation and inconsistent (re)loading conditions. The effectiveness of this set-up was demonstrated by observing the alignment and relaxation of CPCL NaSAL worm-like micelles obtained from in situ time-resolved SANS during extensional flow. This work sets the groundwork for extensional rheo-SAN/XS measures as a complementary technique to shear based rheo-SAN/XS studies and will serve to provide more insight into materials' properties and behaviour during industrially relevant processes.

<sup>1</sup>S. L. Anna, et al., J. Non-Newtonian Fluid Mech., 87, 2 (1999)

<sup>2</sup> J.P. Plog, et al., Appl. Rheol. 15, 1 (2005)

<sup>3</sup> R. Prabhakar, et al., Phys. Rev. Fluids, 2, 011301 (2017)

<sup>4</sup> J. Van Aeken, et al., Rheol Acta. 61 (2022)

#### Friday 9:30 LT 19 (Level 10) / Track 7

EM18

#### **Exploring the extremes of inkjet ink rheology and jetting performance in industrial printing** <u>Tri R. Tuladhar</u>

#### Trijet Limited, Cambridge CB1 9xQ, United Kingdom

Understanding the rheological characteristics of inkjet fluids is crucial for the overall inkjet process, as they influence various jetting conditions. Traditional oscillatory rheometers face limitations, especially when dealing with inkjet fluids that exhibit non-Newtonian behaviour at elevated frequencies, influencing their jet-ability.

Optimising the bulk properties of inkjet inks based on printhead specifications alone is insufficient for reliable jetting. Ink experiences extreme rheological conditions during the jetting process, such as high frequencies (approximately 100 kHz) and shear rates (around  $10^6$  s<sup>-1</sup>) and high inflight extensional. Slight variations in ink components can profoundly impact the viscoelastic properties during waveform actuation and ejection, affecting fluid flow dynamics, jetting, and breakup behaviour.

Our presentation introduces the TriPAV high-frequency piezo axial rheometer and the TriMaster extensional rheometer, offering a 0.1-10,000 Hz frequency range. These instruments quantify the complex rheological properties of inkjet fluids, closely resembling conditions in the printhead channel and during in-flight printing. Real case examples demonstrate the applications of complex rheological analyses in ink development, quality control, and providing recommendations for optimal printing conditions.

Direct rheological measurements at high frequencies, capable of discerning subtle changes in microseconds, hold immense value across various applications.

#### Friday 9:50 LT 19 (Level 10) / Track 7 Keynote Preliminary results from ROJER-X, an orthogonal imaging extension of the Rayleigh-Ohnesorge Jet

**Extensional Rheometry (ROJER) technique** 

Tobias Threlfall-Holmes<sup>1</sup>, Tobias Threlfall-Holmes<sup>2</sup>, Nik Kapur<sup>3</sup>, Damien Vadillo<sup>4</sup>, Phil Threlfall-Holmes<sup>1</sup>, and Phil Threlfall-Holmes<sup>3</sup>

<sup>1</sup>TH Collaborative Innovation, Liverpool L8 3SD, United Kingdom; <sup>2</sup>The University of Liverpool Maths School, Liverpool L69 7SH. United Kingdom: <sup>3</sup>School of Mechanical Engineering. The University of Leeds. Leeds LS2 9JT. United Kingdom: <sup>4</sup>Corporate Research Analytical Laboratory, 3M, Minneapolis, MN 55144-1000, United States

We report preliminary results with PEO solutions from a commercial realisation of the ROJER-X developed by TH Collaborative Innovation, an enhancement of the Rayleigh-Ohnesorge Jet Extensional Rheometer in which simultaneous orthogonal views of the jet are obtained. We dub this ROJER-X denoting both an eXtended method and the crossed views on the jet. Initially conceived primarily to address the numerous usability challenges that have limited the adoption of ROJER, especially in industrial research laboratories, we have additionally found exciting enhancement in capability, with the instrument specified to measure down to sub-microsecond extensional relaxation times. Whereas conventional capillary thinning techniques (CaBER, DoSER) observe a single fluid bridge, ROJER measures thousands of bridges a second, for many minutes of fluid flow from a syringe. This radically improves the statistics of diameter-decay curve fitting, and fluid sampling of real commercial formulations inhomogeneous on a single liquid bridge lengthscale. Such jets won't remain stable in frame and focus for minutes: ROJER-X makes it practical to reposition and refocus on-the-fly: obtaining a time history of measured rheological parameters quantifying non-homogeneity. The two images of the same bridge can be synchronised, or be in fixed or phase delay from each other and/or the jet resonance driving frequency. This permits (a) cross-checking that it is truly a rheometric flow in 3D and remains in Rayleigh resonance, and of the image-analysis-derived rheological parameters; (b) a direct measure of jet speed, independent from a pump flowrate derivation; (c) a "coarse and fine" delay, simultaneous construction of the filament thinning curve, with more rapid acquisition and less fluid consumption. Since all these operating modes are programmatically controlled through the custom controller, with streamed image acquisition, it also enables the development of real-time analysis, and eventually automatic setup and adjustment of parameters.

#### Friday 11:00 LT 19 (Level 10) / Track 7

Attempts to mitigate edge fracture instability in sheared polymers

**EM20** 

**EM21** 

**EM19** 

Li Benke<sup>1</sup>, Mavromanolakis Antonis<sup>1</sup>, Dimitris Vlassopoulos<sup>1</sup>, and Dimitris Vlassopoulos<sup>2</sup> <sup>1</sup>Institute of Electronic Structure and Laser, Foundation of Research and Technology Hellas, Heraklion, Greece; <sup>2</sup>Department of

Materials Science and Technology, University of Crete, Heraklion, Greece

The measurement of nonlinear shear response of viscoelastic materials in rotational rheometers remains challenging, since it is often hindered by edge fracture instabilities. The phenomenon was first addressed theoretically by Tanner and Keentok and ever since has attracted the interest of rheologists. More recently, Fielding and co-workers proposed a stability criterion based on realistic constitutive equations, and proposed the use of a bathing fluid that reduces the stress gradient between sheared material and the environment as a means to mitigate fracture. This idea was found to be effective for weakly viscoelastic liquids. Here, we focus on strongly viscoelastic materials (polymer melts) and compare different mitigation strategies using cone-and-plate and cone-partitioned plate (CPP) geometries. In addition to bathing fluids we use a specially made outer collar that is attached to the sample's edge. Assessing the different transient signals during measurement, we find that the combination of CPP and collar is the most promising. It is proposed that it may help to measure highly elastic materials over an extended range of shear rates and contribute to reliably measuring the normal stress coefficients.

Friday 11:20 LT 19 (Level 10) / Track 7

Effective mechanical system approach for effects of inertia on the viscoelastic properties Xuesi Gao<sup>1</sup> and Wook Ryol Hwang<sup>2</sup>

<sup>1</sup>School of Mechanical Engineering, Gyeongsang National University, Jinju City, Gyeongsang Namdo 52828, Republic of Korea; <sup>2</sup>School of Mechanical Engineering, Gyeongsang National University, Jinju-si, Gyeongsangnam-do 52528, Republic of Korea

The identification of viscoelastic properties in SAOS tests can be affected by fluid inertia, particularly at high oscillation frequencies. Here, we introduce an effective mechanical system to assess how fluid inertia affects the identification of viscoelastic properties during SAOS test using a parallel plate (PP) geometry. This approach entails the mapping of the viscous (polymer viscosity and solvent viscosity), elastic, and inertial properties of the fluid system onto the damping, elastic, and mass components of the effective mechanical system. By utilizing this method, we have created an analytical framework for characterizing the viscoelastic behavior of fluid systems. The analysis of the frequency response of the modulus, obtained by the effective mechanical system, reveals that the fluid inertia only affects the storage modulus and not the loss modulus. Additionally, we explore the relationship of this storage modulus bias with the gap size, oscillation frequency, and disk radius and observe that it is independent of the disk radius while strongly dependent on the gap size and frequency. To validate our findings, we compared the results with both the analytical solution of the Maxwell model with solvent viscosity and numerical simulations using two linear viscoelastic models (Oldroyd-B model and Giesekus model). This comparison confirms the accuracy of our approach in identifying viscoelastic and inertial properties using the effective mechanical system with a maximum error of 2.7%. Funding

Acknowledgement: This study was supported by the National Research Foundation of Korea (NRF) (No: 2019R1A2C10039741561382116530105).

**EM22** 

**EM23** 

#### Friday 11:40 LT 19 (Level 10) / Track 7

#### **Application of Numerical Differentiation to Creep Ringing**

Jehyeok Choi<sup>1</sup> and Kwang Soo Cho<sup>2</sup>

<sup>1</sup>Department of Polymer Science and Engineering, Kyungpook National University, Daegu, Republic of Korea; <sup>2</sup>Department of Polymer Science and Engineering, Kyungpook National University, Daegu, Republic of Korea

Cho and coworkers invented a numerical method which extracts linear viscoelastic function from creep data with ringing. Their method is to use Laplace transform and the Boltzmann superposition principle. Since creep ringing is originated from that the difference between material stress and machine stress is the inertia force which is proportional to the second order time-derivative of measured strain. We know the machine stress because it is the stress applied by stress-controlled rheometer and we know strain because it is measured. Hence, if we have an effective numerical method for time derivative of experimental data then we can determine the material stress which the material feels. Although the Laplace transform method invented by Kim et al., J. Rheol. (2015) is applicable to only linear regime, the method of numerical differentiation is applicable to nonlinear regime such as LAOS. We will introduce the numerical differentiation method which is stable for experimental errors in this talk.

### Friday 12:00 LT 19 (Level 10) / Track 7

### Development of rheometry based on ultrasonic velocity profiling

Yuji Tasaka<sup>1</sup>, Kohei Ohie<sup>1</sup>, and Taiki Yoshida<sup>2</sup>

<sup>1</sup>Faculty of Engineering, Hokkaido University, Sapporo, Japan; <sup>2</sup>AIST, Tsukuba, Japan

Usual torque rheometer has been sophisticated and now is recognized as a reliable tool to evaluate rheology of many kinds of complex fluids. But the strong assumption of constant shear rate acting on test fluids in a thin layer causes tight restriction on measurable objects. Slip on the surface of sensing part, occurrence of shear banding, elastic instability, and etc. would be a source of critical error on the evaluation. Some kinds of velocity-profiling-assisted rheometry were thus proposed to determine local shear rate in a widened fluid layer. Our research group has advanced the methodology based on ultrasonic velocity profiling as velocimetry, which is capable to opaque and heterogeneous media. Rheological properties can be determined by substituting velocity profile information measured in an oscillating/rotating cylinder filled with a test fluid into an equation of fluid motion with a constitutive equation. The most advantage of the proposed rheometry is quasi-instantaneous evaluation and it can capture transient behavior of complex fluids. We demonstrated it to evaluate transient behavior of shear-thinning viscosity of rice gruel (porridge) under hydrolysis by digestive enzyme. The viscosity curves are approximated by power law and the transient behavior is expressed by variations on parameter space of consistency and power law index.

#### Friday 12:20 LT 19 (Level 10) / Track 7

EM24

**Coupling rheology, DMA, and Raman spectroscopy for the characterization of smart and functional materials** José A. Rodríguez Agudo<sup>1</sup>, Andrés Posada Murcia<sup>1</sup>, Jan Haeberle<sup>1</sup>, Christopher Giehl<sup>1</sup>, Ferdinand Cerbe<sup>2</sup>, Dominik Fauser<sup>3</sup>, Holger Steeb<sup>3</sup>, Gunther Arnold<sup>1</sup>, and Jörg Läuger<sup>1</sup>

<sup>1</sup>Anton Paar Germany, Ostfildern, Germany; <sup>2</sup>Institute of Mechanics and Adaptronics, Technische Universität Braunschweig, Braunschweig 38106, Germany; <sup>3</sup>Institut für Mechanik, Universität Stuttgart, Stuttgart 70569, Germany

Functional polymer materials exhibit precise and controlled responses to external stimuli, with changes occurring at the molecular level and translating into macroscopic behavior. Dynamic mechanical analysis (DMA) is an essential tool for monitoring the mechanical properties of these materials throughout the stimulation process. In this context, combined torsional-axial rheometers with both rotational and linear drives have significantly advanced the characterization of functional polymer materials [1]-[4].

This contribution showcases applications of advanced rheometric techniques in the characterization of smart materials, including bio-based shape memory polymers (SMPs), metallopolymers, or liquid crystal elastomers (LCEs). Complementing rheology and DMA with Raman spectroscopy allows correlation of changes in material morphology with related changes in macroscopic mechanical properties. In-situ Raman measurements during DMA reveal morphological changes such as crystallization in SMPs like PLA or liquid crystal mesogen orientation in LCEs. These morphological changes induce anisotropic macroscopic structural changes that can be effectively quantified using combined torsional-axial DMA [4]. We introduce a novel dynamic parameter, ?, derived from the phase shift angles in torsion ( $d_T$ ) and extension ( $d_E$ ), which offers a new perspective on viscoelasticity quantification for anisotropic structures as found in functional or smart materials.

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[2] D. Fauser and H. Steeb, J. Mater. Sci., 57, 20, 9508-9524 (2022)

[3] P. Mainik, L.-Y. Hsu, C. W. Zimmer, D. Fauser, H. Steeb, and E. Blasco, Adv. Mater. Technol., 8, 23, 2300727 (2023)

[4] J. R. Agudo, J. Haeberle, M. Müller-Pabel, A. Troiss, A. Shetty, J. Kaschta and C. Giehl, J. Rheol., 67, 6, 1221-1250 (2023)

#### Friday 12:40 LT 19 (Level 10) / Track 7

**EM25** 

### Rheology of water-based methylcellulose systems: a data-driven approach with Gaussian Process Regression Isaac Y. Miranda-Valdez, Tero Mäkinen, Juha Koivisto, and Mikko J. Alava

Department of Applied Physics, Aalto University, Espoo, Finland

Gaussian Process Regression (GPR) is a machine learning technique that uses Bayesian inference to model a target function as a Gaussian process. In rheology, GPR can be used to model the rheological behavior of complex viscoelastic materials by conditioning a random function on the experimental data. This approach combines prior knowledge with likelihood functions based on a training dataset, allowing for a probabilistic

prediction of rheological material functions. This study focused on the rheology of water-based methylcellulose systems, which undergo thermally induced gelation. We evaluated their oscillatory viscoelastic response during dynamic mechanical analysis (DMA) at multiple temperature steps, thus characterizing their rheological behavior during the liquid-to-gel and gel-to-liquid transitions. Our findings demonstrate that GPR is an effective tool for analyzing rheological data and discovering time-temperature (and concentration) relationships in viscoelastic materials. In fact, GPR described the thermoreversible gelation process of water-based methylcellulose systems; the approach showed that methylcellulose exhibits a frequency-dependent behavior that gives rise, for example, to a butterfly-shaped thermal hysteresis in the loss modulus function. The data-driven approach provides a framework to capture the rheology of complex systems, whose viscoelastic response is conditioned to multiple factors. Overall, our study highlights the potential of GPR as a valuable tool in rheology and remarks on its potential applications in other viscoelastic systems.

### Symposium NF

#### Non-Newtonian Fluid Mechanics and Flow Instabilities

Organizers: Marco Ellero, Stylianos Varchanis and Helen Wilson

Friday 8:50 LT 24 (Level 10) / Track 8

Similarity theory for turbulent planar jets of viscoelastic fluids

Mateus C. Guimarães<sup>1</sup>, Fernando T. Pinho<sup>2</sup>, and Carlos B. da Silva<sup>1</sup>

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A similarity analysis of the Reynolds-averaged governing equations for the mean conformation tensor (Cij) is carried out for turbulent planar jet flows of viscoelastic solutions described by the FENE-P model allowing the development of a theory for the evolution of components of Cij in the far-field. Numerical results used to assess the theory were obtained through accurate DNS at different Reynolds (Re) and Weissenberg numbers (Wi), solvent to total viscosity ratios (B) and maximum polymer extensibilities (L). Downstream the near-field region, where the jet transitions from laminar to turbulent flow and turbulence becomes fully-developed, the far-field is composed of two main regions of viscoelastic effects, the highly elastic region, where polymer are highly stretched and viscoelastic effects are strong, and the nearly-coiled regime of low elasticity. In the highly elastic region the total rate of dissipation of turbulent kinetic energy (k) is predominantly by the polymer and the solvent dissipation reduction (SDR) attains a maximum independent of Re, Wi, ß and L. Polymers are highly stretched initially in anisotropic and subsequently in nearly-isotropic configurations. In the nearly-coiled region, the polymer configuration is nearly-isotropic as the flow proceeds to the low elasticity region and subsequently viscoelasticity goes into a final region of decay. In both nearly-coiled sub-regions polymer stretching is predominantly imposed by approximately isotropic intermediate and small-scale turbulence. In both nearly-coiled sub-regions the solvent and polymer contribute to the total rate of dissipation of k, with the polymer essentially assuming a Newtonian-like dissipative role proportional to its zero-shear-rate viscosity. Here we find the lowest value of SDR, linearly proportional to 1-β, but independent of Wi and Re. In each sub-region the mechanisms leading to Cij are identified and theoretical reasoning produces normalized scaling laws, which are valid regardless of Wi and are confirmed by DNS data.

Friday 9:10 LT 24 (Level 10) / Track 8

NF31

NF30

## Exploring transitional pathway in von Karman swirling flow of polymer solutions: from laminar to elastic turbulence

<u>Xiaoxiao Yang</u><sup>1</sup>, Darius Marin<sup>1</sup>, Charlotte Py<sup>1</sup>, Anke Lindner<sup>2</sup>, and Sandra Lerouge<sup>1</sup> <sup>1</sup>MSC, CNRS UMR 7057, Université Paris Cité, Paris, France; <sup>2</sup>Laboratoire de Physique et Mécanique des Milieux Hétérogènes, CNRS, ESPCI PSL, Sorbonne Université, Université Paris Cité, Paris, Île-de-France 75005, France

In elasticity-driven viscoelastic polymeric fluids, Steinberg's group discovered the existence of elastic turbulence (ET). ET is characterized by increased flow resistance with a hysteresis loop, alongside the excitation of flow across a broad range of frequencies and wavenumbers. Despite significant interest in the characteristics of ET over the past two decades, the exploration of intermediate secondary flow patterns preceding the onset of ET remains sparse. In this work, we study the complete transitional pathway from laminar to fully developed turbulent state in von Kármán swirling flow experimental conditions, very similar to those used in the original studies. Using ramp and step flow protocols under imposed shear rates and shear stresses, we focus on benchmark polymer solutions made of polyacrylamide (PAAm) dissolved in sugar viscous solvent in salted water. Our study reveals a succession of bifurcations, similar to the one reported in more concentrated PAAm solutions. However, fluorescent dye visualization in the (r,z) plane reveals the coexistence of two toroidal vortices in the base flow state, contrasting with the singular elasticity-driven toroidal vortex mentioned in prior literature. This phenomenon features an elasticity-driven peripheral vortex and an inertia-driven central one. The linear increase in flow resistance during the laminar regime correlates with the expansion of the elasticity-and inertia-driven vortices. Persisting into the fully developed ET state, outwardly propagating spiral vortex patterns at large scale can still be found. The explored transition sequence to ET, coupled with the evolution of secondary flow structures, provides critical insights into the underlying mechanisms of elastic instability and the associated cascade of bifurcations.

**NF32** 

#### Friday 9:30 LT 24 (Level 10) / Track 8

Drag reduction of superhydrophobic surfaces with shear-thinning fluids

Linsheng Zhang<sup>1</sup>, Colin R. Crick<sup>2</sup>, Henry Ng<sup>1</sup>, and Robert J. Poole<sup>1</sup>

<sup>1</sup>University of Liverpool, Liverpool L69 3GH, United Kingdom; <sup>2</sup>Queen Mary University of London, London, United Kingdom

Significant research efforts have been dedicated to exploring laminar and turbulent drag reduction using superhydrophobic (SHO) surfaces, particularly in the context of water as the fluid medium. However, the practical transportation of fluids involves a wide range of substances with distinct physical properties. These variations include low surface tension fluids (such as surfactant-contaminated water), elevated-viscosity fluids (such as aqueous ethanol solutions), and complex fluids with shear-thinning properties (such as semi-rigid polymetric solutions). This diversity in fluid characteristics underscores the need for a comprehensive investigation to extend the applicability of SHO surfaces across various industrial scenarios. In a recent publication, we presented results indicating that a decrease in fluid surface tension leads to a reduction in drag reduction/slip length, at a constant fluid viscosity. Meanwhile, an increase in viscosity results in an increased slip length. Experimental results support a linear relationship between slip length with capillary length and viscosity. More recently, our research has progressed to explore the impact of shear-thinning behaviour on drag reduction using SHO surfaces. Drag reduction experiments were conducted in a cone-and-plate rheometric system and dilute/semi-dilute solutions of a semi-rigid polymer - xanthan gum - have been employed as a working fluid. Preliminary results confirmed the effectiveness of laminar drag reduction in shear-thinning fluids. Consistent with the results in our previous work, due to higher viscosity, the slip lengths for xanthan gum solutions are higher than those with water. For the same level of drag reduction, the slip length calculated using a power-law model is higher than that determined assuming a Newtonian fluid at a fixed shear rate. Given the limited experimental work of SHO drag reduction with non-Newtonian fluids, this study aims to contribute insights into the broader drag reduction and rheology community.

#### Friday 9:50 LT 24 (Level 10) / Track 8

NF33

**NF34** 

# Large Scale Direct Numerical Simulation of Turbulent Drag Reduction in Isotropic and Channel Flows Xue-Feng YUAN

Institute for Systems Rheology, Guangzhou University, Guangzhou, Guangdong 510006, China

An OpenFOAM-PETSc based viscoelastic numerical solver has been developed for revealing the multiscale dynamic coupling mechanisms between polymeric chain molecules and the eddies of different sizes in Turbulent Drag Reduction (*TDR*). By using FENE-P and FENE-CD2 constitutive equations for modelling dilute and semi-dilute entangled polymer solutions, large scale direct numerical simulations of *TDR* under isotropic and channel flows in various Reynolds number (*Re*) and Weissenberg number (*Wi*) as well as polymer concentrations and the extensibility of chain molecules have been carried out. The numerical results reproduce the main dynamic features of *TDR* experimentally observed in the literature. The spatiotemporal power spectra of multiple physical field fluctuations, including the kinetic energy, elastic energy, energy dissipation, pressure, the ratio between the normal stress difference to shear stress, polymer conformation, are analyzed and compared to reveal pathways of energy transfer between different dynamic scales. The scaling relations between the exponents of the power-law decays of kinetic and elastic energies, pressure and shear stress fluctuations are also verified. The time-resolved turbulent coherent structures are analyzed by a third generation of vortex identification method and correlated with the polymer conformation field for better understanding polymer and turbulent flow interactions. A machine learning method for super-resolution reconstruction of turbulence is proposed and the validation results are also reported.

#### Friday 10:10 LT 24 (Level 10) / Track 8

## Large scale structures in a drag-reduced turbulent pipe flow with surfactant additives <u>Henry Ng</u> and Robert J. Poole

#### University of Liverpool, Liverpool L69 3GH, United Kingdom

Large scale structures of a drag-reduced turbulent pipe flow are investigated using high-speed stereoscopic particle imaging velocimetry and compared to their Newtonian analogues at matched bulk friction Reynolds numbers. The working fluids are aqueous solutions of cationic surfactant cetyl-trimethyl-ammonium bromide (CTAB) balanced with sodium salicylate at a surfactant/salt ratio of 0.5. Fluid systems with CTAB surfactant concentration up to 800 parts-per-million were tested and turbulent drag reduction of up to 50% achieved at a bulk flow Reynolds number of 35,000. Complementary measurements of Newtonian turbulent pipe flow at matched friction and matched bulk Reynolds numbers facilitated the direct comparison of drag-reduced flows with surfactant additives to their Newtonian counterparts through the lens of both ``matched-drag" and ``matched-mass-flow". Instantaneous fields reveal that the near-wall streaky structure of streamwise-aligned azimuthally-alternating low- and high-momentum zones, the signature of large- and very-large-scale motions (LSM/VLSM), are appreciably elongated in drag-reduced flow but with an attendant reduction of the vortical structures associated with the low-speed streaks. Two-point spatio-temporal velocity correlations yielded three-dimensional structure characteristics revealing average structures up to approx. 80% longer than their Newtonian counterparts in the logarithmic region. Further, it appears that the LSMs and VLSMs, while elongated, are confined to the near-wall region of the pipe and do not penetrate beyond the logarithmic layer into the outer layer.

#### Friday 11:00 LT 24 (Level 10) / Track 8 NF35 Hammering at the entropy: A GENERIC guided approach to learning rheological constitutive equations using PINNs

David Nieto Simavilla<sup>1</sup>, Andrea Bofanti<sup>2</sup>, Imanol Garcia de Beristain<sup>3</sup>, Pep Español<sup>4</sup>, and Marco Ellero<sup>5</sup>

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Applied Mathematics, Bilbao, Spain; <sup>4</sup>Fundamental Physics, UNED, Madrid, Spain; <sup>5</sup>Basque Center for Applied Mathematics - BCAM, BILBAO, BIZKAIA 48009, Spain

We present a GENERIC guided approach using PINNs to determine the entropy leading to the constitutive equation for the stress in rheological models. Our methodology uses the eigenvalue decomposition of the evolution equation of the conformation tensor to instill physical knowledge into a neural network approximation of the real function. We evaluate: 1) the validity of the traditional approach using steady-state rheometric flows to establish rheological models 2) the region of the eigenvalues of the conformation tensor eigenvalues space where the approximated model offered by our PINNs method can be safely extrapolated from the model learned from only steady-state. For 1) we show that transient and complex flows can explore regions of the space away from the steady-state. For 2) we evaluate the error in the model predictions of the entropy and the stress in the region covered by simulations of complex flow around a cylinder at moderate Wi numbers.

### Friday 11:20 LT 24 (Level 10) / Track 8

NF36

**NF37** 

**NF38** 

# A mesh-free framework for high-order simulations of viscoelastic flows in complex geometries Jack King and Steven J. Lind

School of Engineering, University of Manchester, Manchester, United Kingdom

Despite decades of research effort, the determination of accurate viscoelastic flow solutions remains a key challenge in computational rheology, particularly at high Weissenberg numbers and elasticity levels. Pseudo-spectral methods can provide very high levels of accuracy, but are limited to simplified geometries. Methods based on unstructured meshes can handle complex geometries, but with the exception of spectral element methods, are generally limited to second order accuracy. Further, the construction of a high-quality body-fitted mesh can be a complex and time-consuming task. Mesh-free methods provide greater geometric flexibility, but usually only provide low levels of accuracy. In this work we introduce a high-order mesh-free framework for simulations of viscoelastic flows. Spatial accuracy is up to 10th order, and variable resolution is included naturally in the formulation. Alongside direct integration of the conformation tensor evolution equation, we investigate formulations based on the log-conformation and Cholesky decompositions, with which the scheme is capable of providing accurate simulations at high Weissenberg numbers (up to Wi=128). The performance of the method is demonstrated for canonical flows in trivial geometries (e.g. Poiseuille flow, Kolmogorov flow), and more complex flows, including those past periodic cylinder arrays, and representative porous geometries. The ability of the method to capture symmetry breaking and the onset of viscoelastic instabilities is highlighted. This mesh-free approach offers an alternative pathway towards high-fidelity DNS viscoelastic flows and flow instabilities in realistic geometries.

### Friday 11:40 LT 24 (Level 10) / Track 8

#### A Conservative Level-Set Method for Multiphase Viscoelastic Flows

William Doherty<sup>1</sup>, Timothy N. Phillips<sup>2</sup>, and Zhihua Xie<sup>1</sup>

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The development of stable numerical schemes for viscoelastic multiphase flows has implications for many areas of engineering applications. We present a novel conservative level-set framework for simulating multiphase flows accurately and efficiently. The conservative level-set method enables the interface between fluid phases to be defined implicitly and is fully integrated into the mathematical framework of viscoelastic flow. The log-conformation formulation is utilised to enhance stability so that approximations can be obtained for large values of the Weissenberg number. The numerical scheme is validated with reference to several benchmark problems. These include the motion of a gas bubble rising in a viscoelastic and the drop impact problem. In the case of the former flow features such as the discontinuity in the bubble rise velocity at a critical bubble volume, the development of filaments and cusps and the generation of negative wakes is explored. Numerical predictions are compared against experimental measurements and excellent agreement is found.

Friday 12:00 LT 24 (Level 10) / Track 8

### Computational rheology in hierarchical grids: the HiGFlow software

Hugo A. Castillo-Sánchez and Antonio Castelo

Institute of Mathematics and Computer Sciences, University of Sao Paulo, Sao Carlos, Sao Paulo, Brazil

The HiGFlow system (HiG stands for hierarchical grids) is a Computational Fluid Dynamics (CFD) software that was recently developed at the Institute of Mathematics and Computer Sciences (ICMC) from the University of Sao Paulo (USP). This software is able to simulate single and multi-phase flows of both Newtonian and Non-Newtonian fluids using a new finite difference method with meshless interpolations in tree-based hierarchical grids. The system is being developed in a modular way, allowing new techniques and models to be easily implemented. In this talk, I will briefly describe the constitutive equations for complex fluids that have been tested and implemented in HiGFlow (i.e. rheological models for thixotropic-viscoelastic fluids, shear-banding micellar solutions, elastoviscoplastic materials and suspensions of spherical particles with and without shear-thickening behaviour). Numerical simulations of these models were carried out in HiGFlow using different flow geometries (i.e. shear-driven flows, 2D channels, planar-contraction 4:1, contraction-expansion 4:1:4 and lid-driven cavity flows). Our simulations were validated by comparing them with results reported in the literature and with those predicted by other numerical CFD tools, such as the RheoTool system,

the foam-extend software (both of these based on OpenFOAM) and an in-house code that uses the Vorticity-Velocity-Formulation. Lastly, I will present some of our results to show that the HiGFlow system offers flexibility and stability in the numerical simulations of complex fluids to successfully reproduce rheological behaviour of high interest to the rheology community.

#### Friday 12:20 LT 24 (Level 10) / Track 8

NF39

**NF40** 

#### **Smoothed-particle hydrodynamics simulations of integral multi-mode and fractional viscoelastic models** <u>Luca Santelli<sup>1</sup></u>, Adolfo Vázquez-Quesada<sup>2</sup>, and Marco Ellero<sup>1</sup>

<sup>1</sup>Basque Center for Applied Mathematics - BCAM, BILBAO, BIZKAIA 48009, Spain; <sup>2</sup>Facultad de ciencias, Universidad Nacional de Educación a Distancia - UNED, Madrid 28015, Spain

To capture specific characteristics of non-Newtonian fluids, during the past years fractional constitutive models have become increasingly popular. These models are able to capture in a simple and compact way the complex behaviour of viscoelastic materials, such as the change in power-law relaxation pattern during the relaxation process of some materials [1].

Integral models have been proposed more than thirty years ago [2] and are appealing when the material shows a very broad relaxation spectrum, like power-law behaviour. Integrating along a streamline is easier in a Lagrangian framework than an Eulerian one, which requires the flow history to be reconstructed. Therefore, we use a Lagrangian Smoothed--Particle Hydrodynamics (SPH) method [3] to easily track particle history; this allows us to solve integral constitutive models in a novel way that does not rely on complex tasks.

Hence, we develop here a SPH integral viscoelastic method which is first validated for simple Maxwell or Oldroyd-B models under Small Amplitude Oscillatory Shear flows (SAOS). By exploiting the structure of the integral method, a multi-mode Maxwell model is then implemented. The method is furthermore extended to include fractional constitutive models [4], validating the approach by comparing results with theory under SAOS. Finally, simulations for more complex flows will be presented.

[1] Pan Yangb, Yee Cheong Lama, Ke-Qin Zhub, J. Non-Newtonian Fluid Mech.165, 88 (2010).

[2] X.-L. Luo and R. I. Tanner, International J. Numerical Methods in Engineering 25, 9 (1988).

[3] Luca Santelli, Adolfo Vazquez-Quesada, and Marco Ellero, arXiv preprint arXiv:2311.13173 (2023). (Submitted to JNNFM) [4] A. Jaishankar and G. H. McKinley, J. Rheo. 58, 1751 (2014).

#### Friday 12:40 LT 24 (Level 10) / Track 8

Microstructural Smoothed Particle Hydrodynamics Model and Simulations of Discontinuous Shear-Thickening Fluids

Peter Angerman<sup>1</sup>, Sandnes Bjornar<sup>2</sup>, Sagaya S. Prasanna Kumar<sup>3</sup>, Ryohei Seto<sup>4</sup>, and Marco Ellero<sup>5</sup> <sup>1</sup>Complex Fluids Research Group, Department of Chemical Engine, University of Swansea, Swansea, Wales SA1 8EP, United Kingdom; <sup>2</sup>Complex Fluids Research Group, Department of Chemical Engine, University of Swansea, Swansea, Wales SA1 8EP, United Kingdom; <sup>3</sup>Basque Centre for Applied Mathematics, Bilbao, Basuqe, Spain; <sup>4</sup>Wenzhou Key Laboratory of Biomaterials and Engineering, Wenzhou Institute, University of Chinese Academy of Sciences, Wenzhou, Zhejiang 325000, China; <sup>5</sup>Basque Center for Applied Mathematics - BCAM, BILBAO, BIZKAIA 48009, Spain

In this work, we present the first implementation of a microstructural DST model in Smoothed Particle Hydrodynamic (SPH) [1] simulation. The scalar model with a mixed local and non-local evolution was implemented in an SPH scheme and tested in simple shear geometry. Three distinct ratios of local to non-local microstructural effects were probed: weak, moderate, and strong non-locality. Strong non-locality was found to produce boundary driven instability resulting in periodic solutions. This instability was validated with the theoretical framework of Richards et al.[2]. SPH model was found to be in an excellent quantitative agreement and capable of reproducing the stability properties associated with this mechanism. Banding patterns reminiscent of the work of Nakanishi et al. [3] were observed in the case of moderate non-locality. Weak non-locality produced stress-splitting instability, resulting in discontinuous stress fields. The mechanism of the stress-splitting has been explored and contextualised by the interaction of local microstructure evolution, the stress-control scheme and non-local effects. Impact of instabilities on construction of flow curves in simple shear, along with interactions between the instability mechanisms and possibility of aperiodic solutions, is discussed.

Peter Angerman, Sagaya Prasanna Kumar, Bjornar Sandnes, Ryohei Seto, and Marco Ellero. Microstructural smoothed particle hydrodynamics model and simulations of discontinuous shear-thickening fluids, 2023

James Richards, John Royer, B. Liebchen, Ben Guy, and Wilson Poon. Competing timescales lead to oscillations in shear-thickening suspensions. Physical Review Letters, 123, 07 2019. doi: 10.1103/Phys-RevLett.123.038004

Hiizu Nakanishi, Shin-ichiro Nagahiro, and Namiko Mitarai. Fluid dynamics of dilatant fluids. Phys. Rev. E, 85:011401, Jan 2012. doi: 10.1103/PhysRevE.85.0

### **Friday Afternoon**

### Symposium PL

#### **Plenary Lectures**

Friday 2:20 Conference Auditorium / Plenary Lectures **Protorheology** <u>Randy H. Ewoldt</u>

Department of Mechanical Science and Engineering, University of Illinois Urbana Champaign, Urbana, IL 61874, United States

We all instinctively poke, bounce, scoop, and observe materials to understand rheological properties quickly. Yet, these observations are rarely analyzed quantitatively. To address this, here we introduce the paradigm of protorheology: approximate quantitative inference from simple observations (from proto- (first); Latin proto-; Greek p??t?-. see also: prototype).

Several case studies demonstrate how protorheology is an inclusive entry to rheology for a broad range of practitioners, strengthens the confidence and interpretation of rigorous rheometry (e.g. to avoid bad data), and supports high-throughput characterization. We survey a range of creative tests according to which rheological phenomenon is revealed. Caution is required regarding assumptions and working equations. We derive several cautionary checks and new working equations; comparison across different methods reveals a pattern of best practices that we call the Protorheology Checklist.

This establishes a framework to enable increased use of photos, videos, and quantitative inference and to support the increasing interest in digital image analysis, inverse methods, and high throughput characterization being applied to rheological properties.

Hossain, M. T., and R. H. Ewoldt, "Protorheology," J Rheol (2024) https://doi.org/10.1122/8.0000667

PL3

### **Poster Session**

### Symposium PO

### **Poster Session**

Organizers: Oliver Harlen, Rob Poole and Daniel Read

#### Wednesday 5:00 Exhibition Hall / Poster Session **Brownian dynamics simulation of colloidal film in drying process** Yeji Kim<sup>1</sup>, Jiyun Jeong<sup>2</sup>, and Young Ki Lee<sup>3</sup>

<sup>1</sup>School of Food Biotechnology and Chemical Engineering, Hankyong National University, Anseong, Gyeonggi-do 17579, Republic of Korea; <sup>2</sup>School of Food Biotechnology and Chemical Engineering, Hankyong National University, Anseong, Gyeonggi-do 17579, Republic of Korea; <sup>3</sup>School of Food Biotechnology and Chemical Engineering, Hankyong National University, Anseong, Gyeonggi-do 17579, Republic of Korea

We study the drying process of colloidal films using Brownian Dynamics (BD) simulation. We focus on the effects of process variables, for example, drying rate and sedimentation force on microstructures by drying colloidal suspensions. To understand the trend under each condition, we adopted a dimensionless number of Ns, which describe the relative effects of drying and sedimentation. BD simulations are performed at various Ns to observe changes in particle distribution along film height. Here, the initial particle volume fraction is set to phi=0.1, and the particles are assumed to be hard spheres. It is confirmed that Ns have a huge effect on local particle structure during the drying process. With increased Ns, sedimentation is more dominant than evaporation, so thicker particle layers are formed near the substrate. Conversely, a large accumulation of particles is observed near the liquid-gas interface as Ns decreases. To verify these results, we compare and analyze the 1-dimensional model proposed by Wang and Brady. The BD simulation results and 1-dimensional modeling are consistent across most drying process, a few numbers of particles form the structure, while most particles form the structure after it reaches the final stage. Different crystalline structures are formed depending on the drying and sedimentation variables. For example, with slow evaporation conditions, most colloidal particles form HCP structure, but more FCC structures are evolved by tuning the sedimentation force.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### **Structure and dynamics of a glass-forming liquid with pinning disorder** Saumya Suvarna and Madhu Priya

Physics, Birla Institute of Technology, Mesra, Ranchi, Jharkhand 835215, India

We study the role of pinning disorder on the structure and transport properties of a system of particles interacting via a soft-repulsive plus attractive potential. The particles are pinned using the template pinning protocol, and its impact on the equilibrium structure of the system is investigated by computing the radial distribution function and the hexatic order parameter for the different extents of particle pinning. We further explore the effect of pinning on the dynamics of the system by computing the self-intermediate scattering function and mean-squared displacement. We also analyze the onset of the glassy behavior in the system by varying the packing fraction and the fraction of the pinned particles.

Wednesday 5:00 Exhibition Hall / Poster Session

# Alteration of architecture and mechanics of monomeric folded protein hydrogels through seeding during network formation

Kalila R. Cook<sup>1</sup>, Lorna Dougan<sup>1</sup>, David Head<sup>2</sup>, Matt D. Hughes<sup>1</sup>, Sophie Cawood<sup>3</sup>, Leon F. Willis<sup>3</sup>, Najet Mahmoudi<sup>4</sup>, and David J. Brockwell<sup>5</sup>

<sup>1</sup>School of Physics and Astronomy, University of Leeds, Leeds, West Yorkshire LS2 9JT, United Kingdom; <sup>2</sup>School of Computing, University of Leeds, Leeds, United Kingdom LS29LX, United Kingdom; <sup>3</sup>School of Molecular and Cellular Biology, University of Leeds, Leeds, United Kingdom; <sup>4</sup>ISIS Neutron and Muon Source, STFC, Didcot, United Kingdom; <sup>5</sup>Astbury Centre for Structural Molecular Biology, University of Leeds, Leeds, West Yorkshire LS2 9JT, United Kingdom

Folded globular proteins offer great potential as building blocks in biomaterials such as hydrogels. Gaining an understanding of the design parameters of these materials is important in the field of biomedicine where protein hydrogels can be engineered and tuned as responsive scaffolds in tissue engineering. Here, we use a combined computational and experimental approach to explore whether folded globular protein hydrogels can be modelled as colloidal networks. This multi-modal approach has allowed for a cross-length and time scale understanding of the properties of the gels and the mechanisms involved in their formation. Rheological and SANS results of bovine serum albumin (BSA) folded protein hydrogels have shown that the presence of protein oligomers (a small number of folded protein monomers that have self-associated) in an otherwise

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monomeric protein pre-gel solution has a significant effect on the structure and mechanical response of formed hydrogels. A non-monotonic trend in gel stiffness as a function of monomer:oligomer ratio is attributed to a 'seeding' effect of oligomers at low fractions and the increased thermal stability of oligomers at higher oligomer fractions. A coarse-grained model was developed to simulate protein gelation over long timescales and resultant gel structure, and this has demonstrated that the gelation of folded protein monomers can be well approximated with a colloidal approach [1]. However, the presence of oligomers leads to a non-trivial relationship which a pure colloidal treatment is unable to reproduce. This work has highlighted the need for greater consideration of the impact of folded protein oligomers on the formation and properties of hydrogels and protein networks. More broadly, decoding the assembly, properties and impact of oligomers in other hierarchical protein structures may serve to inform novel biomedical therapeutics such as for the prevention of amyloids in neurodegenerative diseases.

[1] Soft Matter 2023, 19, 2780-2791.

#### Wednesday 5:00 Exhibition Hall / Poster Session

PO4

Attractive carbon black dispersions: structural and mechanical responses to shear Julien Bauland<sup>1</sup>, Louis-Vincent Bouthier<sup>2</sup>, Arnaud Poulesquen<sup>3</sup>, and Thomas Gibaud<sup>1</sup>

<sup>1</sup>Laboratoire de physique, ENSL, Lvon 69007, France: <sup>2</sup>CEMEF, Université PSL, Paris, France: <sup>3</sup>CEA, Marcoule, France

The rheological behavior of colloidal dispersions is of paramount importance in a wide range of applications, including construction materials, energy storage systems and food industry products. These dispersions consistently exhibit non-Newtonian behaviors, a consequence of intricate interplays involving colloids morphology, volume fraction, and inter-particle forces. Understanding how colloids structure under flow remains a challenge, particularly in the presence of attractive forces leading to clusters formation. In this study, we adopt a synergistic approach, combining rheology with ultra small-angle X-ray scattering (USAXS), to probe the flow-induced structural transformations of attractive carbon black (CB) dispersions and their effects on the viscosity. In practice, flow curve tests were conducted on CB suspensions in oil with volume fractions ranging from 0.6 to 4.1 %. Our key findings can be summarized as follow. First, in the hydrodynamic regime at high shear rate, CB particles are structured into fractal clusters, which size conforms to a power law of the shear rate, with the breaking constant m = 0.5. Second, drawing insights from the fractal structure of clusters, we compute an effective volume fraction and find that microstructural models adeptly account for the hydrodynamic stress contributions. For each volume fraction, we identify a critical shear rate at which the clusters percolate to form a dynamical network, that sets the lower bound of the hydrodynamic limit. Third, we show that under fast transient shear, the apparent yield stress measured at low shear rates inherits its properties from this percolation point.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### High pressure effects in the sol-gel transition of gelatin dispersions

<u>Nikolaos A. Burger</u><sup>1</sup>, Nikolaos A. Burger<sup>2</sup>, Gerhard Meier<sup>3</sup>, Dimitris Vlassopoulos<sup>1</sup>, Dimitris Vlassopoulos<sup>2</sup>, and Benoit Loppinet<sup>1</sup>

<sup>1</sup>IESL-FORTH, Heraklion 71110, Greece; <sup>2</sup>Materials Science & Technology, Unicersity of Crete, Heraklion 70013, Greece; <sup>3</sup>Jülich, Biomacromolecular Systems and Processes, Jülich, Germany

The effect of pressure and pressure cycle on the rheology of gel forming systems are not that well-established. Gelatin an archetype of gelling systems, is a biopolymer, derived from the hydrolysis of Collagen. We report the effect of hydrostatic pressure on the sol gel transition of gelatin dispersions. We use dynamic light scattering and light scattering probe microrheology to monitor the evolution of the visco-elastic response during isothermal gelation. It provides easy identification of the sol gel transition and the corresponding isothermal gelation time (tg). We established that tg decreases with increasing pressure, i.e. gelation speeds up. The temperature dependence of tg follows the well-established empirical rule up to 100MPa. It is governed by the distance to the collagen denaturation temperature (~314 K, at 0.1MPa) which is known to increase with pressure by 0.04 K/MPa. We also report the time evolution of the viscosity (elastic modulus) in the sol (gel) state. The rates of increase, increases with pressure. Using reverse quenching approach, we observe slowdown of the gel melting when pressure is increased. Our results confirm that the rheological evolution directly relates to the helix formation process. The interplay between temperature and pressure with origin in the helix-coil thermodynamics induce changes in the mechanical properties of gelatin gels.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### Rheology of organoclay dispersion gels formed upon flow cessation

<u>Nikolaos A. Burger</u><sup>1</sup>, Nikolaos A. Burger<sup>2</sup>, Benoit Loppinet<sup>2</sup>, Andrew Clarke<sup>3</sup>, George Petekidis<sup>1</sup>, and George Petekidis<sup>2</sup> <sup>1</sup>Materials Science & Technology, Unicersity of Crete, Heraklion 70013, Greece; <sup>2</sup>IESL-FORTH, Heraklion 71110, Greece; <sup>3</sup>Schlumberger Cambridge Research, Cambridge CB3 0EL, United Kingdom

Clay minerals are one of the most abundant natural materials used widely in coatings, construction materials, soil chemistry, ceramics, as well as being part of drilling fluids. Here, we present the effect of steady and oscillatory shear on organophilic modified clay particle gels in synthetic oil. The platelets-VG 69 and needles-Pangel B5 are both precursors of drilling fluid formulations. The concentration dependence of VG-69 and B5 are for the plateau modulus  $G_P \sim c^{3.9}$ , yield stress  $\sigma_y \sim c^{2.9}$  and yield strain  $\gamma_y \sim c^{-1.71}$ . In mixtures we observed an intermediate behavior: at low concentrations the dispersion follow Pangel B5 alone, whereas at higher concentrations mixtures have a weaker power law behavior of their elasticity and yield strain. Furthermore, upon varying the pre-shear history, the gel viscoelastic properties (and their underlying structure) can be largely tuned. At lower (higher) clay concentrations, pre-shear induces hardening (softening) of the dispersions but the sheared dispersions always yield at lower shear strains. In B5 pre-shear at varying shear strain amplitudes resulted in changes from  $G_P \sim c^{3.9}$  to  $G_P \sim c^{2.5}$  and from  $\gamma_y \sim c^{-1.7}$  to

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 $\gamma_y \sim c^{-1}$ . Preliminary Rheo-SAXS did not reveal big structural changes (in the covered q-range). Consequently, our findings open possibilities to design colloidal organoclay dispersions with the mechanical response which can be enhanced or weakened at will.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### **Yielding, Shear Banding, and Thixotropic Behaviour of a Cellulose Nanocrystal (CNC) Gel** Jourdain H. Piette, Xiaohan Jia, and Savvas G. Hatzikiriakos

Chemical and Biological Engineering, The University of British Columbia, Vancouver, BC V6T1Z3, Canada

This study focuses on the rheological behavior of a cellulose nanocrystalline gel. Adding electrolytes into a cellulose nanocrystal (CNC) suspension transforms it into an attractive glass (gel). This system (5 wt% CNC + 20 mM NaCl) is proved to be thixotropic, as expected for such glassy materials. From strain-controlled experiments, a non-monotonic steady-state flow curve with a minimum stress value of ~33 Pa is found, and the negative slope indicates the existence of shear bands. From stress-controlled experiments, the true yield stress is determined to be  $62.5 \pm 2.5$  Pa. This difference proves that the minimum stress of the flow curve does not coincide with the true yield stress by creep tests ( $62.5 \pm 2.5$  Pa). However, this minimum stress can maintain the flow provided that the material has already yielded. The size of the aggregates of the glassy material is related to the shear rate. At nominal shear rates below about 100 1/s, the shearing is suggested to be localized in a shear band rather than in the whole material. Therefore, the real shear rate in the band is different from the nominal shear rate. At low nominal shear rates, a layer of critical thickness explains the negative slope of the flow curve: a higher nominal shear rate also represents a higher real shear rate within the band, breaks the aggregates of CNC rods lead to the detachment of the sample from the wall and thus to severe wall slip. Our finding of the robust connection between yielding, thixotropy, wall slip, and shear banding shall shine new light on the nature of the non-monotonic flow curves of yield stress, and thixotropic materials

#### Wednesday 5:00 Exhibition Hall / Poster Session

Yielding mechanisms in Mg(OH)2 suspensions with spherical nanoparticle additives

Olivia Pickup<sup>1</sup>, Martyn Barnes<sup>2</sup>, Alexander Lockwood<sup>2</sup>, Timothy Hunter<sup>1</sup>, and David Harbottle<sup>1</sup>

<sup>1</sup>School of Chemical and Process Engineering, University of Leeds, Leeds LS2 9JT, United Kingdom; <sup>2</sup>Sellafield Ltd, Warrington, United Kingdom

A significant activity of the UK nuclear sector is decommissioning, which involves removing and packaging legacy wastes for their long-term safe storage. Intermediate level waste materials include magnesium alloy fuel cladding that has corroded over time in underwater storage to form a complex suspension with a high solids content, leading to difficulties in the disposal process due to high yield stresses. To mitigate these issues, the current research has explored the mechanism behind the use of silica (SiO<sub>2</sub>) to lower the yield stress of concentrated magnesium hydroxide (Mg(OH)<sub>2</sub>) suspensions and investigated the effects of altering SiO<sub>2</sub> particle size. The yield stress of a 28 vol% Mg(OH)<sub>2</sub> suspension was 70 Pa, but when blended with 2 vol% 100 nm SiO<sub>2</sub>, the yield stress decreased to 38 Pa. This rheological modification using SiO<sub>2</sub> was explored using different sized particles at 250, 500 and 800 nm, which also reduced the yield stress to 38, 53 and 60 Pa respectively. Synchrotron x-ray computed tomography (X-ray CT) was used to determine the dispersion of SiO<sub>2</sub> throughout the Mg(OH)<sub>2</sub> network. The size and number of clusters was found to decrease with increasing SiO<sub>2</sub> particle size. The number ratio of Mg(OH)<sub>2</sub> to SiO<sub>2</sub> particles showed to be significant in this case, where there was a minimum number of SiO<sub>2</sub> needed to induce significant yield stress reduction (from 70 Pa to 38 Pa). Large amplitude oscillation strain rheology (LAOS) was used to further explore the effects of SiO<sub>2</sub> particle size on the yielding mechanisms and modification within the binary suspension. From analysis of Lissajous plots, the size of silica had significant effect on the energy dissipation within the system, which is likely linked to interparticle friction and its influence on the yield stress reduction.

#### Wednesday 5:00 Exhibition Hall / Poster Session

### Rheology of graphene oxide suspensions in xanthan gum

José Carlos C. Rodriguez<sup>1</sup>, Lorena R. Moraes<sup>1</sup>, Ricardo Andrade<sup>2</sup>, Hélio Ribeiro<sup>2</sup>, and <u>Mônica F. Naccache<sup>1</sup></u> <sup>1</sup>Mechanical Engineering, Pontificia Universidade Catolica do Rio de Janeiro, Rio de Janeiro, RJ 22451900, Brazil;

<sup>2</sup>Engineering School, Mackenzie Presbyterian University, São Paulo, Brazil

The introduction of graphene oxide (GO) nanosheets into drilling fluids can significantly improve many important properties of the base fluids. This work aims to study the effects of the GO nanosheets on the rheology of aqueous dispersions of xanthan gum (XG). Xanthan gum is used as a viscosifier in water-based drilling fluids, and it is also used in many other industries, from food to cosmetics. In the present work, the graphene oxide was prepared using the modified Hummer's method. The suspension preparation method involves three steps: initially, the GO exfoliation in 30% wt of the total water; in parallel, the hydration and homogenization of XG in the remainder of the water for one hour; the final suspension is then obtained by mixing both dispersions through mechanical stirring for 30 minutes. The effect of NaCl brine on the rheological behavior of aqueous xanthan gum dispersions is also analyzed. The influence of NaCl on rheology is particularly important because water-based drilling fluids are usually prepared in saturated solutions. Rheological characterizations were conducted on a TA Instruments ARES-G2 rate-controlled rotational rheometer, using the parallel plate geometry with the sandpaper surface and 1 mm gap. Rheological tests were carried out in steady and oscillatory flows, and the viscosity, storage, and loss moduli were obtained for different temperatures. The analysis conducted at room temperature indicates that the dispersions are modeled by the Carreau-Yasuda equation. Incorporating GO nanosheets into XG tends to increase viscosity. However, adding NaCl induces the opposite effect. Tests with 4, 50, and 80°C are being conducted. As drilling fluids are made up of many components, comprehensive and in-depth analysis of the rheological effects of incorporating GO into more basic model fluids, in addition to being considered fundamental studies, can be of great value for the development of new nanofluids, with application in a wide range of industries.

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### Wednesday 5:00 Exhibition Hall / Poster Session **A double rigidity transition rules the fate of drying colloidal drops** <u>Matteo Milani</u>, Christian Ligoure, Luca Cipelletti, and Laurence Ramos

University of Montpellier, CNRS, MONTPELLIER 34000, France

The evaporation of drops of colloidal suspensions plays an important role in numerous contexts, such as the production of powdered dairies, the synthesis of functional supraparticles, and virus and bacteria survival in aerosols or drops on surfaces. The presence of colloidal particles in the evaporating drop eventually leads to the formation of a dense shell that may undergo a shape instability. Previous works propose that, for drops evaporating very fast, the instability occurs when the particles form a rigid porous solid, constituted of permanently aggregated particles at random close packing. To date, however, no measurements could directly test this scenario and assess whether it also applies to drops drying at lower evaporation rates, severely limiting our understanding of this phenomenon and the possibility of harnessing it in applications. Here, we combine macroscopic imaging and space- and time-resolved measurements of the microscopic dynamics of colloidal nanoparticles in drying drops, measuring the evolution of the thickness of the shell and the spatial distribution and mobility of the nanoparticles. We find that, above a threshold evaporation rate, the drop undergoes successively two distinct shape instabilities. While the second instability is due to the permanent aggregation of nanoparticles, as hypothesized in previous works on fast-evaporating drops, we show that the first one results from a reversible glass transition of the shell, unreported so far. We rationalize our findings and discuss their implications in the framework of a unified state diagram for the drying of colloidal drops.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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## Effect of carboxymethyl cellulose and temperature on rheological properties of anode slurries for lithium-ion batteries

#### Seung Chang Lee, Jun Seo Lee, and Seong Jae Lee

Department of Chemical and Materials Engineering, The University of Suwon, Hwaseong, Gyeonggi 18323, Republic of Korea

Lithium-ion batteries (LIBs) are used as energy storage devices, including electric vehicles, due to their high operating voltage, capacity, and longlife cycle. To improve the performance of LIBs, most efforts are focused on developing active materials, but research on the rheology of electrode slurries composed of active materials, conductive materials, binders, and solvents is insufficient. In this study, the rheological characteristics of binder molecular weight (MW), temperature, and time were examined for anode slurry. The flow curve, yield stress, and thixotropy were analyzed using three types of molecular weight (90K, 250K, and 700K) of carboxymethyl cellulose (CMC), a binder, and the storage modulus, loss modulus, and complex viscosity under small amplitude oscillatory shear were analyzed. When the MW of CMC was small, shear thickening behavior was observed as the shear rate increased. As the MW increased, this phenomenon was suppressed and a distinct yield stress appeared. In oscillatory shear strain sweeps, strain hardening behavior was observed in the strain range 100-300%. The temperature dependence of the slurry viscosity was evident at a shear rate of 10-100/s. At low shear rates, the influence of particles was dominant, whereas at high shear rates, the influence of the binder was dominant. To understand the time dependence, a hysteresis loop was constructed and thixotropy was examined. The slurry showed thixotropy overall, but also showed some rheopexy at high shear rates. Understanding the rheological properties of electrode slurry can help not only understand the microstructure of the slurry but also improve stable productivity.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### **Self-supported MOF/cellulose-nanocrystals materials designed from ultrafiltration** <u>Lorenzo Metilli<sup>1</sup>, Héloïse Ugo<sup>2</sup>, William Chèvremont<sup>3</sup>, Cyril Picard<sup>4</sup>, and Frédéric Pignon<sup>5</sup></u>

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Metal-organic frameworks (MOFs) are promising materials to address critical issues such as petrochemical and gas separation, filtration, catalysis, sensing and energy storage [1]. Large-scale deployment of MOFs is however hampered by their crystalline powder state, which results in poor processability. Recently, the hybridization of MOFs with biopolymers has emerged as a greener, biocompatible strategy with improved processability into membranes, films, and porous materials [2]. Nevertheless, the physiochemical properties of the biopolymer-MOF mixtures, and their relationship to the composite structure and functionality, need to be investigated to promote their use in real applications.

The present work focuses on the preparation of thin nanocomposites from commercial ZIF-8 (a synthetic zeolite) and cellulose nanocrystals (CNCs) aqueous suspensions using membrane ultrafiltration. An investigation carried out with a rheo-optical setup show that the addition of small quantities of CNCs (1:20 CNC:ZIF-8 vol. ratio) affect significantly the flow of ZIF-8 particles, endowing the suspension with colloidal stability through the electrostatic interactions between the two materials. The CNCs removed the yield stress observed in the ZIF-8 suspensions, prevent their aggregation into a percolated network, and enabled their deposition on the membrane during filtration. The process was also investigated with in situ SAXS, which tracked the concentration of CNCs along the deposit height. Here, the deposit was formed by layers of CNCs containing ZIF-8 particles, perpendicular to the membrane. Finally, the porosity of ZIF-8 to water was tested with a custom-made porosimeter [3], showing that the adsorbed CNCs did not affect the intrusion/extrusion pressure, or the available pore volume in the hybrid material.

#### References:

[1] Freund et al., Angew. Chem. Int. Ed. 2021, 60, 23975-24001.

- [2] El Hankari et al., Prog. Mater. Sci. 2019, 106, 100579-100616.
- [3] Guillemot et al., Rev. Sci. Instrum., 2012, 83, 105105

Poster Session

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### Multi-scale study of chiral self-assembly of cellulose nanocrystals during ultrafiltration

Samuel Mandin<sup>1</sup>, Lorenzo Metilli<sup>2</sup>, Nicolas Hengl<sup>1</sup>, Bruno Jean<sup>2</sup>, and Frédéric Pignon<sup>1</sup>

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Rodlike cellulose nanocrystals (CNCs) exhibit significant potential as building blocks for creating uniform, sustainable materials. However, a critical hurdle lies in the need to enhance existing or devise novel processing that provides improved control over the alignment and arrangement of CNCs across a wide spatial range. In previous works, it has been evidence that membrane filtration processes can be used to efficiently control the concentration polarization layer of anisotropic nanoparticles, both in terms colloidal concentration and orientation |1,2]. Nevertheless, until now in situ characterization during the filtration process has only been achieved at nanometer length scales. Consequently, the aim of this work was to access to multi-scale organization under ultrafiltration from nanometer to micrometer length scales by in situ monitoring using small-angle X-ray scattering (SAXS) and small-angle light scattering (SALS) respectively. During filtration, it was demonstrated that in the polarization layer, CNCs align parallel to the membrane, while forming chiral nematic (cholesteric) structures oriented with their helical axes oriented perpendicular to the membrane pressure) revealed CNC diffusion throughout the volume, influenced by colloidal forces and particle osmotic pressure. This phenomenon led to a homogenization of the concentration while maintaining the orientation induced by the filtration process over a wide range of scales, from CNCs to cholesteric structures. Finally, the observation of the concentrated deposit developed during the ultrafiltration process.

[1] Rey C. et al. ACS Sustainable Chem. Eng. 7, 10679-10689 (2019)

[2] Semeraro et al. Colloids and Surfaces A: Phys. and Eng. Aspects, 584, 124030 (2020)

#### Wednesday 5:00 Exhibition Hall / Poster Session Multiply scattered sound and strain in a granular suspension Ibrahim Awada

#### Université Gustave Eiffel, Champs sur marne, France

Granular materials are systems for which describing mechanical properties poses a major challenge due to the complexity of their amorphous structure. The lack of knowledge on the nature of the elementary mechanisms responsible for plasticity makes the understanding of how they deform a daunting task. It is nevertheless essential to better predict landslides and earthquakes. Advanced characterization approaches, such as optics and X-ray tomography, offer the possibility to study deformation at the scale of the particle but are hampered by their complexity and long acquisition times. Characterizing the deformation of granular materials is challenging, particularly in sudden phenomena such as avalanches. In this context, scattering methods may provide complementary information to scanning methods. Namely, acoustic waves benefit from long propagation distances and are able to effectively probe in the bulk systems such as granular suspensions. Here, we study a dense granular suspension in the elastoplastic regime, inserted in a rotating drum oscillating at angles inferior to the angle of repose. To probe the structure of the suspension, some ultrasound pulses are emitted from one side of the drum and the transmitted signal is collected at the other side. Experimentally, the granular suspension compacts, leading to rearrangements of grains and modifications of transmitted ultrasound signals. In order to measure strain, we propose a new method based on the correlation of transmitted multiply scattered ultrasound waves, sensitive to volumetric deformation of 10^-5. The method can detect local strain at the scale of a few grains and can therefore be applied in systems with heterogeneous deformations.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### Rheology of surface flow of granular materials over erodible heaps: DEM simulations

<u>Neiladri S. Ray</u> and Devang V. Khakhar

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Surface flow of granular material on a heap, defined as the flow of a shallow layer of particles over an erodible bed of the same material, has relevance, both industrially as well as in understanding several natural phenomena such as landslides, avalanches etc. Erosion or deposition of materials into the bed, is governed by the difference between the surface angle and *neutral* angle (surface angle when there is no erosion or deposition) of the heap, as given by the BCRE [1] and BRDG [2] models. The presence of sidewalls is known to affect such flows significantly [3,4]. Here, we present the rheology of steady surface flows on quasi-two-dimensional erodible heaps, by performing computations using the Discrete Element Method (DEM). The effect of variation of different parameters such as the mass flow rate, particle stiffness, inter-particle friction and particle diameter are considered in the simulations. The surface angle of the heap at steady state corresponds to the equilibrium *neutral* angle and the only parameter that affects the magnitude of the surface angle is inter-particle friction. The velocity profile in the shallow flowing layer decays linearly up to a certain depth inside the heap from the free surface and then creeps exponentially deeper inside the heap. The length scale of decay in this 'creeping' zone remains unaffected by mass flow rate and particle stiffness, but increases with inter-particle friction and particle diameter. The transition from the flow to the no flow state is shown to be analogous to a melting-freezing kind of phase change, and the viscosity near the transition is found to follow a modified Vogel-Fulcher relation.

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P. Jop, et al., J. Fluid Mech. 541, 167 (2005).
 D.V. Khakhar, et al., J. Fluid Mech. 441, 255 (2001).

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### **Investigating Granular Flow Dynamics in Wedge-Shaped Hoppers: A Discrete Element Method Approach** <u>Afroz F. Momin<sup>1</sup> and Devang V. Khakhar<sup>2</sup></u>

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The gravitational flow of particulate solids, resembling liquid behaviour, is a typical phenomenon in nature and various industries. This study examines the flow of granular materials through wedge-shaped hoppers under radial gravity. We use the discrete element method (DEM), which considers particle-particle interactions to simulate the system and validate existing theories. Our DEM simulations that solve Newton's equations agree with the seminal work of Savage (1965) and Prakash and Rao (1988), except in the exit region where viscous and frictional effects coexist. A parametric analysis is performed to understand this behaviour, varying orifice diameter, wedge angle, friction coefficient, and stiffness. The results show that velocity increases with flow rate but decreases with wedge angle and friction coefficient, while variations in stiffness have minimal effects on the velocity. Stresses at the exit occur due to the momentum transfer during particle collisions. These collisions involve the momentum exchange between particles, resulting in a change in their velocities. Furthermore, a power-law correlation with the inertial number can describe the relationship between friction coefficient and volume fraction, collapsing into a single curve. An empirical continuum model for granular flows provides valuable insights into the behaviour of these materials within industrial processes.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### Simulation of Fluid-Particle suspension using the Immersed Boundary Method

Azim Babu V. Memon<sup>1</sup> and Devang V. Khakhar<sup>2</sup>

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The Immersed Boundary Method (IBM) is a numerical technique for simulating fluid-structure interactions. It handles complex solid-fluid interactions by embedding boundaries in a fluid domain and using interpolation for fluid forces. IBM finds applications in bioengineering, aerospace, and biomechanics, enabling the study of physiological processes, aerodynamics, and biomechanical interactions. With the aim to model such flows, this work proposed to extend the Signed Distance Function Immersed Boundary Method (SDFIBM) developed by Chenguang [1], which is based on OpenFOAM v6. The suggested pyramid decomposition approach and signed distance field representation of the solid shape enable precise calculation of the volume fraction field generated by solids overlapping with a random unstructured fluid mesh. The objective of the current research work is to carry out a detailed analysis of the dependence of time step, grid size, and Reynolds Number on the results of SDFIBM simulations. The SDFIBM results are compared with OpenFoam's Finite Volume Method (FVM) results to assess the accuracy and reliability, identify potential discrepancies or differences in the predicted fluid-structure interaction behaviors, and gain insights into the strengths and limitations of the method. The work emphasizes on analysis of two-dimensional study of flow past a circular cylinder and three-dimensional simulation of flow past a sphere for different particle Reynolds Number between 0.1 and 100. The work is also extended to analyze the flow through an array of cylinders. The comparison between array of fixed cylinders and vibrating cylinders for different particle Reynolds Number and volume fraction is shown.

Wednesday 5:00 Exhibition Hall / Poster Session

#### Origin of two distinct stress relaxation regimes in shear jammed dense suspensions

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Many dense particulate suspensions show a stress induced transformation from a liquidlike state to a solidlike shear jammed (SJ) state. However, the underlying particle-scale dynamics leading to such striking, reversible transition of the bulk remains unknown. Here, we study transient stress relaxation behaviour of SJ states formed by a well-characterized dense suspension under a step strain perturbation. We observe a strongly nonexponential relaxation that develops a sharp discontinuous stress drop at short time for high enough peak-stress values. High resolution boundary imaging and normal stress measurements confirm that such stress discontinuity originates from the localized plastic events, whereas system spanning dilation controls the slower relaxation process. We also find an intriguing correlation between the nature of transient relaxation and the steady-state shear jamming phase diagram obtained from the Wyart-Cates model.

#### Wednesday 5:00 Exhibition Hall / Poster Session

Time- and space-resolved stress heterogeneities in shear thickening suspensions

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Discontinuous shear thickening (DST) is associated with a sharp rise of a suspension's viscosity with increasing applied shear rate or stress. Key signatures of DST, highlighted in recent studies, are the very large fluctuations of the measured stress in time, as the suspension thickens with increasing the rate. A clear link between microstructural development and the dramatic increase of the stress fluctuations has not been established yet. To identify the microstructural underpinnings of this behavior, we perform simulations of sheared dense suspensions. Through an analysis of

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the particle contact network, we identify a subset of constrained particles that contribute directly to the rapid rise in viscosity and the large stress fluctuations. Indeed, both phenomena can be explained by the growth and percolation of constrained particle networks---in direct analogy to rigidity percolation. A finite size scaling analysis confirms this is a percolation phenomenon and allows us to estimate the critical exponents. Based on the new understanding of the specific microstructural self-organization transition that underlies DST, we then explore quantitatively the link to spatial stress heterogeneities observed in experiments.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### Dynamics of semi-flexible and breakable fibers under Poiseuille flow

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In the processing of fiber-reinforced polymer composites such as in injection molding, the fiber's orientation, length, and distribution vary depending on its properties and flow field, which affects the performance of final products. To investigate the behavior of semi-flexible and breakable fiber suspensions under Poiseuille flow, we used the hybrid simulation approach, multiparticle collision dynamics-molecular dynamics (MPC-MD), which takes hydrodynamic interactions and fiber properties (strength, flexibility) with it. Fibers that are non-breakable and rod-like align well along the direction of flow and exhibit greater alignment in close proximity to walls. The length and orientation of fibers greatly depend on their characteristics as they become flexible and/or breakable. The interesting phenomenon is specifically seen for breakable and semiflexible fibers, where the orientation of the fiber exhibits unusual behavior depending on the flow rate. This complex behavior emphasizes the importance of comprehending the dynamics of various kinds of fibers and necessitates further investigation into the optimal conditions for injection molding.

#### Wednesday 5:00 Exhibition Hall / Poster Session

Complex viscoelastic behavior of aluminum-bridged oleic acid compounds

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School of Chemical Science and Engineering, Tongji University, Shanghai, Shanghai 200092, China

In this study, we investigated the viscoelastic properties of a low molecular weight aluminum-bridged oleic acid compound and its organo-gel formed by dispersing the compound into squalane. The compound and the gels exhibit remarkably high viscoelasticity, with rheological properties similar to those of entangled polymer melts. At low frequencies, these materials behave like liquids, conforming to the Maxwell model with G' ~  $\omega^2$  and G" ~  $\omega^1$ . At high frequencies, the storage modulus G' of these materials exhibits a polymer-like entangled plateau, where the loss modulus G" displays a minimum. The platform modulus  $G_N^0$  shows a volume fraction dependence of  $G_N^0 \sim \phi^{2.9\pm 0.1}$ , but the power law exponent of 2.9 is clearly outside the range of 2.0 to 2.3 of entangled polymer solutions reported in the literature. The aluminum-bridged compound in solutions forms nanosized particles that connect in the form of an elastic network, as discovered by the SEM and AFM microscopes. Furthermore, the addition of a small amount of the compound (i.e., 10 parts per hundred parts of rubber) to carbon black-filled rubber formulations significantly improves the formulation properties such as tensile strength and rolling resistance. Keywords: viscoelasticity, organo-gels, aluminum-bridged compounds; rubber; additives.

Wednesday 5:00 Exhibition Hall / Poster Session

### Equation for rheological characterization of temperature-responsive polymers based on gelatin and poly(Nisopropylacrylamide)

#### Tilen Kopac

#### Department of Chemical Engineering and Technical Safety, University of Ljubljana, Faculty of Chemistry and Chemical T, Ljubljana 1000, Slovenia

This study introduces a mathematical model for characterizing the rheological behavior of temperature-responsive polymers. It focuses on critical temperature ( $T_c$ ), baseline viscosity or storage modulus ( $\eta_c$  or G'<sub>0</sub>), hysteresis ( $T_1$ ), and parameters m and p, delineating polymer types (LCST or UCST). The model demonstrates how  $T_c$  influences the sol-gel transition temperature and how ( $\eta_c$  or G'<sub>0</sub>) values depict baseline states. It further explores post-critical temperature behaviors ( $\eta_{\infty}$  or G'\_{\infty}) and the effects of temperature (T<sub>2</sub>) and flow behavior parameter (s). The model's application is showcased through detailed analysis of GEL and pNIPAm polymers. GEL exhibits UCST-type behavior around 28 °C, and pNIPAm displays LCST-type behavior at 37 °C. Changes in polymer concentration affect T<sub>c</sub>, with notable hysteresis in their thermoresponsive properties. The study analyzes the rate of change in viscosity and storage modulus (m and p) near T<sub>c</sub> and the concentration's impact on crosslink density. Higher concentrations, especially in pNIPAm, lead to deviations due to complex gel network formations. The study provides insights into the temperature-dependent behavior of these polymers, beneficial for applications requiring precise temperature control.

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#### Wednesday 5:00 Exhibition Hall / Poster Session Impact of the Host Polymer Linear Viscoelastic Properties on the Magnetic Hyperthermia of Polypropylene/Fe<sub>3</sub>O<sub>4</sub> nanocomposites

Laura Ea<sup>1</sup>, Vivien Truchot<sup>2</sup>, Gildas Coativy<sup>3</sup>, and Guilhem P. Baeza<sup>1</sup>

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We investigate magnetic hyperthermia of  $Fe_3O_4$  nanoparticles embedded in polypropylene of various molecular weights. While irradiating these materials with a high frequency magnetic field is expectedly seen to increase their temperature through magnetic hysteresis loss, an extra heating mode is systematically observed above the melting point of the host polymer. This phenomenon is assigned to friction between the nanoparticles and the matrix as a consequence of the gigantic drop of viscosity that enables nanoparticles rotation.<sup>[1]</sup>

In this work, we focus our attention on the impact of the linear viscoelastic properties (including the zero-shear viscosity) of the host polymer on the specific power generated by nanoparticles friction. In particular, we will show how the induction heating profiles (temperature vs. time) enable to extend the current theoretical understanding proposed for Newtonian liquids.<sup>[2]</sup>

In perspectives, we will present preliminary results on polystyrene and polymethyl methacrylate based nanocomposites where the glass transition is used as an alternative to melting to investigate the impact of dramatic viscosity drops.

<sup>[1]</sup> Pommella, A.; Griffiths, P.; Coativy, G.; Dalmas, F.; Ranoo, S.; Schmidt, A. M.; Méchin, F.; Bernard, J.; Zinn, T.; Narayanan, T.; others Fate of Magnetic Nanoparticles during Stimulated Healing of Thermoplastic Elastomers. ACS nano 2023, 17, 17394-17404.

<sup>[2]</sup> Helbig, S.; Abert, C.; Sánchez, P. A.; Kantorovich, S. S.; Suess, D. Self-consistent solution of magnetic and friction energy losses of a magnetic nanoparticle. Physical Review B 2023, 107, 054416.

#### Wednesday 5:00 Exhibition Hall / Poster Session

### Interplay of Polymer Matrix Molecular Weight and Functionality in Polymer Vitrimer

#### Hao Wang<sup>1</sup>, Charles-André Fustin<sup>2</sup>, and Evelyne van Ruymbeke<sup>3</sup>

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Vitrimers, developed by Leibler and his colleagues<sup>1</sup>, exhibit interesting mechanical performance and recyclability, making them an attractive alternative to traditional polymers, as well as a promising solution to decrease the environmental impact of polymer materials. With the increasing focus on the development of new chemistry in vitrimers, it is now crucial to better understand the relationship between their composition and their viscoelastic properties, to allow their rational design as well as for their large-scale processing and manufacturing.

Indeed, today, the impact of the molecular weight of the polymer precursor, the crosslinker density of the vitrimer, and the functionality of the vitrimer on its viscoelasticity remains unclear. Therefore, the objective of this project is to synthesize and examine the viscoelastic properties of well-defined model vitrimers in order to elucidate the effects of these composition parameters on the material's rheological behavior.

Entangled and unentangled vitrimers containing diol pendant groups<sup>2</sup> and appropriate ratio of crosslinkers have been synthesized. In such a way, we can investigate and discuss the influence of each of these parameters separately, and determine what are the key ingredients to control their dynamics.

1 D. Montarnal, et al., Science. 334, 965 (2011). 2 Röttger, Max, et al. Science 356.6333 (2017).

#### Wednesday 5:00 Exhibition Hall / Poster Session

# Vitrimers as additive to improve the dynamics and crystallization process of degraded semi-crystalline polyolefins

<u>Anthi Sapouna</u><sup>1</sup>, Renaud Nicolay<sup>2</sup>, Nathan van Zee<sup>3</sup>, George Floudas<sup>4</sup>, and Evelyne van Ruymbeke<sup>5</sup> <sup>1</sup>Inst. on Condensed Matter & Nanosciences, Dept. of Physics, Université Catholique de Louvain, University of UOI, BSMA, Louvain-La-Neuve 1348, Belgium; <sup>2</sup>Chimie Moléculaire, Mottérieux, ESPCI, Université PSL, Paris, France; <sup>3</sup>Chimie Moléculaire, Mottérieux, ESPCI, Université PSL, Paris, France; <sup>4</sup>Dept. of Physics, Institute of Materials Science & Computing, University of Ioannina, University Research Center of UOI, Ioannina, Greece; <sup>5</sup>IMCN, BSMA, Université catholique de Louvain, Louvain-la-Neuve, Belgium

A major recent advance, which can serve as a very promising solution to the problem of mechanical degradation of plastic waste, is the development of associative dynamic covalent networks called "vitrimers". 1 As the exchange reactions can occur between different positions along the polymer chains, macroscopic flow is achieved at long times without compromising the network coherence. Since these exchange reactions are thermally activated, the exchange dynamics are slow at lower temperatures, imparting strength to the material akin to thermosets. 2 At higher temperatures used for processing, the exchange reactions are fast, allowing the material to flow. Consequently, vitrimers can be reshaped and reprocessed at wish, while maintaining the desired chemical and mechanical resistance of crosslinked polymers, which is of prime interest for developing recyclable elastomers.3 Vitrimers have a huge potential as agents to improve the mechanical properties of recycled polymers and can serve as critical ingredients for novel materials. However, despite the large progress in developing new vitrimers, these materials are not yet used, today, to improve commodity plastic recycling.4 This requires, first, to understand how the addition of vitrimers in a degraded polymer will affect

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its properties. Within this direction, we selected two different grades of High-Density Poly(Ethylene) (HDPE) and studied their degradation after several cycles of processing, based on their linear viscoelastic properties. We then investigate the influence of the addition of different amounts of HDPE vitrimers on the dynamics, creep resistance, crystallization and structure of the degraded samples.

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#### Wednesday 5:00 Exhibition Hall / Poster Session

#### **Evaluating the dynamics of polyolefin vitrimers**

Anahita Karimi<sup>1</sup>, Huidi Sun<sup>2</sup>, Nathan J. Van Zee<sup>2</sup>, Renaud Nicolaÿ<sup>2</sup>, Sina Ghiassinejad<sup>1</sup>, and Evelyne van Ruymbeke<sup>1</sup> <sup>1</sup>IMCN, BSMA, Université catholique de Louvain, Louvain-la-Neuve, Belgium; <sup>2</sup>Chimie Moléculaire, Macromoléculaire, Matériaux, ESPCI Paris, Paris, France

Polyolefins, as thermoplastics, play an essential role in worldwide plastic consumption but pose challenges in recycling due to issues such as thermal degradation and significant loss in their mechanical properties during the recycling process. To address these issues, polyolefin treatment has emerged as a key solution to enhance their properties. In particular, incorporating dynamic covalent bonds within the polymer chains has appeared as a novel and promising approach. Indeed, the material obtained, which are called vitrimers, combine the properties of thermosets and thermoplastics, thanks to the exchange reactions of their dynamic bonds, which allows topology changes while maintaining a constant number of chemical bonds. By adjusting the timescale of the dynamic covalent bonds through temperature, a wide range of properties can be obtained, and this makes vitrimers appealing in diverse applications which require recyclability, self healability, and re-processability. This study focuses on polyethylene (PE) vitrimers as PE is a widely used plastic. Adding dynamic covalent bonds (crosslinkers) suppresses system crystallinity and rheological analysis indicates that network elasticity is largely influenced by the crosslinker content. Networks with higher crosslinker concentrations also display enhanced creep resistance, and improved recovery in creep-recovery measurements. These findings underscore the high potential of this material for sustainable applications. Moving forward, our research aims to explore and rationalize the influence of the crosslinking density and the lifetime of the exchange bonds on the network dynamics and its mechanical properties.

#### 5:00 Exhibition Hall / Poster Session Wednesday

Correlation between relaxation behavior and transesterification catalyst structure in epoxy vitrimers

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In this contribution the impact of the catalyst type on the linear rheological behavior of various anhydride cured epoxy vitrimers is analyzed. Vitrimers represent a specific class of materials which exhibit an intermediate behavior between thermosets and thermoplastics. These intermediate features are enabled by the combination of the network structure and fixed crosslink density, distinctive of thermosets, with the presence of dynamic covalent bonds allowing thermally activated structural rearrangement. This study aims to correlate network structure and rheological properties in the linear regime. The vitrimers studied here are based on both commercial (petroleum-derived) and bio-based epoxy resins, thermally cured using anhydride hardeners and containing a series of transesterification catalysts. The effect of various catalysts [1] is directly related to the stress relaxation ability of the studied vitrimers and their re-processability. Specifically, the relationship between the structure of selected metal catalysts and the linear rheological behavior of the investigated epoxy vitrimers is here addressed. A quantitative relationship between the structural properties and the viscoelastic behavior is established through the application of a mathematical model to the experimental results. The model introduced in this work is based on a continuum free energy theory proposed by Meng et al. [2] and adapted to the obtained experimental data for describing their rheological behavior.

[1] W.Liu et al. Ind. Eng. Chem. Res. 56, 10, 2667-2672 (2017) [2] F. Meng et al. Macromolecules 52(19) 7423-7429 (2019).

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### Using machine learning to deduce molecular weight distribution from rheology

Robert Elliott<sup>1</sup>, Daniel J. Read<sup>1</sup>, Luisa Cutillo<sup>1</sup>, and Johan Mattsson<sup>2</sup>

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Recent advances in rheological modelling have enhanced our ability to quickly and accurately predict rheology given an arbitrary polydisperse polymer blend. We seek to reverse this prediction, using machine learning techniques to infer molecular structure from rheological measurements, which combine easy accessibility with richness in molecular information. This ease of accessibility enables data to be gathered cheaply, which would be hugely beneficial for the classification of polymers in the recycling industry. Our goal is to train and optimise neural network models for inferring the molecular weight distribution. This, however, requires large data sets, beyond what is readily available experimentally. We use forward modelling - more specifically the LP2R software [Das and Read, J. Rheol, 2023] - to produce large data sets of storage and loss moduli for a range of molecular weight distributions. We consider the optimal format for presenting data to the models, addressing challenges posed by irregular formats and noise. We present a solution by fitting the rheological curves with regularised Maxwell modes and evaluate the 'information

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capacity' of this alternate data type when optimised for machine learning. This research forms part of the research programme of DPI, project #861.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### Non-linear time-dependent behavior of amorphous polymers: experimental and theoretical approach Lidija Slemenik Perse and Alen Oseli

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Polymers are widely used in various industrial and research areas due to their numerous benefits (light weight, chemical resistance, ease of production). However, as plastic production grows, the amount of plastic waste significantly burdens the environment. Although the sustainability of plastic products may be experimentally evaluated in several ways, numerical simulations offer numerous benefits over real time testing (time and energy consumptions). The prediction of plastic product behavior usually addresses linear viscoelastic constitutive laws (proportionality and additivity of stress/strain response over time); therefore, the theory is significantly limited to a magnitude of external loading conditions (within linear viscoelastic domain). The focus of the research was to develop the methodology for predicting non-linear viscoelastic behavior, utilizing Schapery constitutive equation (non-linear model derived from non-equilibrium thermodynamic theory), extended to amorphous (rheodictic) polymers incorporating irrecoverable flow term. Schapery's non-linear coefficients were determined by conducting long-term creep tests of amorphous thermoplastics at extreme temperatures, representing stress dependent irrecoverable instantaneous, creep and recovery deformations as well as creep acceleration factor. The results showed precise predictions (within experimental error ~4%) at extreme loading conditions, compared to linear viscoelastic laws, which can deviate from experimental stress and time windows (maximal error ~7%). By utilizing the presented methodology, it is possible to predict long-term material behavior at extreme loadings and environmental conditions, enabling improved sustainability for various applications, like plastic products, complex shaped products and decreased material consumption, which is in line with current directives on sustainability and climate neutrality.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### Scaling of the Linear Viscoelasticity of Entangled Polymer Solutions

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It is well known that linear viscoelasticity of entangled polymer solution can be characterized by two scale factors such as the plateau modulus and reptation relaxation time. The two scale factors are considered as functions of concentration and molecular weight. If the dynamic modulus of entangled polymer solution is scaled by the two scale factors then it is expected that the plot of dimensionless modulus as a function of dimensionless frequency is invariant with respect to concentration and molecular weight because this superposition is found for linear viscoelasticity of unentangled polymer solutions. If adopting the two scale factors from entangled dynamic theory of monodisperse polymers, the quality of superposition is not good. On the other hand, if two scale factors are obtained from zero-shear viscosity and steady state compliance then better superposition if found. However, the mean modulus (the inverse of steady state compliance) and the mean relaxation time (the product of steady state compliance and zero-shear viscosity) do not show clear functional relations with respect to concentration and molecular weight. Hence, we applied numerical method for superposition and determined horizontal and vertical shift factors. The shift factors correspond to the scale factors. We found that the numerically determined shift factors show clear functional relations with respect to concentration and molecular weight.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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#### Linear and non-linear rheology of blends of linear and pompom molecules : a molecular tube model Benoît Blottière

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A molecular tube-based model is derived for the linear and non-linear rheology of blends of pompom molecules with linear chains in the melt. The polymer architecture has a strong impact on relaxation mechanisms depending on the degree of branching in polymer melts. For monodisperse linear chains it was shown experimentally that the terminal relaxation time scales as M^3.4 and was well described with the reptation model(1) and the tube model(2). For monodisperse star molecules it was shown experimentally that the longest relaxation time scales as the exponential of molecular weight of the arm Ma. The dynamic dilution concept(3) qualitatively enhance the previous theoretical predictions. From these two architectures (linear and star) melts of blends were investigated both experimentally and theoretically and the theory based on the tube model for linear polymer and dynamic dilution for star was successful to describe the relaxation mechanisms(4). More complex topological architectures were achieved with the H-shaped molecules and that was generalized with the pompom (two branched points) molecules. The pompom model(5) was successful to describe experimental results on the monodisperse samples. In this pompom model, a hierarchy of relaxation time scales was introduced: first the arms relax via free-end retraction inside the initial tube followed by a modified reptation of the inner backbone. Adding linear chains together with pompom molecules will affect both rheological behaviors. The model that is implemented in this study - the L-P model - takes into account all molecular parameters to predict both linear and non linear viscoelasticity. It is compared to some experimental results.

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Wednesday 5:00 Exhibition Hall / Poster Session

PO36

### Effect of a new multibranched polymer topology on melt rheological and foaming properties

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Polymer topologies have a significant impact on rheological melt and foaming properties. For instance, long-chain branching usually leads to shear thinning and strain hardening in elongation flow. In this study, a new star-like branch-on-branch polymer topology with longer arms for the first generation (>3 M<sub>e</sub>) and shorter arms for the second generation (1-2 M<sub>e</sub>) was synthesized to enhance the foaming abilities of polystyrene polymers. The molecular structure of this branch-on-branch stars consists of 3 to 6 armed in the first generation, with a number q of side chains grafted onto the end of each arm, as the second generation. The rheological and mechanical properties of this new topology were studied by synthesizing it with low dispersity via anionic polymerization and grafting-onto method. The chain length of the first generation arms are approximately  $M_{w,bb} = 50$  kg/mol. At each end of the first generation arms a similar number of side chains ranging from q = 3 to 6 (the second generation of arms) were grafted onto. The length of the second generation of chains was intentionally varied from unentangled to entangled chains with  $M_{w,a} = 7$  to 21 kg/mol. To determine the rheological and mechanical properties of the samples, we used small amplitude oscillatory shear (SAOS) and uniaxial elongation measurements. Through the uniaxial elongation measurements, the strain hardening factor (SHF) is determined, which is for this new polymer topology bigger than 20. We also investigated the foaming process with supercritical CO<sub>2</sub> and correlated the volume expansion of the foamed polymer sample to its molecular and rheological properties.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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**PO38** 

#### **Rheological properties of polystyrene blends comprising rings and matrices of different molecular structure** <u>Katerina Peponaki<sup>1</sup></u>, Daniele Parisi<sup>2</sup>, and Dimitris Vlassopoulos<sup>1</sup>

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The presence of free ends is known to affect the conformation and dynamics of macromolecules. Typically, entangled polymers are characterized by an intermediate rubbery plateau. In the special case of ring polymers, which have no free ends, there is not plateau and stress relxation proceeds self-similarly. At the same molar mass above the entanglement limit, rings relax fast their linear counterparts. Mixing ring and linear polymers results in threading, which maximes the overall entropy of the blend. Hence, understanding the dynamics blends involving rings added in different polymers and the role of branched polymers on the threading of rings is an interesting question with potential impact in tailoring materials properties by changing the architecture, i.e., entropically. Recently, we investigated the linear viscoelastic response of a series of symmetric (same molar mass) ring-linear polymer blends and revealed the important role of constraint release of the linear on the ring, leading to a non-monotonic composition dependence of the zero-shear viscosity. Here, we extend this work by presenting a systematic study of asymmetric ring-linear polymer blends, as well as blends involving rings added to a branched polymer matrix (H- or star-polymer). We assess the effects of constraint release and provide guidelines in terms of molecular parameters to tailor the viscosity of the blend and also, indirectly, control (enhance or reduce) threading events for complex macromolecular architectures. Our results should contribute toward a deeper understanding of constraint release effects and the design of polymeric composites with entropically tunable rheological properties.

#### Wednesday 5:00 Exhibition Hall / Poster Session

The rheology and scattering behaviour of Locust Bean Gum (LBG) in aqueous glucose solutions

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Locust Bean Gum (LBG) is a naturally occurring polysaccharide originating from the seeds of the carob tree. It is widely used e.g. in food, textiles, cosmetics and biopharmaceutical industries. LBG is a galactomannan composed of a linear backbone of mannose units, with added galactose sidegroups, and is typically characterised by a mannose to galactose ratio of approximately 4:1, where the variation in galactose content and distribution leads to a variation in solubility. The industrial interest in LBG is typically due both to its biocompatibility and its strong ability as a thickener. Also, it is known to demonstrate interesting phase behaviour such as the ability to form aggregates and gels upon long-term storage, freeze-thaw cycling, or mixing with other polysaccharides. Moreover, for many applications, the behaviour of LBG in the presence of co-solutes such as monoor di-saccharides (e.g. sugars such as glucose or sucrose), is of great importance, since the presence of sugars changes the hydrogen bonding properties of the solutions, and thus their solvent quality, the polymer aggregation, and flow behaviour. A better understanding of the impact of the presence of sugars in LBG solution behaviour is thus essential for the design of complex LBG-based formulations and thus key for a wide range of applications. Here, we use bulk and micro-rheology, as well as dynamic and static light scattering, to study the behaviour of LBG in aqueous glucose solutions. The rheology, structure and dynamics of the solutions are investigated for varying polymer concentrations, temperatures, and freeze-thaw processing conditions. The results will be compared both to data on aqueous LBG solutions, and to literature results for solutions of other polysaccharides.

Wednesday 5:00 Exhibition Hall / Poster Session

#### Effects of Monovalent and Divalent Cations on the Rheology of Entangled DNA

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DNA-based materials is an emerging field that utilises DNA as a material, rather than genetic, building block. A pioneer in DNA nanotechnology, Ned Seeman described using DNA as not only having the 'bricks' to build a material but also the 'mortar'. This dual role arises from DNA's capacity to function as a polymeric material and its ability to serve as an adhesive through complementary base pairing between two DNA strands. Since DNA is a negatively charged polyelectrolyte, it is well known that the presence of cations will encourage the hybridisation of DNA strands. However, the impact of cations on this process in solutions of concentrated DNA remains largely unexplored. This is significant for potential applications, such as DNA-based vaccines that would require administration at high concentrations [1]. In this talk, I will present our work studying a system of entangled lambda DNA, that possesses two ends that can bind to each other, allowing for the formation of longer chains through hybridization. This system can be described by Cates' reptation model of living polymers [2]. We investigate, how different valency and concentration of cations affect the rheology of entangled lambda DNA solutions and reveal that the distribution of chain lengths is influenced by the type and concentration of cations. Furthermore, we observe indications of intermolecular bridging between DNA strands caused by Mg2+ ions.

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#### Wednesday 5:00 Exhibition Hall / Poster Session Fatigue analysis of rHDPE and PE-PP blends via Fourier Transform of the stress on a modified rubber

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PO41

PO39

#### **rheometer** <u>Anika Goecke<sup>1</sup></u>, Valerian Hirschberg<sup>2</sup>, and Manfred Wilhelm<sup>1</sup>

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Nowadays polymeric materials have invaded nearly all areas of our lives. The most abundantly used polymers are polyolefins i.e., polyethylene (PE) and polypropylene (PP). They are mainly used for packaging and have a short lifespan, generating large amounts of plastic waste that pose a threat when discarded into the environment. Thus, recycling has become an essential aspect of waste management. Post-consumer waste typically consists of a mixture of different polymers due to difficulties during sorting. It is well known that the mechanical properties of PE-PP blends are reduced compared to the pure components due to the immiscibility of PE and PP.<sup>1,2</sup> However, the fatigue properties of PE-PP blends have not been investigated as intensively.

In this work the fatigue properties of PE-PP blends and mechanically recycled HDPE under oscillatory torsion and tension were analysed via the Fourier Transform (FT) of the stress to quantify nonlinear contributions as higher harmonics i.e.,  $I_{2/1}$ ,  $I_{3/1}$ .<sup>3</sup> The tests were performed in the solid state at room temperature on a modified rubber process analyzer (RPA) using rotational bone shaped specimen. The time evolution of the linear parameters (G'(t), G''(t)) and the nonlinear parameters ( $I_{2/1}$  (t),  $I_{3/1}$  (t)) as well as their derivatives and integrals were analysed to detect and describe specific events (e.g. crack growth and propagation), to predict the failure lifetime and to find failure criteria.<sup>1,2</sup>

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#### Wednesday 5:00 Exhibition Hall / Poster Session

Flow-induced by ultrasonic waves on non-degassed polydimethylsiloxane with different viscosity

<u>Manon Favre</u><sup>1</sup>, Emna Helal<sup>1</sup>, Sophie Miralles<sup>2</sup>, Simon Dagois-Bohy<sup>2</sup>, Valery Botton<sup>2</sup>, Éric David<sup>1</sup>, and Nicole R. Demarquette<sup>1</sup> <sup>1</sup>Department of Mechanical Engineering, École de Technologie Supérieure, Montreal, Quebec H3C1K3, Canada; <sup>2</sup>LMFA, UMR5509, Univ Lyon, INSA Lyon, CNRS, Ecole Centrale de Lyon, Univ Cl, Villeurbanne 69621, France

Ultrasound has found extensive application in polymer solutions for various purposes such as cross-linking or degradation of polymers, dispersions of particles, and more. The underlying phenomenon has been thoroughly investigated, elucidating the impact of ultrasound through cavitation. Essentially, the application of a high-frequency pressure cycle generates bubbles within the solvent that collapse forcefully. Researchers have also explored the use of ultrasound in molten polymers for similar objectives as in polymer solutions. They have observed polymer degradation, leading

to a reduction in molar mass, and a potential enhancement dispersion of nanoparticles. However, the mechanism behind these effects is still subject of ongoing discussion within the research community, given that molten polymers represent distinct fluids compared to polymer solutions.

This study delves into the impact of ultrasound on polydimethylsiloxane (PDMS) with varying viscosity. Ultrasound treatments were applied at different amplitudes (ranging from  $9\mu$ m to  $31,5\mu$ m) to PDMS with viscosities of 5, 30, and 300 Pa.s. The power consumption of the probe to apply a certain amplitude was recorded. Ultrasound treatment created a cloud of dissolved gas bubbles forms near the vibrating surface. The propagation of the bubbles, driven by the ultrasound beam, was filmed using an optical camera combined with a laser source. This setup facilitated visualization exclusively in the laser plane, specifically in the midplane of the probe. A Matlab program was used to extract the flow penetration.

The results unveiled distinctive flow behaviors depending on the polymer viscosity. For identical flow behaviors, an increase in viscosity decreased the flow penetration depth, however the power consumption was identical. Finally, the probe depth has also impacted the flow penetration depth and the power consumption. These findings will help to extend the study and to understand the effect of ultrasound on the molten polymers.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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# Rheological Assessments of the Preparation and Aging of CTAB/NaNO3 Solutions Targeting Reproducible Flows of Wormlike Micelles

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The peculiar rheology of shear-banding fluids is attributed to the emergence of elastic instabilities and turbulence (ET) in non-inertial flow regimes. The steady shear flow curves of such fluids are segmented into two dominant viscosity regions, linked with a mid-shear rate range plateau in the stress value. This setting leads to two sources of complexity in the flow of shear-banding fluids: (i) the segmentation of the field based on the dual-valued local viscosity, and (ii) ambiguity of the governed shear rates corresponding to the imposed shear stress, leading to velocity fluctuations. As a category of material exhibiting shear-banding characteristics, Wormlike Micelles (WLMs) formed by surfactants with extended and more flexible hydrophobic tails, are commonly studied for their rapid-alternating of the entangled chains of linear thread-like structures, reptating and bifurcating as a living micro-structure system, giving birth to elastic attributes of the fluid at macro-scale levels. Thus, WLMs are suitable candidates to investigate ET in canonical flow settings. Nevertheless, due to their non-equilibrium nature, they are extremely responsive to preparation protocols. They aggregate into elongated structures when electrostatic forces are screened by salts and a gel-like structure can be formed by their entangled network, giving the solution a higher viscosity and elasticity. The rheological properties of the resulting colloids are determined by the entanglement of elongated micellar structures, which are highly dependent on stress, temperature, surfactant concentration, salt addition, and can undergo complex aging phases. Here we address the asymptotical state curves of CTAB/NaNO3 solution as a commonly used shear-banding fluid prone to elastic instabilities, targeting reproducible experiments. Deviation from the Maxwell model is considered as a monitoring parameter for the state of preparation/deterioration. The findings propose reliable protocols for sample preparation under consistent flow conditions.

#### Wednesday 5:00 Exhibition Hall / Poster Session

**PO43** 

## Investigation of the effect of the gradual increase in strain in a step shear experiment on the molecular dynamics of hydrogels

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A combined rheology and low-field time domain (TD) NMR setup allows for simultaneous monitoring of molecular dynamics (<10 nm) by TD  $^{1}$ H NMR relaxometry and rheological properties and thus direct correlation of both. <sup>1</sup> In previous work this setup was used to study the relation between polymer segmental mobility and mechanical properties during hydrogel synthesis by measuring the T<sub>2</sub> relaxation under oscillatory shear deformation.<sup>2</sup> The measured T<sub>2</sub> relaxation curves are directly related to segmental motions in the KHz range.<sup>2,3,4</sup>

Based on this, the current research focuses on the investigation of the polymer segmental mobility of poly(acrylic acid) hydrogels under rheological deformation in the form of step strains in the linear and nonlinear viscoelastic regime. Preliminary results indicate, that applying strains of different magnitudes on the hydrogel network induces reorientation and stretching of the polymer chains, restricting segmental motion and leading to faster  $T_2$  relaxation times. But drying out of the samples by water evaporation during the measurement led to a less pronounced similar effect. The objective now is to prevent the samples from drying out in further experiments by using a custom-designed geometry, eliminating problems such as wall slip and testing different sample compositions. Therefore, poly(acrylic acid) hydrogels with varying parameters (degree of crosslinking, degree of neutralization, water content) are synthesized and subjected to logarithmically increasing step strains.

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#### Wednesday 5:00 Exhibition Hall / Poster Session

# Kinetik calculations for understanding the impact of thermo-mechanical treatment on the rheological properties of HDPE

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As consequence of the growing circular economy polymers should be processed multiple times. High stress, excessive temperature, and a long residence time in processing equipment (e.g. extruders) are therefore critical conditions, because thermal and mechanical degradation of the polymer material will occur. Therefore, it is important to understand how and which conditions during processing influence the degradation of polymers and to quantify this degradation process. A commercially available closed-cavity rheometer (CCR) is used, to emulate processing conditions, monitor the behaviour due to thermo-mechanical treatment of a high-density polyethylene and investigate potential degradation and changes of the polymer topology of the HDPE. The different treated samples were analysed rheologically by determining the crossover angular frequency and the activation energy of flow from in situ linear viscoelastic measurements at specific temperatures and reaction times during the treatment. The molecular characterization was performed by a high-temperature size exclusion chromatography (HT-SEC) afterwards. It was found that the simulated processing conditions by the CCR lead to an increase of the polymers molecular weight. The in-phase shear stress and the temperature were varied to investigate their specific impact on degradation. A model to calculate the change of the molecular weight is suggested, which is derived from the works of Goldberg et al. and Zhurkov et al. An activation energy for the initiation reaction of degradation of 267 kJ mol-1 without shear was found. This value is in good agreement with the activation energy for the HDPE degradation calculated from thermogravimetric analysis (TGA).

#### Wednesday 5:00 Exhibition Hall / Poster Session

Mechanical degradation of polymer solutions in lubricating mineral oil

<u>Salvatore Coppola</u><sup>1</sup>, Arcangela Russo<sup>2</sup>, Giusy Cozzolino<sup>2</sup>, Giovanni Ianniruberto<sup>2</sup>, and Giuseppe Marrucci<sup>2</sup> <sup>1</sup>Versalis SpA, Ravenna, Italy; <sup>2</sup>Department of Chemical, Materials & Production Engineering, University of Naples Federico II, Naples, Italy

Polymers are often used as rheology modifiers of lubricating mineral oils. A major issue in this application, is the mechanical degradation of polymer solutions caused by scission events in the polymer chains, when subjected to stress in complex flow fields. While the phenomenon is known and has been studied for some decades, the structure - property relationships are not crystal clear yet. In this work, we try to elucidate the different mechanical degradation of two polydisperse, linear Ethylene-Propylene random copolymers (EPR), characterized by two different propylene content. We prepared solutions in the semidilute concentration regime and subjected them to flow in a capillary rheometer, with the aim of causing degradation in a controlled way with a flow field taking place in a rather simple geometry. We adopted different capillaries, with varying L/D ratio, and different piston velocities. The degradation was characterized by online measurement of the pressure before the die entrance, and by offline measurement of the solution viscosity in rotational rheometer. Results from these two characterizations were not redundant as the online pressure measurement is also influenced by the degradation process itself and by possible non-isothermal effects. We observed effects of the shear flow in the capillary die and of the contraction flow at the die entrance. The solutions showed decreasing molar mass, and decreasing polydispersity index, with increasing capillary die L/D and piston velocity. At low piston velocity, L/D effect was insignificant, but the degradation was still evident. This means that at low piston velocity the degradation was dominated by the entry flow. Even if both the polydispersity and weight average fully stretched chain size of the two polymers were almost the same, we observed significantly different degradation for the two polymers. Eventually, a simple dumbbell model was implemented to gain a deeper understanding of the degradation phenomenon caused by the entry flow.

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**PO46** 

#### Wednesday 5:00 Exhibition Hall / Poster Session

Impact of aging on Chemical and Rheological Characteristics of self-healing asphalt binder

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Microcapsule-based self-healing asphalt binders have emerged as a promising technology for extending the lifespan and improving the sustainability of asphalt pavements. However, the long-term performance of these materials under aging conditions remains unclear. This study investigated the impact of the RTFOT aging on the chemical and rheological characteristics of self-healing asphalt binders containing silane microcapsules and recycled engine motor oil. The study employed various analytical techniques, including Fourier-transform infrared spectroscopy (FTIR), dynamic shear rheometer (DSR), and bending beam rheometer (BBR), to evaluate the changes in chemical composition, viscosity, and low-temperature cracking resistance. The results revealed that aging led to an increase in the carbonyl functional group index and hence, complex shear modulus (G\*), indicating the occurrence of stiffening and oxidation. Additionally, the self-healing ability of the binders decreased with aging, which can be attributed to the reduced mobility of the healing agent within the microcracks. Despite these changes, the self-healing binders retained a significant self-healing capability which can exceed the 93% even after aging, suggesting their potential for improving the durability of asphalt pavements. The study offers valuable insights into the long-term performance of self-healing asphalt binders and contributes to the development of more sustainable and resilient pavement systems.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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# Multiscale viscoelasticity of locust bean gum in aqueous and sucrose solutions as probed using bulk and microrheology

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Locust bean gum (LBG) is a high molecular weight polysaccharide of great industrial interest due to its natural origins and strong thickening ability. LBG is known to demonstrate interesting phase behaviour such as gelation upon long-term storage or freeze-thaw cycling, although uncertainties remain around its solution state properties. For example, existing literature shows sucrose-a common co-solute for LBG in industrial applications-to have a minimal impact on intrinsic viscosity and viscosity-concentration scaling while having a greater effect on gelation, a discrepancy which is not fully understood. To probe LBG solution behaviour, and the effect of sucrose, the linear viscoelasticity of LBG was measured in semidilute solutions with and without 40 % sucrose using a combination of bulk rheology, video particle tracking microscopy (PTM), dynamic light scattering (DLS), and diffusing wave spectroscopy (DWS). Good consistency was found between the techniques, which together accessed over six decades in frequency-revealing relaxation dynamics far beyond the terminal regime, as probed by bulk rheology alone. With increasing LBG concentration, the dynamics transition from predominantly Zimm-like to Rouse-like, with an additional elastic response contributing at the highest probed concentrations where physical entanglements become important. The presence of 40 wt % sucrose enhanced the complex specific viscosity and dynamic moduli within the terminal regime but had little impact on high-frequency dynamics, beyond the slowing caused by the greater solvent viscosity. Furthermore, the crossover into the terminal regime, where observed, occurred at longer timescales in the presence of sucrose. These observations were attributed to the kosmotropic effect of sucrose giving rise to strengthened and longer-lasting hydrophobic associations, which must dissociate for whole chain relaxation to take place. Our study suggests a potential origin for the enhanced LBG gelation behaviour typically observed in sucrose solutions.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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## The effects of liquid crystallinity and mechanical deformation on the molecular relaxations and viscoelasticity of liquid crystal elastomers

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Liquid Crystal Elastomers (LCEs) combine the anisotropic order of liquid crystals with the properties of elastomers, providing unique physical characteristics, such as stimuli responsiveness [1], and a recently discovered molecular auxetic response. [2,3] The LCE presented in this work

exhibits a molecular auxetic response in its nematic state, and chemically identical nematic or isotropic samples can be prepared by cross-linking. Hence, our LCE is an excellent system for studying the effects of liquid crystallinity on the properties of LCEs. We here determine how the molecular relaxation dynamics and viscoelasticity are affected by the LC phase using broadband dielectric relaxation spectroscopy, calorimetry and rheology. [4] We find that both the glass transition temperatures and dynamic fragilities are very similar in both LCE phases. Both LCE phases also demonstrate a cross-over in the structural relaxation behaviour at a similar temperature. For temperature below this cross-over, the behaviour becomes Arrhenius for the nematic phase LCE, but only more Arrhenius-like for the isotropic phase LCE; we present evidence supporting that this behaviour is due to pre-transitional fluctuations locked in during polymerization. Moreover, the effects of applied strain on both the relaxation dynamics and the mechanical response of the LCE is investigated; this is important since the molecular auxetic response occurs under elongational deformation. We find that for strains large enough to induce an auxetic response, significant changes in the structural relaxation and complex moduli are observed. Based on these results, we suggest that the molecular auxetic response is related to a strain-induced out-of-plane rotation of the mesogens, an interpretation which is consistent with relaxation and rheology data.

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Wednesday 5:00 Exhibition Hall / Poster Session

#### **3D Printed Liquid Crystal Elastomers**

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Liquid Crystal (LC) refers to phases of matter in between solid and liquid. Materials showing LC phases are said to be comprised of 'mesogenic molecules', often rod-like in shape, which can flow like a liquid while also possessing some crystalline qualities. A Liquid Crystal Elastomer (LCE) combines LCs and elastomers, where mesogenic groups are bonded to an elastomeric polymer network[1]. LCE networks can still exhibit orientational order and show properties from elastomers and LCs. More unique are the emergent properties like shape actuation[2] and energy dissipation. Energy dissipation is a crucial property of effective shock-absorbing materials. LCEs are, therefore, a promising new material to be used in protective gear, such as helmets[3].

A current challenge in the application of LCEs is their production. Monodomain-aligned LCEs are desired for their dissipation characteristics; however, they are traditionally produced as thin sheets to ensure quality alignment and molecular ordering[4]. 3D printing offers a solution to producing monodomain LCEs without traditional dimension restrictions. When 3D printing, the mesogens are shear aligned in the nozzle during extrusion so that alignment is ensured at the point of construction[5]. Although 3D printing monodomain bulk LCEs have shown success, more information is needed on how the print methodology and operational parameters, such as nozzle size and geometry, extrusion rate, print speed, and temperature, affect the flow and order of the LCEs and therefore, the LCEs properties and characteristics. This is what the project aims to understand. The poster will show my preliminary results for mechanical, physical and rheological tests of 3D-printed LCE components.

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Wednesday 5:00 Exhibition Hall / Poster Session

#### Phan-Thien-Tanner model applied to different materials extrusion

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Polymer processing applies trial and error strategies to improve process parameters and the quality of the final result. This has been rendered obsolete thanks to CFD simulations which exploit rheological models [1]. An interesting application of this field lies in Additive Manufacturing (AM). In AM, these models are applied to improve 3D simulations of Fused Filament Fabrication printing technique. One of the main issues with polymer melt during printing is focused on the ability of the melt to swell at the die exit. This has been proven to be caused by instabilities during processing. More recently, nonlinear differential models were applied for CFD simulations of the polymer melt. The Phan-Thien-Tanner (PTT) model in its exponential form, has proven multiple times to give the best fit for non-linear rheological properties of polymer melt [2][3]. In this work, we have selected three materials: Polypropylene (PP), Polyethylene terephthalate glycol (PETG) and Polylactic acid (PLA). The fit with the PTT model was employed for the data series depicting its validity. All the materials were compared based on the rheological properties that most influence the model parameter; namely the elongational viscosity and the first difference of normal stresses. This data together with SAOS measures were fitted to get the model parameter and the variation of this parameter was evaluated based on the polymer structure and flow properties. The second part of the work is dedicated to the 3D simulation of the material through a slit die. This allowed us to evaluate the swelling for the different systems.

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#### Wednesday 5:00 Exhibition Hall / Poster Session Effect of radical initiators on the phase structure and rheological properties of PLA/PHA blends Hyungsu Kim

#### Dankook University, Yongin, Gyeonggido, Republic of Korea

In the blend of poly(lactic acid) (PLA) and poly(3-hydroxybutyrate-co-4-hydroxybutyrate) (PHA), PHA alleviates the brittleness of PLA and improves processability due to the flexibility introduced by the 4-hydroxybutyrate repeating unit. A radical initiator added to the polymer meltmixing process extracts hydrogen from the polymer chain, initiating various modification reactions. The lifetime and stability of the radicals depend on the structure of the initiator, potentially leading to polymer decomposition or crosslinking. In this study, we introduced three types of radical initiators into the PLA/PHA blend to investigate changes in the phase structure and rheological properties of the blend. The selected radical initiators were AIBN (azobisisobutyronitrile), BPO (benzoyl peroxide), and DCP (dicumyl peroxide). In PLA, melt viscosity decreased due to decomposition reactions with AIBN and BPO, while it increased due to crosslinking reactions with DCP. Meanwhile, in PHA, the predominant effect of the initiators was a crosslinking reaction, leading to an increase in melt viscosity. The radical initiators, when added during the melt-mixing process of the PLA/PHA blend, acted as effective compatibilizers, reducing the size of the dispersed phase. Additionally, they significantly altered the rheological properties of the blend. Notably, the elongation viscosity of the PLA/PHA/DCP blend greatly increased compared to the PLA/PHA blend. This effect is expected to be beneficial in processing applications such as 3D printing.

#### Wednesday 5:00 Exhibition Hall / Poster Session

# The study of rheological properties of catalyst-free upcycled polyethylene terepthalate(PET) vitrimer from PET bottles

#### Seunghyeon Jin and Kyu Hyun

School of Chemical Engineering, Pusan National University, Busan, Republic of Korea

Polyethylene terephthalate (PET) is a thermoplastic material that is widely used in a variety of applications, including packaging, construction, and electronics [1]. Due to diverse applications, PET is a significant contributor to global solid waste. Recycling of PET is one of the important to solve the environmental pollution caused by plastic waste. Among plastics, PET bottles are currently being recycled the most. However, compared to other materials, the recycling rate is significantly lower, and the recycling life is short because the viscoelastic and mechanical properties of the raw materials are significantly degraded with each recycling. So, there are limitation when it comes to long-term recycling. In this study, we aimed to improve the processability and viscoelastic and mechanical properties of PET by introducing a dynamic network bonding. By combining PET matrix with tertiary amine structure and diepoxy in an eco-friendly method without using external catalysts, we upcycled PET bottles into a vitrimer that can be continuously reprocessed via torque rheometer. So, we took commercially available PET bottles and made them into flakes and then upcycled them. Rheological tests were conducted to investigate the linear viscoelastic and thermodynamic properties of the PET vitrimer as a function of crosslinking density by varying the amount of tertiary amine and diepoxy.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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# Evaluation of the Degree of Graphite Dispersion in Lithium-Ion Battery Anode Slurries Using Nonlinear Rheological Properties via FT-Rheology

#### Jeonghyeon Lee and Kyu Hyun

#### School of Chemical Engineering, Pusan National University, Pusan, Republic of Korea

The particle stability of lithium-ion electrode slurries is a key factor in manufacturing processes. Anode slurry is composed of conductive additives, active materials, binders, and a solvent, all of which affect particle dispersion and microstructure. Numerous studies have been conducted on the linear viscoelastic behavior of electrode slurries. However, slurries undergo exposure to exceedingly high shear rates and deformation during the production process. Consequently, investigation of nonlinear rheological behavior can provide a comprehensive insight into microstructural changes of electrode slurries under large deformation in the real industry.

This study focuses on the effects of different molecular weights of binders and different shapes of active materials on the nonlinear rheological properties of anode slurries. Materials used in this study are as follows: carbon black(CB) as a conductive agent, two different graphites(flake and spherical shape) as active materials, three carboxymethyl celluloses(CMCs,  $M_w$ =90, 250, 700 kg/mol) as binders, and water as a solvent.

We conducted a large amplitude oscillatory shear(LAOS) test to measure the nonlinear rheological properties of anode slurries. From the LAOS test, we classified various LAOS types of anode slurries, such as two-step thinning in G' and overshoot in G". In addition, via the Fourier Transform(FT) rheology, we calculated intrinsic nonlinearity( $Q_0$ ) and the nonlinear linear viscoelastic ratio(NLR) value for the anode slurries using various molecular weights of CMCs and different types of graphites. These approaches to characterize the nonlinear rheological properties enabled us to quantify and compare the degree of graphite dispersion in the CMC/CB suspensions.

To further observe the dispersion of particles, we investigated the morphologies of slurries and dried films using an optical microscope and field emission scanning electron microscope images, respectively.

### Wednesday 5:00 Exhibition Hall / Poster Session

#### Silica ionogels as smart materials for energy storage devices

<u>Carlos A. Gracia Fernández</u><sup>1</sup>, Antia Santiago<sup>2</sup>, Jose M. Antelo<sup>3</sup>, Villanueva María<sup>4</sup>, Juan J. Parajó<sup>5</sup>, and Salgado Josefa<sup>6</sup> <sup>1</sup>TA Instruments, Madrid, Madrid 28049, Spain; <sup>2</sup>2ABCR LABORATORIOS, Forcarei, pontevedra 36550, Spain; <sup>3</sup>ABCR LABORATORIOS, Forcarei, Pontevedra 36550, Spain; <sup>4</sup>NAFOMAT Group Instituto de Materiales iMATUS USC, Santiago de Composte, La Coruña 15782, Spain; <sup>5</sup>NAFOMAT Group Instituto de Materiales iMATUS USC, Santiago de Composte, A Coruña 15782, Spain; <sup>6</sup>NAFOMAT Group Instituto de Materiales iMATUS USC, Santiago de Composte, A Coruña

The efficient energy storage, mainly electric batteries or supercapacitors, has emerged as a crucial element in transforming our world towards a more sustainable and clean energy-focused society. Currently, the reliance on renewable energy sources such as solar and wind has created a critical need for efficient and reliable energy storage systems. Smart electrochemical devices have become the backbone of this energy transition, enabling the storage of energy generated during surplus periods and releasing it when renewable energy sources are unavailable. The search for new materials is essential to enhance storage capacity, lifespan, and the sustainability of these devices. Innovations in nanotechnology, composite materials, and advanced designs are playing a crucial role in this process. The need to find alternatives to traditional materials, such as lithium, is driving research towards more abundant and environmentally friendly components. Optimizing materials not only improves device efficiency but also contributes to reducing dependence on scarce resources and mitigating the environmental impacts associated with their extraction and production. The design of the novel electrolytes for commercially available energy storage devices is one of the major scientific challenge nowadays. The characteristic properties of ionic liquids (ILs) as their extremely high electrochemical and thermal stabilities, their non-flammability and low vapour pressure, their high ionic conductivity, which can be tuned by means of the addition of cosolvents, make them optimal candidates for energy applications, as, for example, electrolytes for next generation high-voltage batteries, advanced supercapacitors or last generation fuel cells. However, liquid electrolytes present important problems of safety and performance and recent studies suggest the possibility of the confinement of these fluids in gel matrices, achieving high mechanical stability while preserving liquid state transport properties. This work includes the proposal of a

#### Wednesday 5:00 Exhibition Hall / Poster Session **Decisive External Factors Affecting the Electrorheological Effect of Waxy Oil** Hongying Li, Yiwei Xie, and Jinjun Zhang

China University of Petroleum Beijing, Beijing, China

Waxy crude oil exhibits poor flowability at normal temperature during pipeline transportation, posing great threats to flow assurance, such as high energy consumption, restart failure after extended shutdown, etc. Improving the flowability of waxy crude oil is the fundamental solution to flow assurance problems. Electric treatment is an emerging approach to improve the cold flowability of waxy crude oils with high efficiency and low consumption, whose basic physical principle is the negative electrorheological effect of crude oil. The impacts of the applied external parameters on electrorheological effects have been thoroughly studied. The rheological behaviors of crude oil respond quickly to an electric field greater than 0.2 kV/mm, the viscosity can be reduced immediately by applying electric field to oil sample for seconds to minutes, whether for static or flowing oil sample, and whether for electric field parallel to the flow direction or perpendicular to the flow direction. Comparatively, the electrorheological effect of an AC electric field is somewhat weaker than that of a DC field under equivalent conditions of treatment time and temperature. Subjecting a waxy oil to varying strengths (0.1~5 kV/mm) for sufficient time achieves a consistent maximum viscosity reduction at the same temperature, regardless of the applied field strength. An increase in the energy input per unit volume of the oil sample corresponds to an enhanced reduction in viscosity. The inputted energy density is the essential factor determining the viscosity reduction. The energy inputted to the system should work in three stages. Initially, it is used to overcome the energy gap between charged particles and wax particles, causing charged particles to accumulate on the wax particle surfaces and initiating viscosity reduction. The continued inputted energy promotes more particle cohesion until a maximum viscosity reduction is achieved. After that, additional inputted energy will further extend the duration of the viscosity reduction.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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**PO57** 

**Rheological characterization of superplasticizers in self-levelling smoothing compound for flooring applications** Emanuele Carlini<sup>1</sup>, Stefano Carrà<sup>1</sup>, Fabio Curto<sup>2</sup>, and Laura Pellicano<sup>2</sup>

<sup>1</sup>R&D Rheology Lab, Mapei Spa, Milano, MI 20158, Italy; <sup>2</sup>R&D Rheology Lab, Mapei Spa, Milano, MI 20158, Italy

Self-levelling, ultra quick-hardening smoothing compounds are used in interiors for levelling and smoothing differences in thicknesses from 1 to 10 mm on new or existing substrates, preparing them to receive different kinds of floorings. Self-levelling compounds consist in a powder made of special cements, with rapid setting and hydration, and a special admixture of additives. Mixed with water, this powder becomes a fluid and easily workable mortar, perfectly self-levelling, and ultra-fast drying.

Superplasticizers are the most important additives in this kind of formulation. Their purpose is to lower both the viscosity of the mortar and the required water amount, thus increasing spreadability, self-levelling and final mechanical properties of the compound.

Therefore, a rheological characterization of the effect of these superplasticizers during the time from mixing to setting time is key to develop this kind of formulations. This time corresponds to the early stage of the cement hydration. The final mechanical properties of the mortar are highly dependent on the different crystalline phases that are formed in the first hydration steps.

In this work, the effect of different superplasticizers was evaluated by using rheological analysis; in one case, the percentage of additive was also varied to determine the influence of its quantity. Both continuous and oscillatory measurements were performed with a rotational rheometer. Flow

curves were used to obtain information about the initial workability. The time sweep test turned out to be especially useful to provide important information on the setting time and on kinetics of the hydration reaction.

Another important property of self-levelling compounds is called "healing"; it is the ability to obtain a flat final surface on two consecutive pours, while the product is still fresh. Healing was evaluated by using a rheological measurement protocol, which allows to obtain more precise information with respect to traditional empirical tests.

#### Wednesday 5:00 Exhibition Hall / Poster Session

Rheological studies of thermally reactivated cements mixed with varying water-cement ratios

Susanne Helmich, Neshable Noel, Anne Z. Gierth, Stefan Nawrath, Tommy Mielke, and Doru C. Lupascu Institute for Materials Science, University of Duisburg-Essen, Essen, North Rhine-Westphalia 45145, Germany

One of the most prominent challenges of today's world is the reduction of CO<sub>2</sub>-emissions. A promising approach to lower the carbon footprint of the construction industry is based on the reactivation of cement stone from concrete waste via thermal treatment and thus recovering the hydration ability. Various researchers have noted that the water demand of reactivated cement is higher than for industrially produced Portland Cement (OPC). However, the quantification of the rheological properties of rehydrated reactivated cements has been neglected. This research sets the objective to improve the understanding of the rheological behavior of reactivated cements and intends to create a data basis as a reference point in further research. Reactivated cement powders have been produced using different reactivation temperatures and have then been mixed with varying water/cement-ratios to obtain the cement pastes for measurement. The rheological parameters were determined in a rotational rheometer and evaluated according to the Bingham model. Additionally, spread flow measurements using a Haegermann cone as per DIN EN 1015-3 were performed with each prepared cement paste. The results highlight the differences in water demand between OPC and reactivated cements. It can be observed that the water demand increases with the reactivation temperature at which the cement has been treated.

#### Wednesday 5:00 Exhibition Hall / Poster Session

# Investigating the effect of agitator geometry on the performance of sawtooth impellers for processing high solid pastes

<u>Shreyasi Deshpande</u><sup>1</sup>, Shreyasi Deshpande<sup>2</sup>, Zoe Berisford<sup>1</sup>, Joseph Mills<sup>3</sup>, Thomas Abadie<sup>2</sup>, Andrew Ingram<sup>2</sup>, Bettina Wolf<sup>2</sup>, and Jonathan O'Sullivan<sup>1</sup>

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Formulated products exhibiting characteristics of pastes, as used in the personal care industry, are liquid systems with a high-volume fraction of undissolved solid particulates. Sawtooth impellers are a mixer geometry commonly used in the processing of such multi-component formulated products possessing complex non-Newtonian rheology. They are especially useful in manufacturing processes where high shear is required to break-up powder agglomerates. It is important to understand which aspects of the mixer geometry drive this function and the associated power draw for process understanding and optimisation. In this study, the effect of the mixer geometry (height and number of teeth) on the power draw characteristics of sawtooth impellers is studied, as a function of fluid rheology.

Custom mixer geometries were 3D printed and the torque capabilities of a rotational rheometer were used to scale-down a mixing setup. Tests were conducted using a glycerol solution (60 wt. %) and a Carbopol solution (1.5 wt. %), and the relationship between the dimensionless power number and Reynolds number was established for twelve geometries. Results show a decreasing power draw as rotational speed increases, and this trend holds across the transitional and turbulent regimes. Increasing the number of teeth increases the power draw. This increase is more pronounced from 0 to 8 teeth and is attenuated from 8 to infinite teeth.

Establishing the torque response as a function of rotational speed, geometry and rheology is to assist in the development of accurate computational fluid dynamics models. The resulting flow visualisation can help understand shear rate profiles and the dynamics of powder break-up during the manufacture of multi-component formulated paste products.

Wednesday 5:00 Exhibition Hall / Poster Session

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#### Large Amplitude Oscillatory Shear of Elastoviscoplastic (Saramito) fluids

Rishav Agrawal<sup>1</sup>, Esther García-Tuñón<sup>1</sup>, Robert J. Poole<sup>1</sup>, and Claudio P. Fonte<sup>2</sup>

<sup>1</sup>School of Engineering, University of Liverpool, Liverpool L69 3GH, United Kingdom; <sup>2</sup>University of Manchester, Manchester, United Kingdom

Direct ink writing (DIW) is a 3D printing technique which requires the printing materials to have both yield stress and shear thinning properties so that they can flow through narrow nozzles and recover their original strength after printing to support the printed structure[1]. In the DIW community, these yield stress fluids are commonly modelled using the simple Bingham or Herschel-Bulkley constitutive models, which can simulate the yield stress effects but do not consider the time-dependent/viscoelastic behaviour that some of the inks can exhibit. There have been recent developments of constitutive equations that incorporate both viscoelastic and plastic (yield stress) effects, the so-called elastoviscoplastic (EVP) models. An EVP model proposed by Saramito[2] which describes the unyielded state of the material as a Kelvin-Voigt solid and the yielded state as a non-linear viscoelastic material, has been widely implemented for various flow simulations[3] and has the potential to be utilized for modelling DIW processes. With this aim, we numerically investigate the large amplitude oscillatory shear (LAOS) of EVP fluids using the

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Saramito model. The LAOS study is carried out by applying an oscillatory shear strain and then the resulting stress response is studied for varying relevant dimensionless parameters, Weissenberg number, Deborah number, Bingham number (Bn) and solvent viscosity ratio. The first-order storage and loss moduli are in good agreement with the theoretical values in the limiting cases of Bn = 0 (viscoelastic Oldroyd-B model) and Bn = 8 (Kelvin-Voigt-like response). We quantify the non-linearities in the stress response using FT-rheology[4], the plastic dissipation ratio[4], and the sequence of physical processes[5] framework.

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#### Wednesday 5:00 Exhibition Hall / Poster Session

#### Fluid-mediated impact of soft solids

<u>Jacopo Bilotto</u><sup>1</sup>, Joaquin Garcia-Suarez<sup>2</sup>, John M. Kolinski<sup>3</sup>, Brice Lecampion<sup>4</sup>, Jean-François Molinari<sup>5</sup>, and Ghatu Subhash<sup>6</sup> <sup>1</sup>Civil Engineering, EPFL, Lausanne, Vaud 1015, Switzerland; <sup>2</sup>Civil Engineering, EPFL, Lausanne, Vaud 1015, Switzerland; <sup>3</sup>Mechanical engineering, EPFL, Lausanne, Vaud 1015, Switzerland; <sup>4</sup>Civil Engineering, EPFL, Lausanne, Vaud 1015, Switzerland; <sup>5</sup>Civil Engineering, EPFL, Lausanne, Vaud 1015, Switzerland; <sup>6</sup>Mechanical and Aerospace engineering, University of Florida, Gainesville, FL 32611, United States

A viscous, lubrication-like response can be triggered in a thin film of fluid squeezed between a rigid and flat surface and the tip of an incoming projectile. We develop a comprehensive theory for this viscous approach stage of fluid-mediated normal impact, applicable to soft impactors. Under the assumption of mediating fluid being incompressible, the impacting solid displays two limit regimes: one dominated by elasticity and the other by inertia. The transition between the two is predicted by a dimensionless parameter, which can be interpreted as the ratio between two time scales that are the time that it takes for the surface waves to warn the leading edge of the impactor of the forthcoming impact, and the characteristic duration of the final viscous phase of the approach. Additionally, we assess the role of solid compressibility and elucidate why nearly-incompressible solids feature (a) substantial "gliding" prior to contact at the transition between regimes, (b) the largest size of entrapped bubble between the deformed tip of the impactor and the flat surface, and (c) a sudden drop in entrapped bubble radius past the transition between regimes. Finally, we argue that the above time scale ratio (a dimensionless number) can govern the different dynamics reported experimentally for a fluid droplet as a function of its viscosity and surface tension.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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### **Particle equilibrium positions in the microchannel flow of the Carbopol solution, a model yield stress fluid** Daekwon Jin<sup>1</sup>, Ju Min Kim<sup>1</sup>, and Jaewook Nam<sup>2</sup>

<sup>1</sup>Department of Energy Systems Research, Ajou University, Suwon, Republic of Korea; <sup>2</sup>Department of Chemical and Biological Engineering, Seoul National University, Seoul, Republic of Korea

Yield-stress fluids are complex rheological materials that exhibit a shift from soli-like to fluid-like properties. In recent times, yield-stress fluids have been found in a variety of industries, such as cement slurries, food, and lithium-ion batteries. Furthermore, most of these fluids contain particles and are subject to shear and elongational flows, or their combined flows, during material processing. Meanwhile, particles suspended in a liquid experience forces in a direction perpendicular to the flow direction due to particle-particle or particle-fluid interaction, resulting in the spatial heterogeneity of particles. Because this heterogeneity alters the rheological properties of the suspension and acts as an irregularity in liquid-flow processes, it is vital to study particle dynamics in various flow conditions. In this study, we investigate particle migration in the microchannel flow of yield stress fluid. Carbopol solution, which is a transparent fluid with yield stress for optical observations, was used as a suspending medium. The spherical polystyrene particles were suspended at dilute concentrations (0.1 wt%) to minimize the particle-particle interaction. For precise investigation, we used a dual-view imaging system that was able to reconstruct the particle cross-sectional distribution by observing the particle in vertical and horizontal directions. From this system, we observed that the particles in the carbopol solution migrated to the corners in a square microchannel and to the wall in a circular microchannel. Interestingly, these equilibrium positions of particles are unique compared to those in other viscoelastic fluids, such as Boger fluid and shear-thinning fluid. It seems that flow instability due to the microstructure change of the carbopol solution in the shear flow induced these unique distributions. We anticipate that our findings will contribute to understanding the flow behavior of particulate-complex fluid systems.

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### **Exploring the relative importance of strain rate and strain in governing yielding in oscillatory experiments** <u>Manuel Helfer<sup>1</sup></u>, Hans M. Wyss<sup>2</sup>, and Veronique Trappe<sup>1</sup>

<sup>1</sup>Department of Physics, University of Fribourg, Fribourg, Fribourg 1700, Switzerland; <sup>2</sup>Microsystems Section, Department of Mechanical Engineering, Eindhoven University of Technology, Eindhoven, Noord Brabant 5612 AE, The Netherlands

We investigate the mechanical properties of a dispersion of Carbopol-microgels in propylene glycol. This dispersion belongs to the class of yield stress materials, which are materials that are solids when at rest, yet flow when the shear stress applied exceeds a certain threshold value.

In this work we explore the yielding process by applying a sinusoidal strain of varying amplitude and compare the results with those obtained by applying triangular oscillations, in which the magnitude of the shear rate is maintained constant.

Our experiments reveal that the yielding condition are not only determined by strain, but also by strain rate. Moreover, we find that the elastic load within the system becomes strain-independent at large enough strains.

Wednesday 5:00 Exhibition Hall / Poster Session

Network modelling of yield-stress fluid flow in randomly disordered porous media

Elliott M. Sutton<sup>1</sup>, Kohei Ohei<sup>2</sup>, Maziyar Jalaal<sup>3</sup>, Yuji Tasaka<sup>4</sup>, Claudio P. Fonte<sup>5</sup>, and Anne Juel<sup>6</sup>

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Yield-stress fluid flow occurs in many industrial applications, including in porous systems during enhanced oil recovery and membrane emulsification. However, the high computational cost of computational fluid dynamics (CFD) simulations for yield-stress fluids hinders the study of these large-scale systems. In this work, we develop a fully predictive network model for flow through 2D disordered porous media that obtains a solution in minutes on a single CPU core, considerably faster than the days or weeks required by CFD with multiple CPU cores. Our model generates network nodes using a Voronoi tessellation, extracts geometric parameters accounting for the topology of the domain, and numerically solves mass and momentum conservation equations with a Newton-Raphson solver. We accurately predict the pressure drop and velocity field of flows with and without wall slip, validating our results against CFD simulations produced in-house and from the literature. Wall slip is ubiquitous in real-world flows of these materials, dramatically decreasing friction at solid-fluid interfaces and changing the overall resistance in porous networks. Thus, it was imperative to account for this phenomenon in our model.

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#### Wednesday 5:00 Exhibition Hall / Poster Session

#### Rheological properties of clay-polymer composite hydrogels for bioremediation

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Contamination by heavy metals poses many risks to the ecosystem (e.g., soil, aquifers) and humans, affecting food chain safety, food quality, and the ability to use the land for agricultural production. Despite the presence of high concentrations of heavy metals, highly contaminated environments are reservoirs of heavy metal tolerant microbes with biotechnological application in bioremediation. In particular, clay minerals from the group of bentonites are characterized by the presence of diverse groups of microbes including bacteria (such as Stenotrophomonas bentonitica) with high heavy metal tolerance and attractive potential in the recovery of critical elements (selenium, uranium, lead, etc.). Within this context, in this work we describe the synthesis and rheological characterization of alginate-cellulose hydrogels at different polymeric proportions, with best results for 30/70 alginate/cellulose proportion and final concentration of 1%wt. For this concentration, we added bentonite at 1%wt to encapsulate S. bentonitica cells. A hydrogel precursor was created by adding CaCO3 in combination with D-glucono-d-lactone . Finally, hydrogels were created by additional cross-linking with CaCl2 solution. Hydrogel of this composition were used to perform selenite (Se (IV)) bioremediation assays, which showed the formation of less toxic Se(0) nanoparticles. The rheological properties under shear of the hydrogels were analysed for the different conditions before and after inoculation with bacteria. Moreover, traction and compression assays were also carried out to completely characterize the mechanical properties of the hydrogels. The results were related with microstructural changes analysed by Scanning Electron Microscopy.

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#### Wednesday 5:00 Exhibition Hall / Poster Session **Designing athermal disordered solids with automatic differentiation** Mengije Zu

#### Institute of Science and Technology Austria, Klosterneuburg, Austria

Functional material is one of the essential goals of material science, which has been revitalization by taking advantage of the advanced development of nanotechnology and machine learning in the last few decades, particularly in the framework of inverse design. Given a desired functionality or structure and design parameters, inverse design searches for optimal solutions in the landscape of objective function. This approach is popular in the design of the self-assembly of multi-component systems to specific structures, while the complex and ambiguous relation between building blocks and properties hinders the inverse design of the specific property of materials directly. Here, we develop an effective and robust approach for the design of disordered solids with target properties, particularly one of the intrinsic mechanical properties - Poisson's ratio, by tuning model parameters. These parameters are adjusted by the gradients information, which directly exhibits the relationship between the property and the interaction between components. The explorations of various target Poisson's ratios and the number of components demonstrate the effectiveness and accuracy of our approach. Moreover, our approach allows us to easily manipulate different properties simultane- ously, such as Poisson's ratio and pressure, providing a promising avenue to design multi-functional materials.

**PO70** 

#### Wednesday 5:00 Exhibition Hall / Poster Session

#### How does synergy play a role in the mechanics of a double-network hydrogel?

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Composite materials are found almost everywhere in nature. The synergistic effect of collagen, which is good at absorbing and dissipating energy but is highly inextensible, and elastin, which is extensible but very poor at dissipating energy, helps tendons in our body transfer force from muscles to bone structure. Double-network (DN) hydrogel is a soft and tough material consisting of two interpenetrating polymer networks with contrasting properties [1]. It provides a way for us to design materials that can use the same synergistic effect for various applications. However, due to a lack of rheological studies, the fundamentals of the non-linear mechanics of double-network hydrogels are still not well understood, which prevents the design of materials for advanced practical applications.

In this poster, we illustrate the pivotal role played by individual networks in shaping the non-linear properties of a DN (double network) hydrogel, employing Large Amplitude Oscillatory Shear (LAOS) rheology [2]. Our presentation highlights the intricate interplay between the two networks, revealing the synergistic impact on the linear characteristics and failure mechanics of a DN hydrogel.

#### References:

 Gong, J. P., Katsuyama, Y., Kurokawa, T., & Osada, Y. (2003). Double-Network Hydrogels with Extremely High Mechanical Strength. In Advanced Materials (Vol. 15, Issue 14, pp. 1155-1158). Wiley. https://doi.org/10.1002/adma.200304907
 Ewoldt, R. H., Hosoi, A. E., & McKinley, G. H. (2008). New measures for characterizing nonlinear viscoelasticity in large amplitude oscillatory shear. In Journal of Rheology (Vol. 52, Issue 6, pp. 1427-1458). Society of Rheology. https://doi.org/10.1122/1.2970095

Rheological behaviour of syneresis evolution in HydroxyPropyl(Methyl)Cellulose (HPMC) hydrogels doped

#### Wednesday 5:00 Exhibition Hall / Poster Session

PO73

PO74

### with porous silica particles

### Saray Perez-Robles, Claudia Carotenuto, and Mario Minale

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Hydroxypropyl Methylcellulose (HPMC) undergoes hydration when introduced into cold water, transitioning into a gel when heated. This thermogelation process is reversible, allowing the gel to return to its sol state as it cools down. Porous particles can be added to the gel with possible applications to, e.g., drug release, water remediation, and enhancement of oil recovery. Silica porous particles are used at different concentrations, from the very dilute ones to moderate concentrations, where particle-particle interactions cannot be neglected, and a percolated particle microstructure can be obtained. The effect of the porous particles is investigated following the hydrogel viscoelastic changes during the thermogelation with SAOS tests with an ARES-G2 rheometer (TA Instruments). Thermal ramps from 30 to 90°C using the multiwave method were run to obtain isochronal frequency sweeps during heating. The gel point and the final gel strength were estimated with the Winter-Chambon and Friedrich Heymann criteria. In addition, the thermal stability of the different gels is investigated by following the syneresis process at 90 °C for 14 days. Syneresis involves the contraction of the gel, releasing liquid. At days 0, 2, and 7 the expelled liquid is analysed in terms of density and viscosity measurements with a combined densimeter/rolling-ball viscometer Lovis DMA 4500 (Anton Paar), while the viscoelasticity of the hydrogel exposed at 90°C is investigated by cooling it down and subsequently following its thermogelation upon heating with SAOS tests. Results showed that adding only 1% of silica porous particles reduced syneresis by about 10%.

Wednesday 5:00 Exhibition Hall / Poster Session

#### Rheological innovative analysis of CNPs coatings

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Carbon nanoparticles (CNPs) are extremely small carbon-based particles with dimensions typically ranging from 1 to 100 nanometres. The nanometric size and fractal structure endow these nanoparticles with properties that promote applications across various disciplines such as materials science, electronics, medicine, and environmental science. In particular, in this work CNPs are used to yield coatings with specific behaviors, particularly for superhydrophobic surfaces. The coatings were produced through the deposition of CNPs generated in a rich flame onto a selected substrate. This substrate is inserted intermittently into the flame to allow nanoparticle capture and deposition by thermophoresis. This phenomenon occurs thanks to the thermal gradient of approximately 10^6 K/m between the flame (approximately 1800 K) and the substrate, considered at room temperature given the very short flame insertion time. The resulting films are made up of an aggregation of CNPs, randomly arranged on the substrate, whose physical properties vary based on the collection mechanisms and flame conditions. To assess the wettability of the samples produced using the described experimental procedure, the contact angle was evaluated with a goniometer using the "sessile drop" method. To establish a correlation between the flame conditions and the nanostructure of the film, tests were conducted with different liquids characterized by different properties such as surface tension, density and viscosity. Furthermore, through innovative techniques that involve the use of a rotational rheometer, it is possible to appreciate the "self-similar" structure resulting from thermophoretic flame deposition and evaluate the interparticle adhesion, a crucial aspect to be taken into account for the practical use of this kind of coatings.
#### PO75

**PO76** 

**PO77** 

#### Wednesday 5:00 Exhibition Hall / Poster Session

An investigation of Liver Viscoelasticity through Dynamic Mechanical Analysis

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Advanced in-vitro liver models serve as valuable tools for advancing pharmacological research. These models imply the growth of liver cells within a 3D scaffold which aim at mimicking the complex biochemical and mechanical properties of the liver. Considering the mechanical behaviour, disparate results are reported due to variations in testing conditions and sample origins. This study proposes an analysing the viscoelastic properties of liver by applying dynamic mechanical analysis, both in shear and compression. Porcine livers were sourced from local butchers and underwent testing within 24 hours of slaughter. Both physiological livers and livers affected by either steatosis or hepatitis were considered. Dynamic mechanical analyses in shear were performed using an Anton-Paar MRC502 rheometer; a TA RSAIII dynamic analyser was used for the characterization in compression. In both cases, a 25mm plate-plate geometry was adopted. All tests were performed in the linear regime, at 37°C. To assess intra-species variability, multiple livers were examined and the effects of freezing and thawing on the mechanical properties were investigated. The analysis revealed no significant differences between sample depending on orientation or sampling position. Liver tissue exhibits solid-like behaviour. Interestingly, G' varied between 0.1-0.4kPa and E' between 30-35kPa. This unexpected difference is probably due to the complex liver structure, that may modelled as a foam, with closed cells filled with an incompressible liquid. Finally, dynamic mechanical characterization in shear allowed to distinguish between physiological, steatotic and hepatitic livers; compressive tests instead allowed to distinguish hepatitic livers from physiological ones, while stetotic livers have the same properties than the physiological ones.

#### Wednesday 5:00 Exhibition Hall / Poster Session

# **3D** bioprinting for building spatially distributed microbial populations

<u>Shagufta Rehmat</u>, Daniele Tammaro, and Pier Luca Maffettone Università degli Studi di Napoli Federico II, Naples, Italy

Microbial consortia refers to a community of different microbial species that interact and function together. In nature, microbial consortia thrive in a variety of habitats, including biofilm, soil, and food products. Traditionally mixed culturing techniques are used to produce these communities however, these conventional co-culturing techniques have been insufficient in the understanding of the true nature of interaction and communication, and how an equilibrium is achieved between most microbial communities. This study aims to design and construct a sustainable microbial consortium that mimics the natural spatial distribution of the micro-organisms using the 3D printing technique. To achieve this, a state-of-the-art printhead is designed along with a microbe-laden hydrogel formulation (Lactobacillus species and Saccharomyces cerevisiae are used for formulating the hydrogel attributing to their co-fermentation abilities to produce sour beer). The rheological properties of the microbe-laden hydrogel provide insights into the behavior of a printed microbial community in the context of rheology can offer insights into a) the effects of microbes on the intricacies of hydrogel and b) how changes in the hydrogel formulation can alter microbial interactions within a complex matrix. To evaluate the production efficiency of the microbial community created using the state-of-the-art printhead, a comparative analysis will also be conducted with its counter-microbial community created using conventional techniques. In conclusion, this study is expected to play a significant role in advancing the creation of intricate polybacterial microsystems, tissue-microbiota interactions, and the production of bioengineered materials with increased complexity.

Wednesday 5:00 Exhibition Hall / Poster Session

# Rheological insight on 3D printable biomaterials

<u>Mercedes Fernandez-San Martin</u><sup>1</sup>, Itziar Insua<sup>2</sup>, Itxaso Calafel<sup>2</sup>, Robert Aguirresarobe<sup>2</sup>, Juliana Nunes<sup>3</sup>, Damien Dupin<sup>3</sup>, and Arantxa Acera<sup>4</sup>

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The dynamic covalent bonds are mechanically stable but capable of exchanging, dissociating, or switching in response to external stimuli, providing the material with a responsive and adaptive nature, which gives hydrogels self-healing properties, injectability and suitability for postprocessing and additive manufacturing. This study investigated the dynamic response of dynamic hydrogels based on thiol-functionalized hyaluronic acid formulated with coordinative gold (I) or silver (I), that meet all the requirements to be printed without the need for external stimuli [1]. At rest, the hydrogel viscoelastic network was characterized by good macroscopic mechanical stability. The structure evolved to a liquid network under strain, facilitating the deformability and printability of these materials. The rheological analysis revealed different behavior depending on the disulfide, gold-and silver-thiolate interactions existent in these hydrogels. Oscillatory shear in the linear viscoelastic regime and steady flow curves accounted for the soft-solid and still liquid nature of the network. In superposition experiments, the steady and oscillatory shear motions were applied simultaneously, which allowed the detection and quantification of the flow induced acceleration of the network's relaxation modes. The specific signature of the large amplitude oscillatory shear (LAOS) was also dependent on the different interactions. Analysis by Fourier Transform Rheology (FTR) showed the highest intrinsic nonlinear response of Au-thiolate interactions compared to Ag-thiolate. The interpretation in terms of the known nonlinear elastic behavior (stiffening-softening) and the nonlinear viscous behavior (thickening-thinning) was also investigated by means of stress signal decomposition (SD) methodology and Chebyshev polynomials. The nonlinearity parameters provided useful information related to the printability criteria of these materials. [1] Aitor Díaz et al., Macromol. Rapid. Commun. 2022, 43 (21), e2200449, doi: 10.1002/marc.202200449

Wednesday 5:00 Exhibition Hall / Poster Session

Applying Cryo-SEM with in-situ rheology measurements synergistically

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Bovine serum albumin (BSA), a globular protein, photochemically crosslinks via dityrosine bonding to form a hydrogel exploiting protein function [1]. Disulphide bonds hold the folded state of BSA together which is altered by reducing agents like dithiothreitol (DTT) or denaturing agents like urea causing complete or partial protein unfolding respectively. This control over protein unfolding during network formation enables the design of materials with distinct properties. Mechanical characterisation using a stress-controlled rheometer in parallel plate configuration shows a 4-fold increase or 6-fold decrease in storage modulus G' due to complete or partial protein unfolding respectively (from 1.9 kPa to 7.5 kPa or 0.3 kPa). The loss ratio, energy dissipation and relaxation behaviour all vary significantly with manipulating protein unfolding. BSA-based systems have been characterised by small angle scattering (SAS) techniques that indirectly measure the network structure [1-3]. This study will perform Cryo-Scanning Electron Microscopy (Cryo-SEM) to directly image the hierarchical architecture of protein-based networks from macro-pore sizes, matrix wall thickness and potentially down to folded protein distributions within the matrix. DTT and urea were used to alter the BSA folded state before gelation. Cryo-SEM revealed that all networks have a macro pore structure altered by manipulating protein (un)folding. A folded protein gel displays the smallest average pore areas  $(1.8 \ \mu m^2)$ , while protein unfolding forms larger average pore gels  $(16.1 \ \mu m^2)$ . Cryo-SEM indicates the folded protein (un)folding in network formation and hydrogel mechanics. Our structure-property analysis aims to enhance the performance of these gels in biomedical applications.

1.ACSNano,2021,15,11296 2.SoftMatter,2020,16:6389 3.Biomacromolecules,2020,21:4253

# Wednesday 5:00 Exhibition Hall / Poster Session

Motion of Semi-Flexible Fibres Under Shear Flow

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The cylindrical fluctuating semi-flexible fibre (as a representation of a semi-flexible fibre in cell's cytoskeleton) immersed within a fluid can bind to other filaments via some mechanisms like cross-linking and span the whole system to construct a fibrous random network. The shear stress imposed by the flow has large effects on the rheology of the immersed fibrous network and makes it behave similar to viscoelastic media. In this research study, Slender Body Theory (SBT) with the Rotger-Prager-Yamakawa (RPY) singularity representation have been implemented to simulate the fluid-structure interaction of immersed semi-flexible fibres under shear stokes flow, where the fibres are cross-linked by small springs representing, e.g. scruin or filamin proteins. For rigid fibres, the model results have been quantitatively validated against the known solution of Jeffrey Orbits. For single flexible fibres it is found that fibres undergo a variety of buckling regimes (and configurations) depending on the bending stiffness, length, diameter of the filament, and shear rate of the flow. Further work will consider interactions of semi-flexible fibres under shear flow including an initial study of cross-linked fibres.

Wednesday 5:00 Exhibition Hall / Poster Session

#### Plasticity of fibrin networks: human versus bovine

Vanessa Geimer<sup>1</sup> and <u>Ursula Windberger<sup>2</sup></u>

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Fibrinogen is a hexameric plasma protein composed of symmetrical halves, each composed of an Aa, Bß, and ? subunit, held together at the center of the molecule by disulfide bonds. Although this general architecture is conserved among the species, species-specific differences in the chemistry of the subunits exist, which are transferred to the fibrin fibers to result in clots with different mechanical behaviors. We chose the comparative approach to analyze the mechanical function of fibrin meshworks with stiffer or softer fibers. We hope that assumptions can be made about the basic behavior of blood clots in patients whose fibrinogen chemistry is altered post-translationally or due to liver diseases. Human and bovine clots were first generated in the rheometer gap (CP50, 0.1mm gap width) by addition of homologous thrombin and calcium, and afterwards insitu exposed to increasing shear stress amplitudes until the clot yielded (LAOStress). Increasing the fibrinogen concentration (FIB) in the samples resulted in a prothrombotic state: the higher FIB, the faster the clot formed and the higher was G'. When these clots were cyclically loaded with increasing shear amplitudes, the bovine systems started to deform plastically at lower shear stresses, stiffened more with shear, and required higher shear stresses to align the fibers in the shear direction. For both species apply: the higher FIB was, the higher was G'0 in the LVER, the shear stiffening response and the threshold for switching from non-affine to affine deformation. This holds true for platelet depleted plasma and for fibrinogen solutions. The fibrinogen concentration determines the stiffness of a fibrin meshwork. However, a qualitative change of fibrinogen - by using fibrinogens from different species or by pre-incubating it with LPS - modifies clot behavior substantially. This must be taken into account when infusing patients with commercially available fibrinogen concentrates or when an alteration in fibrinogen chemistry is suspected in diseases.

PO78

PO80

PO83

# Front Microrheology: relation between blood viscosity and red blood cells biomechanical properties in health and disease

Claudia A. Trejo-Soto<sup>1</sup> and Aurora Hernández-Machado<sup>2</sup>

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The rheological properties of blood depend highly on the properties of its red blood cells: concentration, membrane elasticity, and aggregation. These properties affect the viscosity of blood as well as its shear thinning behavior. Using an experimental analysis of the interface advancement of blood in a microchannel, we determine the viscosity of different samples of blood. In this work, we present two methods that successfully normalize the viscosity of blood for a single and for different donors, first according to the concentration of erythrocytes and second according to the shear rate. The proposed methodology is able to predict the health conditions of the blood samples by introducing a non-dimensional coefficient that accounts for the response to shear rate of the different donors' blood samples. By means of these normalization methods, we were able to determine the differences between the red blood cells of the samples and define a range where healthy blood samples can be described by a single behavior. This hematocrit-independent method has been succesfully used for the detection of beta-thalassemia trait and iron deficiency anemia, through the rheological characterization of whole blood samples for different donors.

#### Wednesday 5:00 Exhibition Hall / Poster Session

PO84

**Living droplets: cell spreading as a wetting problem** <u>Ali Alhadi Wahhod<sup>1</sup></u>, Jocelyn Étienne<sup>1</sup>, and Jonathan Fouchard<sup>2</sup>

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The shape taken by cells is intrinsically linked to the mechanics of their cytoskeleton, particularly the actomyosin cortex, a gel which organizes as a thin sheet localized beneath the cell membrane. Within this cortex, the myosin generates a mechanical tension that can be modeled as a surface tension. However, the 3D mechanical balance at the surface of cells, either isolated or in tissue, is not fully understood. As a cell spreads on a flat substrate, it resembles a water droplet, except that the surface concavity reverses from convex to concave near the triple line formed with the substrate. The change in curvature is a key indicator of the mechanical balance governing cell shape, indicating the presence of additional phenomena that modify cell balance. Using fluorescence microscopy, we find that a structure comprised of intermediate filament proteins, specifically vimentin, forms a cortex beneath the actomyosin layer and appears to influence cellular morphology. In this work, an approach is explored to calculate the shape of cell while spreading, treating it as a solution to the Young-Laplace equation, similar to the problem of wetting of water droplets and capillary bridges. Our methodology views the interfaces within the cells, shaped by the actomyosin and vimentin cortices, as portions of surfaces of revolution of constant mean curvature. Then, using pointwise experimental geometric data, such as the contact radius with the substrate, radius at the inflection point, and the cell height, we solve an inverse problem that leads to the determination of pressures and tensions. As a result, a comprehensive reconstruction of the cell 3D-geometry is achieved, which corresponds well with the observations. The findings from this work open up new possibilities in understanding how cell compartment volumes might be influenced by osmotic processes within the cell. While this study provides significant insights into the static case, it also sets the stage for the explorations into the dynamics of cell shape.

Wednesday 5:00 Exhibition Hall / Poster Session

#### **PO85**

# A Cost-effective Rheo-Optical Compression Assay for the Mechanical Characterization of Cell Spheroids Rosalia Ferraro<sup>1</sup>, Stefano Guido<sup>1</sup>, Manlio Tassieri<sup>2</sup>, and Sergio Caserta<sup>1</sup>

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The investigation into the mechanical attributes of cell spheroids, a pivotal facet within the realms of 3D biology and tumor exploration, has historically encountered limitations stemming from the requisite deployment of intricate apparatus and specialized expertise. Addressing this constraint, our research introduces an innovative, cost-effective, and user-friendly rheo-optical compression assay, thereby facilitating notable strides in the domain of mechanobiology. Our pioneering methodology employs commonplace microscopy glass coverslips for the purpose of applying controlled mechanical stress to cell spheroids situated within conventional cell culture plates. Capitalizing on the capabilities of optical microscopes and smartphones equipped with magnifying lenses, this approach permits meticulous observation and analysis of spheroid deformation under applied loads. The economic viability of our technique renders it accessible to a diverse spectrum of biomedical laboratories, thereby democratizing advanced research capabilities. The efficacy of our assay is substantiated through validation experiments conducted on agarose gel particles, affirming the dependability of the acquired mechanical property measurements. Additionally, our methodology has been extended to diverse cell lines. Through the application of established rheological models to our steady-state measurements, we have elucidated responses in terms of Young's modulus, thereby unveiling noteworthy distinctions in viscoelastic behavior among various cell lines, particularly discernible disparities between tumoral and non-tumoral variants. Moreover, our groundbreaking transient rheo-optical compression tests, employing 'i-Rheo', capture nonlinear dynamic mechanical responses, offering access to extensive information within the frequency domain. The incorporation of this pioneering assay into standard cell culture practices not only streamlines the mechanical characterization process but also unfolds novel avenues for comprehensive biological testing.

### Wednesday 5:00 Exhibition Hall / Poster Session **Adhesion of nanocarriers to endothelial cells under flow: A Machine Learning approach** Joao Maia and Saeed Akbari Shandiz

Macromolecular Science and Engineering, Case Western Reserve University, Cleveland, OH 44106, United States

Developing nanocarriers (NCs) tailored for targeted drug delivery to endothelial cells poses a notable biomedical and pharmacological challenge. Varied surface chemistries of functional NCs, including homogenous (Homo) and Janus NCs, offer distinct opportunities for loading multiple drugs and precisely controlling both targeting and cargo release. In this study, we employ Dissipative Particle Dynamics (DPD) simulations to analyze the flow around chains of the endothelial glycocalyx (EG) layer and associated receptors within a microchannel. Our investigation extends to understanding the impact of flow on the morphology and dynamics of the EG layer, as well as the adhesion of NCs to the receptors. Utilizing an energy calculation method, we systematically investigate the adhesion of Homo and Janus NCs to endothelial cells. In contrast to a stationary system where the penetration of NCs was significantly governed by shell entropy loss, the influence of potential energies takes precedence over entropy losses under flow conditions. Creating a performance space map to depict the penetration depths of NCs in relation to variations in their properties necessitates simulations for all conceivable NCs with the specified properties, which is impractical. To overcome this challenge, we employed a Gaussian Process Regression (GPR)-informed machine learning strategy and conducted an extensive exploration of numerous samples, each representing distinct NC properties. The performance space map reveals that NCs with rod and disc shapes exhibit enhanced penetration capabilities compared to those with a spherical shape. Moreover, it highlights that smaller-sized rod-shaped NCs and larger-sized disc-shaped NCs tend to achieve superior penetration. In the case of smaller NCs, the influence of ligand density appears to be constrained. Conversely, for larger NCs, an increase in ligand density is correlated with greater penetration depth, emphasizing its significant role in shaping their penetration capabilities.

#### Wednesday 5:00 Exhibition Hall / Poster Session

PO87

**PO86** 

# Rheological characterization of artificial erythrocytes with encapsulated tracer particles for cardiovascular flow modeling

Gesine Hentschel<sup>1</sup>, Christina M. Winkler<sup>1</sup>, Florian Rummel<sup>2</sup>, and Birgit Glasmacher<sup>1</sup>

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Objectives: Hemodynamic flow models in cardiovascular applications rely on Particle Image Velocimetry (PIV) experiments with limited accuracy due to the use of single-phase glycerol/water substitutes. This study presents a novel approach to achieve improved flow modelling by using a multiphase blood substitute. These particles (beads), derived from poly-sodium acrylate-co-acrylamide P(SA-Am) and P(SA-Am)/Alginate-Co hydrogel, are suspended in glycerol/water mixtures mimicing blood plasma. To improve the detectability of the beads for PIV experiments, liposomes containing a fluorescent dye are incorporated into the beads as markers. The rheological behaviour of beads with liposomes will be characterised using a rheometer. Methods: Microfluidic systems (MFS) for bead production using mineral oil as the continuous phase. Liposomes were prepared by thin film hydration and extrusion. Liposomes were dispersed in the hydrogel solution at concentrations from 5 to 40vol%. The integration was confirmed by cryo-SEM. In rheological experiments, beads are dispersed in glycerin/water-solutions to elucidate their flow behaviour. A plate-plate rheometer measured dynamic viscosity at shear rates (5-20001/s). Results: Biconcave P(SA-Am) beads were produced with an average diameter of 250µm. Round P(SA-Am)/Alginate-Co hydrogel beads had an average bead diameter of 20 µm. Successful incorporation of liposomes into the hydrogel beads was confirmed. Rheometric experiments confirmed the shear rhinning behaviour of the blood substitute fluid. The concentration of liposomes influences the absolute viscosity of the artificial blood. At high shear rates (>1800 1/s) the integrity of the liposomes was damaged. Conclusions: Liposomes were successfully incorporated into the hydrogel beads, the overall viscosity behaviour can be precisely adjusted.

# Wednesday 5:00 Exhibition Hall / Poster Session

PO88

# Manipulation of the mesoscale architecture of protein hydrogels for controlled release and mechanics: A smart stimuli responsive drug delivery material

<u>Ahmad Boroumand</u><sup>1</sup>, Matt D. Hughes<sup>2</sup>, Dimitra Katrantzi<sup>3</sup>, Leon F. Willis<sup>4</sup>, David J. Brockwell<sup>5</sup>, Sally Peyman<sup>6</sup>, Arwen Tyler<sup>7</sup>, and Lorna Dougan<sup>8</sup>

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Hydrogels are important materials with unique mechanical and structural properties which originate from their 3-dimensional water-filled structure. Photochemically crosslinked hydrogels comprised of folded globular proteins are fascinating systems with inherent biological functionality. By combining protein engineering, electron microscopy (EM), rheology, small angle scattering, single molecule biophysics, dityrosine quantification (DQ), and computational modelling we have explored the importance of crosslinking and protein design parameters on

the structure and mechanics of the protein hydrogel. To exploit this fundamental understanding, we focused on the functionality of the materials for specifical biomedical applications, including a stimuli responsive drug delivery system with tailored structural and mechanical properties. The release of small molecule drugs is highly dependent to the mesh size of a hydrogel. While degradation, swelling, and deformation are typically used to control the release of drugs, here we wish to exploit our knowledge of the protein network mesoscale structure by altering the volume fraction (VF) and average coordination (AC) of Bovine Serum Albumin (BSA) proteins inside the gel to reduce the need for post gelation manipulation. Rheological studies so far have shown that the shear modulus of the system can be tuned from ~ 1000 to 3000 Pa varying the VF from 3.7% to 12.33%. Additionally, release assay of model drug calcein indicates changes in the mechanism of the release from Fickian to quasi-Fickian when increasing number of protein building blocks. Upcoming small angle neutron scattering experiment will provide important structural insight into the mesoscale structure of the hydrogels and inform drug delivery studies for cancer disease modelling. In the future, this system is going to be adopted for drug delivery to collateral cancer cells because of its highly tuneable mechanical and network characteristics and the binding affinity of BSA to attach hydrophobic drugs.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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#### **Shear-force-controlled cancer spheroids deformation and cell migration into 3D collagen-hyaluronic hydrogel** Sara Metwally, Joanna Pabijan, and Malgorzata Lekka

Institute of Nuclear Physics Polish Academy of Sciences, Krakow 31-342, Poland

Cells in tissues experience a plethora of forces that regulate their fate. Recent studies showed that shear stress significantly impacts cancerous cell metastasis. To fully understand the metastasis process, it is crucial to evaluate the dynamic interactions of cells with the mechanics of the surrounding extracellular matrix (ECM). In this study, collagen-hyaluronic acid (Col-HA) hydrogels with embedded spheroids were used as a model to study cell migration in a complex environment mimicking ECM. Spheroids were formed from human non-malignant cancer cells of the ureter (HCV29), transitional cell carcinoma (T24), and bladder carcinoma (HT1376) cells. The study aims to analyze the viscoelastic properties of Col-HA hydrogels and their effect on spheroid deformation and cell migration into surrounding 3D scaffolds. Incorporating spheroids into Col-HA scaffolds did not significantly influence the viscoelastic properties of hydrogels. The mechanical properties of Col-HA scaffolds were determined by their cross-linking density controlled by initial collagen solution density. Our results have shown that the increased mechanical properties of hydrogels inhibited spheroids deformation. The migration of cells was significantly larger for T24 cells, which are highly invasive compared to HCV29 and HT1376 cells. Our results demonstrate that Col-HA-based hydrogels are a suitable model for studying the mechanical resistance of spheroids under applied shear forces. We showed that cell escape from the spheroids' surface to the 3D Col-HA matrix depends both on hydrogel mechanics and cell phenotype. Obtained results might help understand the relationship between physicochemical and biological properties in the cell-ECM interfaces. Acknowledgments This study was financed by the National Science Centre (Poland), project no. UMO-2021/41/B/ST5/03032 (NCN, OPUS-21).

## Wednesday 5:00 Exhibition Hall / Poster Session Bacterial sedimentation: Effects of activity? <u>Hyorin Nam</u>

#### Department of Materials, ETH Zürich, Zürich, Switzerland

Active matter systems, such as bacteria, generate internal stresses that drive them far from equilibrium which might be a critical factor enabling them to regulate both their own behavior and the dynamics of their environment. This self-generated stress causes captivating phenomena such as spontaneous self-assembly or pattern formation but also presents a challenging problem in the theoretical understanding of the complex dynamical behaviors within the realm of statistical physics of soft matter. A new principle that all active matter systems generate an intrinsic swim stress that impacts their dynamic and collective behavior was identified by Brady and coworkers. The origin of the swim pressure is based upon a simple notion - a self-propelled body would swim away in space unless confined by boundaries. The pressure exerted by the surrounding walls to contain the particle is precisely the swim pressure. Its magnitude has been verified for synthetic microswimmers reaching a few pascals for swim speeds in the range of several micrometers per second. However, measuring swim pressure in actual bacterial systems has not yet been carried out. Dedicated to advancing research, our focus is to take a step further and investigate how and whether bacteria can sculpt the soft environment by their own activity. To this end, the project will entail measuring the so-called "swim pressure" generated by real marine planktonic bacterial suspension with very fast swimmers (40-50 micrometers per second), by studying the changes in convection-diffusion equilibrium during sedimentation. Prior to this phase, preliminary image analysis has been conducted to extract parameters that describe the behavior and properties of the selected bacteria which leads to the reorientation time distribution.

#### Wednesday 5:00 Exhibition Hall / Poster Session

# CFD simulation of fouling effect on feed water heater outlet temperature

#### Emmanuel E. Duke

#### Mechanical Engineering, University of Nigeria, Nsukka, Nigeria

This work investigates the effect of fouling on the effectiveness of feed water heater in a steam power plant. The plant studied was the 1320 MW Egbin steam power plant. The component considered were low pressure heater 2 and high pressure heater 5. The model was designed using Autodesk Fusion and a computational fluid dynamic (CFD) analysis was carried out on the heater model when cleaned and fouled. The results showed that the steam temperature at the outlet is higher for the fouled model than for the clean model for each of the feed water heater. The simulation results have confirmed the potential implications of fouling on the overall performance of the feed water heater system. In the low pressure feed water heater 2, it is seen that the outlet temperature of the fluid from the simulation data when cleaned is 118 °C and 126 °C when

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fouling occurred in the feed water heater and for the high pressure feed water heater 5, it is seen that the outlet temperature of the fluid from the simulation data when cleaned is 162 °C and 220 °C when fouling occurred in the feed water heater.

# Wednesday 5:00 Exhibition Hall / Poster Session

# Numerical Approach to Paraffin Gelation in Pipelines

Nezia de Rosso<sup>1</sup>, Cezar Negrao<sup>2</sup>, Silvio Junqueira<sup>3</sup>, and Denis Barbara<sup>4</sup>

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The oil production in deep and ultra-deep waters is highly challenging, mostly due to low temperatures at the subsea bed. During the oil uplift from the reservoir to the subsea wellheads, the oils are subjected to temperatures as low as  $4^{\circ}$ C at the seafloor. Heat transfer from the oil to the marine environment occurs during transportation. As the oil cools down, there is a reduction in specific volume (shrinkage), although this reduction is not linear throughout the entire process. The aim of this study is to numerically model the phase change process (gelation) of paraffin to assess the shrinkage and gas void formation within a pipe. The geometry of interest consists of a pipe. The mixture model is used to solve the phase change in the domain. Enthalpy-Porosity model is used for the simulation of the solidification process in the phase change domains. The study reveals that the shrinkage takes place in two stages: in the first stage, it is due to the thermal contraction of the material, which can be expressed with temperature and occurs until the first appearance of paraffin crystals within the fluid. In the second stage, the reduction occurs due to the phase change of paraffin. The study concludes the change in the shape of the solid fraction does not remain uniform throughout the solidification process.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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# Investigating the Ultrasound-Mediated Rheological Properties of Microcrystalline (CMF) and Nanofibrillated Cellulose (CNF): A Model-Based Approach

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Cellulose nanomaterials, microcrystalline cellulose (CMF), and nanofibrillated cellulose (CNF) have gained attention for their exceptional mechanical, structural, and rheological attributes. This study explores the interplay between ultrasound parameters (time and amplitude) and the resulting modifications in CMF and CNF rheological properties. We investigate the roles of hydrogen bonds and nanofibril size under diverse ultrasound conditions. A consistent and exponential increase in the rheological properties, including shear modulus, viscosity, and critical stress, is observed for both CMF and CNF with escalating ultrasound time and amplitude. Beyond mere empirical observations, this study aims to elucidate the underlying mechanisms governing these changes: the critical influence of hydrogen bond availability and nanofibril size. To encapsulate these findings, our study introduces a comprehensive model that integrates the effects of ultrasound parameters, hydrogen bonds, and nanofibril size on the rheological properties of CMF and CNF. This model not only quantifies observed trends but also establishes a predictive framework for tailoring the rheological behavior of these cellulose nanomaterials. By considering the intricate interplay between ultrasound-induced modifications, hydrogen bonding, and nanofibril dimensions, the model provides valuable insights for designing and optimizing processing conditions for these nanocellulose materials. The outcomes of this study contribute significantly to advancing the fundamental understanding of cellulose nanomaterials. Moreover, they underscore the critical significance of molecular-level interactions in the domain of ultrasound-mediated rheological enhancements. This research bears implications for diverse applications spanning biomedical and industrial sectors, offering a systematic approach to harness the versatile and tunable nature of CMF and CNF in the development of advanced materials and products.

# Wednesday 5:00 Exhibition Hall / Poster Session

# Giant electroviscous effect in ferroelectric nematic liquid crystals

Praveen kumar Mylapalli<sup>1</sup>, J Karcz<sup>2</sup>, Przemyslaw Kula<sup>3</sup>, K Karmakar<sup>4</sup>, and Surajit Dhara<sup>5</sup>

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The electroviscous effect deals with the change in the viscosity of fluids due to the external electric field [1]. Here, we report experimental studies on the electroviscous effects in a ferroelectric nematic liquid crystal. We measure electric field-dependent viscosity under a steady shear rate at different temperatures. In the low field range the increase in viscosity ( $\Delta\eta$ ) is proportional to E<sup>2</sup>, and the corresponding viscoelectric coefficient (f) is two orders of magnitude larger in ferroelectric nematic than the conventional apolar nematic liquid crystals. The apparent viscosity measured under a high electric field shows a power-law divergence  $\eta \sim (T-TC)^{-0.7}$ [2], followed by a drastic decrease below the N-NF phase transition. Experimental results within the dynamical scaling approximation demonstrate rapid growth of elongated polar domains under a strong electric field, which suppresses the N-NF phase transition temperature significantly.

# Effect of viscosity ratio on the behaviour of ferrofluid droplet emulsions

<u>Ghulam Muhammad Ayaz Sultan<sup>1</sup></u>, Paolo Cappobianchi<sup>2</sup>, and Mónica S. Oliveira<sup>3</sup>

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The behaviour of a ferrofluid droplet in a non-magnetisable immiscible liquid of different viscosity was investigated using a Hybrid Volume of fluid - Level Set approach [1]. We consider the droplet is subjected to simple shear flow and study the effect of viscosity ratio in the presence of magnetic field. The accuracy of the code was first assessed against existing experimental and numerical data in absence of magnetic field. We then study the effects produced by a uniform magnetic field applied normal to the flow direction for a range of capillary and Bond numbers. Using the approach of [2], the results are then used to infer the rheology of a dilute emulsion, showing that even relatively small magnetic fields can significantly affect the rheological properties such as the effective viscosity of the emulsion.

[1] Capobianchi, P., Lappa, M., Oliveira, M.S.N. (2018) "Deformation of a ferrofluid droplet in a simple shear flow under the effect of a constant magnetic field", Computers and Fluids, 173, 313-323. [2] Capobianchi, P., Lappa, M., Oliveira, M.S.N., Pinho, F. T. (2021) "Shear rheology of a dilute emulsion of ferrofluid droplets dispersed in a non-magnetizable carrier fluid under the influence of a uniform magnetic field", Journal of Rheology, 65, 1

#### Wednesday 5:00 Exhibition Hall / Poster Session

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**Deformation and breakup of a single droplet in extensional flow studied by numerical simulation** <u>Jiyun Jeong<sup>1</sup></u> and Young Ki Lee<sup>2</sup>

<sup>1</sup>School of Food Biotechnology and Chemical Engineering, Hankyong National University, Anseong, Gyeonggi-do 17579, Republic of Korea; <sup>2</sup>School of Food Biotechnology and Chemical Engineering, Hankyong National University, Anseong, Gyeonggi-do 17579, Republic of Korea

We study the deformation and breakup behavior of a single droplet under extensional flow for various density ratios. While many numerical studies have been conducted on the deformation of droplets in shear flow, relatively few studies have been carried out in extensional flow. Additionally, there is insufficient research on how the density ratios between matrix liquids and droplets affect the deformation and breakup of droplets. Inspired above facts, we explore the effects of density ratios between the matrix liquid and a droplet in extensional flow using numerical simulation. To investigate a target system, the color-gradient lattice Boltzmann method (CG-LBM) which is one of the mesoscale simulation techniques for describing a multiphase system was applied. First, we carry out a simulation test under conditions without a droplet to prove whether the flow is properly imposed. In the simulation, a cross-slot channel flow was adapted to apply an extensional flow in the domain. After validating this, we move to the system where a single droplet is floated in the center of the cross-slot channel. When the density ratio between the droplet and the matrix liquid has a relatively low-density ratio such as 1:1 - 1:100, a total breakup in which a break occurs from the center of the droplet is captured. On the other hand, for a large density ratio such as 1:1000, a tip breakup in which a break occurs from the end of the droplet appeared for all the tested capillary numbers. Additionally, for both large density ratio and high capillary number conditions, total breakup and tip breakup are observed same time.

Wednesday 5:00 Exhibition Hall / Poster Session

# Multiscale dynamics in supercritical Taylor-Couette flow of nanostructured fluids

Reza Ghanbari<sup>1</sup>, Kesavan Sekar<sup>1</sup>, Viney Ghai<sup>1</sup>, Marko Bek<sup>1</sup>, Ann E. Terry<sup>2</sup>, Kim Nygård<sup>2</sup>, and <u>Roland Kádár<sup>1</sup></u> <sup>1</sup>Chalmers University of Technology, Göteborg, Sweden; <sup>2</sup>MAX IV Laboratory, Lund University, Lund, Sweden

We correlate nanostructural and flow pattern dynamics in supercritical Taylor-Couette (TC) flow by novel spectral analysis of temporally resolved small-angle x-ray scattering (SAXS) and polarised light imaging (PLI) experiments in self-assembling nanostructured weakly elastic fluids. Pioneering combined rheo-SAXS experiments on anisotropic clay suspensions under Taylor-Couette flow have been previously employed, together with linear stability analysis, to study the nanoscale ordering of suspended platelet-like particles during onset of the first Taylor-Couette instability mode - Taylor vortex flow (TVF) [1]. However, these experiments were limited in temporal resolution and could not hence address the dynamics of the flow-induced instability. With the advent of diffraction-limited synchrotron sources [2] and the use of high flux x-ray monochromators, we can now probe three orders of magnitude faster dynamics, paving the way for a new era in combined rheological/flow and SAXS experiments. We study two weakly elastic and weakly shear thinning nanoparticle suspensions of different particle morphologies - rod-like cellulose nanocrystals (CNC; 3 w%) and platelet-like graphene oxide nanoparticles (GO; 0.7 w%). We follow the multiscale dynamics of the flow across axisymmetric, azimuthally periodic, frequency modulated and turbulent wavy modes (wavy vortex flow, WVF; modulated wavy vortices, MWV; chaotic wavy vortices, CWV; and turbulent wavy vortices, TWV), all Newtonian-like with non-Newtonian features but also in novel flow patterns identified in the GO suspensions. We find the particle morphology to govern several fundamental aspects of the flow-induced instabilities and most importantly, the SAXS data reveal hitherto hidden, particle-morphology-driven dynamics of nanostructured fluids in supercritical Taylor-Couette flow.

[1] Philippe, A. M., et al. Phys. Rev. Lett. 108, 254501 (2012); [2] Eriksson, M., et al. J. Synchrotron Rad. 21, 837-842 (2014).

**Buoyancy driven flow of droplets in viscoelastic fluids** Giancarlo Esposito, Yannis Dimakopoulos, and John Tsamopoulos

Department of Chemical Engineering, University of Patras, Patras, Greece

The buoyancy-driven flow of deformable objects in complex fluids has stirred the interest of the scientific community in the past decades because of its direct implications in a wide span of industries. Although several experimental and numerical studies have been dedicated to the analysis of a single bubble rising in viscoelastic media [Fraggedakis et al., 2016], little attention has been dedicated to systems where the dispersed phase has a non-negligible viscosity. Previous experiments show that viscous drops rising in viscoelastic materials can form long tails and break in different ways depending on their size and the properties of the continuous phase [Ortiz et al., 2016]. To explore this scenario, we perform a novel computational analysis examining the rise of a Newtonian droplet in a polymeric solution. The rheological behaviour of the suspending fluid is modelled via the e-PTT constitutive equation to incorporate both elastic and shear thinning effects. A parametric study is carried out to elucidate the role of each physical force on the dynamics of the rising drop, exploring the interplay between buoyancy, capillarity, elasticity and viscous dissipation. The initial radius of the droplet is found to be a crucial parameter in the determination of the breakup mode. Increasing material elasticity of the continuous phase turns a spherical drop to one with a cusp and eventually with an extended tail in its rear pole. The viscoelastic extensional stresses developing in the continuous phase are crucial for the generation of these long tails. Low values of the interfacial tension allow the extension of threads of uniform width, but higher values break it under a Rayleigh-type instability. An important conclusion is that viscoelastic materials can be used to encapsulate small drops generated by the tip streaming of liquid filaments. Acknowledgement: This work has been financially supported by the European Union's Horizon 2020 research and innovation program under the Marie Sklodowska-Curie Grant Agreement N°95

#### Wednesday 5:00 Exhibition Hall / Poster Session

### Flow Asymmetry in Planar Flows enhanced by Viscoelasticity

#### <u>Vivaswan ChandraShekar</u>, Alexis Ayala, Baptiste Laclau, Guillaume Maîtrejean, and Hugues Bodiguel Laboratoire Rhéologie et Procédés, Université Grenoble Alpes, Grenoble 38400, France

Planar flow of viscoelastic liquids is a well-established phenomenon be it past a confined cylinder or in other simple geometries like a T-Junction, especially since the former serves as a traditional benchmark problem in numerical studies providing crucial insights into the complex kinematics of these fluids. The significance of the studies around the flow behaviour in these geometies lies in the litany of industrial applications from polymer mixing to offshore oil rigs. The studies of note to this work are surrounding asymmetric flows where a new viscoelastic flow instability has been observed. Above a critical Weissenberg number (Wi, a dimensionless number used to describe the strength of elastic forces to viscous forces), complex fluid flow past a confined cylinder becomes highly asymmetric. This has been seen experimentally and numerically in Worm-like Micelles and shear-thinning viscoelastic liquids 1 2, where the liquids have preferential pathway around the cylinder above a critical Wi. This work explores viscoelastic planar flow but with a twist, with the introduction of a geometric asymmetry. In the case of viscoelastic planar flow past a confined cylinder is placed off-center from the centerline of the channel. With the introduction of this geometric asymmetry, an intensified flow asymmetry arises at higher Weissenber numbers. A critical Weissenberg number is observed beyond which the flow drops almost to zero in the narrower pathway around the cylinder. A similar phenomenon is also observed in a planar flow in a T-junction with asymmetric branches. This phenomenon is studied using two-dimensional FVM simulations of an Oldroyd-B fluid and in experiments of partially hydrolysed Polyacrylamide (HPAM) solutions flowing in microfluidic devices.

# Wednesday 5:00 Exhibition Hall / Poster Session

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### **Experimental study of dilute polymer solution flow in the converging-diverging channels with axial symmetry** Aidar Kadyirov, Ivan Shvedov, and Rinat Zaripov

# Institute of Power Engineering and Advanced Technologies, FRC Kazan Scientific Center, Russian Academy of Sciences, Kazan, Tatarstan 420111, Russia

The geometry of converging-diverging channels can be viewed as a model porous media, since they have the same predominantly extensional flow kinematics. So the present study will be interesting to better understand the flow behavior in such channels. The experimental measurements include the point velocity measurements in various cross-sections of converging- diverging channels as well as the construction the axial velocity distribution along center line. The vortex length for each case was estimated by artificial construction of streamlines by superimposes each subsequent frame on the first one. We proposed modified version of expression to calculate extensional strain rate which can well classified the following modes under viscoelastic flow in convergent-divergent channels: slow flow without vortex, vortex enhancement and divergent flows. For geometries with the highest converging length we observed extreme mode for which the viscoelastic flow behave like divergent flow till reached the vicinity of the orifice between the converging and diverging sectors without presence any vortex area near the pipe wall and still stay stable.

Acknowledgments. The study was supported by a grant Russian Science Foundation No. 23-21-00166, https://rscf.ru/project/23-21-00166/

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#### Wednesday 5:00 Exhibition Hall / Poster Session

# How do polymers stretch in capillary-driven extensional flows?

Vincenzo Calabrese, Fabian Hillebrand, Amy Shen, and Simon J. Haward

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Measurements of the capillary-driven thinning and breakup of liquid filaments are widely used to extract the extensional rheological properties of complex fluids. For viscoelastic (e.g., polymeric) liquids, the existence of an exponential 'elastocapillary' (EC) thinning regime, associated with the unraveling of polymer chains, is used for the determination of the "longest polymer relaxation time". The intimate relationship between the filament thinning dynamics and the polymeric response within the thinning filament are generally assumed, but are based almost entirely on the Oldroyd-B and FENE-P model predictions. We use birefringence measurements in a cross-slot-type device to determine the longest relaxation time ( $\tau$ ) directly from the inverse strain rate for the onset of polymer orientation, and we compare the values against those obtained from the EC regime in capillary thinning experiments ( $\tau_{cap}$ ). For highly flexible polymers, such as polystyrene in a theta solvent, we find  $\tau \approx \tau_{cap}$  and we believe the FENE-P model predictions for the macromolecular dynamics in the fluid filament are largely valid. However, for a semiflexible  $\lambda$ -DNA in a viscous buffer solution we find  $\tau \gg \tau_{cap}$ . In this case, our best estimates indicate that the DNA is likely highly oriented before the onset of exponential thinning; it seems that the dynamics of the fluid filament are not sensitive to the relatively weak entropic elasticity of the DNA in the early stages of orientation. Our results from a wide variety of polymers of different extensibility indicate the assumptions underlying the analysis of capillary thinning measurements are likely only valid for highly flexible polymers, but do not hold in general. Caution is required when using capillary thinning to extract relaxation times for polymeric fluids of lower extensibility, such as solutions of polyelectrolytes in the absence of screening counterions.

#### Wednesday Exhibition Hall / Poster Session 5:00

# Jetting prediction model for weakly viscoelastic Ink by machine learning

Seongju Kim<sup>1</sup>, Sungjune Jung<sup>2</sup>, and Gioele Balestra<sup>3</sup>

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Inkjet printing has considerable potential ability for additive manufacturing technology. The functional inks, which have been commonly used in the industry, lead to the high complexity of predicting the jetting behavior. The complexity is induced by the rheological properties of the ink. However, the printability map composed of Ohnesorge and Weber numbers doesn't contain the rheological features. We present the machine learning-based predictive model of jetting behavior from the Deborah number, Ohnesorge number, and waveform parameters. We prepared the 10 weakly viscoelastic model inks and measured their storage and loss modulus, which were close to the theoretical Maxwell model linear viscoelastic fluid. The relaxation time of viscoelastic inks is obtained by analyzing the Maxwell model equation. The big datasets of the ink jetting at different waveforms were collected from the high-speed images. Deborah number contributed to increasing the prediction accuracy of learning models. Multi-layer Perceptron showed outstanding performance after comparing the prediction accuracy of machine learning models. The final predictive model also exhibited remarkable accuracy of unknown ink according to waveform parameters and the correlation between jetting behavior and ink properties were in reasonable agreement. We proposed a new printability map characterized by the Ohnesorge number and Deborah number through the predictive model for weakly viscoelastic ink.

Wednesday 5:00 Exhibition Hall / Poster Session

# Air entrainment in falling viscoelastic jets: new complex fluid flow phenomena

Thomas John<sup>1</sup>, Jack King<sup>2</sup>, Steven J. Lind<sup>3</sup>, and Claudio P. Fonte<sup>4</sup>

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Falling jets of complex fluids represent both a common industrial problem (e.g. filling of shampoo bottles, laundry capsules, etc.), as well as an interesting fundamental non-Newtonian fluid mechanics problem. Falling jets of viscoelastic fluids, particularly, can exhibit a range of fascinating phenomena, such as buckling and the "leaping shampoo", or Kaye, effect. It is important to understand these phenomena so that we may mitigate challenges in processing such complex fluids in industrial applications. Whilst buckling of jets is known to be an effect driven by viscosity, recent investigations have shown that the Kaye effect is related to both viscoelasticity and air entrainment. We study a falling vertical jet of a shampoo formulation, driven by a syringe pump through a small nozzle, which impacts a pool of the same liquid. For low velocities, the jet spreads axisymmetrically, and the flow remains laminar. As the flow rate is increased (for a fixed height, nozzle diameter, and rheology), we observe that the jet undergoes several transitions. Firstly, the jet penetrates the surface and becomes submerged into the pool, where a thin (cylindrical) layer of surrounding air is driven down with the penetrating jet. This has previously been observed only for Newtonian fluids. At the bottom of the jet, the flow can become unstable, in which case the thin film of air ruptures and is dispersed as bubbles into the surrounding liquid. For even higher velocities, we observe buckling/coiling instabilities at the bottom of the jet, and even a behaviour reminiscent of the Kaye effect, where the jet appears to break symmetry and bends. We also observe, under certain conditions, the formation of "bubbles-on-a-string" structures which, to the best of our knowledge, have not been observed previously with a Newtonian fluid in this type of flow. We capture these fascinating phenomena with high-speed videography, and investigate the role of viscoelasticity and surfactants.

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# Wednesday 5:00 Exhibition Hall / Poster Session Investigation of the Deborah number effect over jetting prediction

Jules Richardot<sup>1</sup>, Seongju Kim<sup>2</sup>, and Sungjune Jung<sup>3</sup>

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Inkjet Printing Drop-on-Demand technology has gained importance over the past few years for its diverse applications varying from display applications to transistors since it enables to precisely deposit small amounts of ink onto a substrate. The rheological properties of the ink are the main factors affecting the final print quality. However, previous jetting behavior predictions have been proven inaccurate due to the increasing complexity of viscoelastic functional fluids. This study examines the "in-channel" rheological effects of weakly viscoelastic fluids during the drop ejection process. To take into account the viscoelastic properties, the Deborah number is used to redefine a jettability map for various polymer-enhanced solutions. To achieve this, the high-frequency shear of the ink was measured using a squeeze-flow rheometer to determine the storage (G') and loss (G") moduli. The relaxation time was obtained through Maxwell model fitting and used to compute the Deborah number. The study captured the jetting demeanors through high-speed camera recordings and correlated it with the rheological properties of the ink via the Deborah number. The moduli were found to follow the Maxwell Model for linear viscoelastic fluids, enabling the extraction of a single relaxation time at their intersection point. This led to the calculation of the Deborah number. The latter was strongly dependent on the polymer concentrations and molecular weight. As those increased, the Deborah number also increased, indicating stronger viscoelasticity. This increase in Deborah number was accompanied by a smaller single-drop zone when compared to the Weber number, and eventually non-ejection for De = 1. These findings emphasize the significance of high-frequency linear viscoelastic rheology in understanding and improving the jetting morphology of viscoelastic fluids.

# Wednesday 5:00 Exhibition Hall / Poster Session

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# Unpicking the SECRETs of kinematically mixed rheology with Shear Extension Combined Rheology Experimental Techniques

# Richard Hodgkinson

Department of Materials Science and Engineering, The University of Sheffield, Sheffield, South Yorkshire S1 3JD, United Kingdom

This poster introduces and summarises a new/current four year experimental fellowship project to expand our understanding of "kinematically mixed" rheological flows - those which contain simultaneous shear and extension.

Three key areas are elaborated:

- Prior work, which showed the effect of extensional flow on the apparent shear viscosity of a polymer solution via a novel two-phase flow technique developed.

- The scope of the fellowship project, to transition this to utilising flow MRI, enabling the study of a wide range of test systems.

- The challenge of correctly measuring shear and extension rates in arbitrary flows.

Suggestions of test systems and rheological regimes of interest are actively sought, as well as discussion surrounding the strain rate measurement challenge.

The author previously developed a two-phase flow technique allowing us to study shear viscosity under not a conventional pure shear flow, but for the first time, one containing a simultaneous combination of shear and extensional deformation; measuring shear stresses within the core of the extensional flow (https://doi.org/10.1122/8.0000380). Study of kinematically mixed rheology is especially relevant to flows of materials which show extension-aligning behaviour, from polymers to anisotropic particle suspensions, but was previously limited due to the use of optical PIV (particle image velocimetry). The variety of materials which may be accessed via flow MRI (magnetic resonance imaging) - e.g. opaque suspensions - stand to inform fluid model development and validation towards improved prediction of complex, industrially-relevant flow fields. Coupled to this, independent measurement of shear and extension rates in arbitrarily orientated and potentially rotating flow fields is surprisingly non-trivial (even in 2D), differing from our apparently well-defined shear and extension rates in the laboratory reference frame: a parallel challenge, relevant to both this work and the wider rheology field.

# Wednesday 5:00 Exhibition Hall / Poster Session

PO107

# Rayleigh-Benard type stability in Non-Newtonian Fluid using RheoTool based PTT Model Simulation

Govind Maurya, Suneet Singh, and Lalit Kumar

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Rayleigh-Benard Convection and its stability has been widely studied for Newtonian fluids. Current numerical study focuses on the stability of RBC in non-Newtonian fluids using Rheotool based PPT model. The critical Rayleigh number for square cavities has been presented and transition from no-flow to flow regime has been shown using isotherms and velocity profiles at mid-height and mid-width of the cavity.

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#### PO108

# Wednesday 5:00 Exhibition Hall / Poster Session

Elastic turbulence in two-dimensional Taylor-Couette flows

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It is widely known that adding small amounts of long-chain polymers can strongly impact the properties of flowing fluids. Recently, experiments have shown a remarkable effect of highly viscous polymer solutions: developing an "elastic turbulence" regime in the limit of small Reynolds numbers and strong elasticity. In this regime, the flow displays rich dynamics typically encountered in high Reynolds number turbulent flow in Newtonian fluids, such as a broad range of active scales and growth of flow resistance in the limit of vanishing Reynolds numbers. Due to the turbulent-like motion, elastic turbulence is proposed as an efficient technique for mixing in very low Reynolds flows, such as in microchannel flows. Despite its great technological interest, elastic turbulence is still only partially understood due to the "high-Wi problem". In this study, we investigate the behaviour of a viscoelastic fluid in a 2D Taylor-Couette geometry using numerical solutions of the Oldroyd-B model in an elastic turbulence regime. By restricting our investigations to two spatial dimensions, we have the simplest implementation of a Taylor-Couette flow. Despite this simplification, significant insight into elastic turbulence can be obtained by performing 2D simulations. We use OpenFOAM to perform direct numerical simulations to solve the Oldroyd-B model up to large Weissenberg numbers. In particular, it can handle the high-Weissenberg-number problem, where numerical instabilities occur at high Wi. We work at a very low Reynolds number appropriate for flows at the micron scale and keep its value fixed during all simulations. Thus, the observed turbulence is only due to the polymeric, i.e., the elastic component of the fluid. We observe a supercritical transition from the laminar Taylor-Couette to an irregular flow at a critical Weissenberg number accompanied by an increase in flow resistance. We illustrate the irregular flow and identify it as turbulent.

#### Wednesday 5:00 Exhibition Hall / Poster Session

# Energy cascade and intermittency in turbulent elastoviscoplastic flows

Mohamed S. Abdelgawad, Ianto Cannon, and Marco E. Rosti

Complex Fluids and Flows Unit, Okinawa Institute of Science and Technology, Onna son, Okinawa 904-0495, Japan

We investigate the turbulence characteristics of elastoviscoplastic (EVP) fluids through high-resolution direct numerical simulations at significantly high Reynolds numbers. Elastoviscoplastic fluids, which demonstrate the complex interplay between their elastic, viscous, and plastic properties, undergo transitions from solid-like to liquid-like states under varying stress levels. Our study focuses on the implications of fluid plasticity on turbulence, particularly its influence on the turbulent energy cascade and flow properties. We observe that as the fluid plasticity increases, there is a shift in the energy distribution across scales: energy increases at large scales while decreasing at small scales, indicating the emergence of a novel intermediate scaling range. This behavior reflects a deviation from the classical Kolmogorov's turbulent energy cascade observed in Newtonian fluids and points to the non-Newtonian nature of EVP fluids influencing energy transfer. Furthermore, our findings reveal an enhancement in intermittency, attributable to the plastic nature of the fluid, which directly impacts the dissipation behavior of the fluid. This increased intermittency has a profound implication for understanding natural phenomena such as landslides and lava flows, where the predicted intensification of extreme events poses greater unpredictability.

Wednesday 5:00 Exhibition Hall / Poster Session

# Direct numerical simulation of roll waves in an elasto-viscoplastic fluid

<u>Arivazhagan G. Balasubramanian</u><sup>1</sup>, Danila Denisenko<sup>2</sup>, Gael Richard<sup>2</sup>, Guillaume Chambon<sup>2</sup>, Ricardo Vinuesa<sup>1</sup>, and Outi Tammisola<sup>1</sup>

# <sup>1</sup>Dept. of Engineering Mechanics, KTH Royal Institute of Technology, Stockholm, Sweden; <sup>2</sup>Univ. Grenoble Alpes, INRAE, IGE, Grenoble, France

The flow of shallow fluid down an incline can become unstable under specific conditions related to fluid inertia, inclination angle and fluid properties. This instability leads to the formation of periodic shock-like disturbances on the free surface, commonly known as 'roll waves'. Since the pioneering experiments conducted by Kapitza &. Kapitza (1949), the study of falling liquid films has been a subject of both fundamental and applied research, spanning a wide range of applications. Existing literature extensively explores roll waves down an incline for Newtonian fluids. However, certain aspects of the phenomenon remain not fully understood, especially in the inertia-dominated regime (F. Denner et al. (2018)). Through a combination of experiments, direct numerical simulation and low dimensional modelling, researchers have delved into the dynamics of solitary waves, providing new insights into observed hydrodynamical behaviour. While the primary focus has historically been on understanding dynamics in Newtonian fluids, many real-world applications in engineering (such as paint, food) and geophysical pheonomena (like glaciers, mud) involve fluids with more complex rheological behaviour (G. Chambon et al. (2014); M.H. Allouche et al. (2017)). Viscoplastic fluids, commonly encountered in these scenarios, exhibit shear-thinning behaviour alongside yield stress. Consequently, this study utilizes DNS to investigate roll waves in viscoplastic and elasto-viscoplastic fluid, aiming to comprehend the influence of elasticity and plasticity on the critical Reynolds number limit to identify the stability of roll waves.

PO109

# Numerical study of elasto-visco-plastic flows past a confined cylinder

Milad Mousavi<sup>1</sup>, John Tsamopoulos<sup>2</sup>, and Yannis Dimakopoulos<sup>3</sup>

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We examine the flow of elastoviscoplastic (EVP) fluids around a cylinder confined between two parallel plates. The Saramito model is solved via the finite-volume method with the OpenFOAM software. As in viscoplastic materials, unyielded regions are observed as two small islands located above and below the cylinder and as polar caps at the two stagnation points on the cylinder. Most interestingly, an elongated yielded area around the midplane is predicted downstream of the cylinder, sandwiched between two unyielded areas, under certain conditions. This surprising result for Carbopol 0.1% appears when considering a blockage ratio of 0.5 (the ratio of the cylinder's diameter to the channel's width) and above a critical elastic modulus (G>30 Pa). An approximate analytical solution for the stress behind the cylinder and at the midplane reveals that its magnitude decays exponentially towards the yield stress and approaches it only asymptotically. The materials behave as highly elastic when the elastic modulus is decreased, hence they deform more before yielding, which results in the expansion of the unyielded areas. Furthermore, by decreasing the elastic modulus below a critical value, the transient variations remain for longer times in all variables, including the yield surface, instead of a monotonic approach to the steady state. The yield surface may exhibit damped oscillations, or irregular shapes even without a plane of symmetry, all under creeping flow conditions. These patterns arise mainly behind the cylinder, where the material elongates the most and triggers a purely elastic instability. Similarly, an instability arises for high yield stress values because of the decreased thickness of the yield film between the cylinder and the unyielded islands.

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# Wednesday 5:00 Exhibition Hall / Poster Session

# Entrance flow of viscoplastic materials in annular tubes in the presence of wall slip

Evgenios Gryparis<sup>1</sup>, Alexandros Syrakos<sup>2</sup>, and Georgios C. Georgiou<sup>3</sup>

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We solve numerically the entry flow of Newtonian and viscoplastic materials in concentric annular tubes assuming the Navier slip occurs at the walls. The entrance length, i.e., the length required for the flow to adjust from a uniform to the fully-developed profile is usually defined as the distance, Lc, required for the maximum velocity to attain 99% of its fully developed value. In the present work, the entrance length is caculated as a function of the radial coordinate, and its maximum value is considered as the global entrance length, Lg. The numerical simulations are carried out using the open-source finite element software FeniCS for Reynolds numbers up to 1000 and various values of the Bingham and slip numbers. The difference between the two entrance lengths (Lc and Lg) becomes more important at high Reynolds numbers. The numerical results reveal that flow development may be slower near a wall than in the region of the maximum fully developed velocity and that the effects of wall slip are more pronounced for small values of the radii ratio.

# Wednesday 5:00 Exhibition Hall / Poster Session Buoyant miscible viscoplastic fluid injections in a closed-end pipe

Soheil Akbari and Seyed Mohammad Taghavi

# Department of Chemical Engineering, Laval University, Quebec, QC G1V 0A6, Canada

We investigate buoyant miscible injection flows of a heavy viscoplastic fluid into a vertical pipe filled with a light Newtonian fluid. The viscoplastic fluid is injected via an eccentric pipe into an outer closed-end pipe filled with a Newtonian fluid. We use mainly experiments, accompanied by complementary numerical simulations and semi-analytical methods, to analyze the effects of the flow and fluid parameters on the flow dynamics. To generalize our results, we present them using the relevant dimensionless numbers of the flow, including the buoyancy numbers (?), the Bingham number (B), the viscosity ratio (M), and the aspect ratio (d). For the partial inlet injection, a filament of the core fluid immersed in the in-place fluid is formed, for which three distinct flow patterns are observed, namely, the breakup, coiling, and bulging (buckling) regimes [1,2]. In the breakup regime, as the injection continues and time grows, when the injected viscoplastic fluid can no longer sustain the buoyancy force, the core fluid is yielded towards thinning and, finally, breaking up. In the bulging regime, the interfacial shear stress and the pressure exerted by the upward flow of the in-place fluid on the core lead to yielding towards the bulging of the core. For the coiling regime, the core fluid and its diameter remain unyielded/unchanged until the fluid deforms into a coiling structure. To gain a deeper understanding, we develop a lubrication approximation model using the Herschel-Bulkley constitutive equation. Based on a reasonable prediction of the yielding onset, the flow regimes are classified based on an elegant combination of the dimensionless flow parameters, including B, ?, M, and d, for which a good agreement with the experiments is achieved. The results of this study help improve the quality of the cement plug placement in the plug and abandonment of oil and gas wells.

1 S. Akbari and S. M. Taghavi, J. Fluid Mech., 940, A42 (2022). 2 S. Akbari and S. M. Taghavi, J. Non-Newton. Fluid Mech., 306, 104836 (2022).

PO111

PO113

Yield stress fluid deformation in vicinity of a porous wall under air current

Nicoleta-Octavia Tanase and Ciprian-Stefan Mateescu

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The paper analyses the dynamics of a yield stress fluid in a confined air current, in the vicinity of a porous wall. The main goal of the study is to determine the fluid linkage through the porous wall under steady and periodic controlled air flows. Experimental investigations of the behavior of an artificial mucus gel in the air current are performed in a straight pipe and a tracheal bifurcation. The evolution of the fluid interface is recorded with a fast camera and the linkage volume is measured with a sensitive scale. The 3D numerical simulations of the gel deformation, using the Herschel-Bulkley model, and the computation of the flow rate transfer through the porous wall are performed with the VoF code implemented in the Fluid Flow (Fluent) software. The numerics are correlated with the visualized deformation over time of the air-gel interface. The distribution of the wall shear stress is analyzed and related to the fluid linkage and interaction with the porous wall. The study has direct applications in modeling the flow of mucus in the tracheal bifurcation during the respiratory cycle.

#### Wednesday 5:00 Exhibition Hall / Poster Session

# Pinch-off of an elasto-visco-plastic filament: Scaling analysis and self-similarity

Pourya Zakeri, Pantelis Moschopoulos, Yannis Dimakopoulos, and John Tsamopoulos

Department of Chemical Engineering, University of Patras, Patras, Greece

Understanding the local dynamics of a fluid thread just before it pinches is vital in advancing industrial operations, such as inkjet and 3D printing. Elasticity and plasticity once deemed mutually exclusive, both must be accounted for in filament stretching of yield stress materials as shown in [1], which studies mainly bulk dynamics, neglecting pinch-off. Therefore, we undertake a computational study to examine the pinching dynamics of elasto-visco-plastic (EVP) materials employing PEGAFEM-V [2]. The confined material between two coaxial disks extends abruptly with the movement of the upper disk. When the thread radius drops below a threshold, capillarity drives the rapid collapse of the thread, making thinning independent of the upper plate movement. We demonstrate a power law evolution of the minimum radius, velocity, and stress with numerical calculation of the respective exponents. The results of our 2D simulations regarding the values of the power law exponents are verified performing asymptotic analysis with the slender filament equations. Also, we find the self-similar solutions during pinching by rescaling properly the 2D results. The effect of yield stress diminishes due to high stresses during pinching. For an EVP filament with very low elasticity, the evolution aligns initially with that of a viscoplastic material but deviates eventually from it as elastic effects become significant. In this regime, scaling exponents attain a universal value, independent of the material elastic modulus, resembling the linear decay in pinching finite extensible viscoelastic threads [3].

[1] P. Moschopoulos, E. Kouni, K. Psaraki, Y. Dimakopoulos, and J. Tsamopoulos, Soft Matter, 2023. [2] S. Varchanis, A. Syrakos, Y. Dimakopoulos, and J. Tsamopoulos, J Nonnewton Fluid Mech, 2020. [3] M. A. Fontelos and J. Li, J Nonnewton Fluid Mech, 2004. Acknowledgment: This Project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 955605.

#### Wednesday 5:00 Exhibition Hall / Poster Session Interacting tandem pairs of bubbles in elastic yield stress materials

Athanasios Kordalis, Yannis Dimakopoulos, and John Tsamopoulos

Department of Chemical Engineering, University of Patras, Patras, Greece

We consider the buoyancy-driven rise and interaction between two initially stationary, tandem bubbles of constant volume inside an elastoviscoplastic material, extending our previous work on the topic [1]. We investigate the dynamics for three different initial separation distances in a wide range of bubble radii and we predict three distinct regimes: approach, separation, and formation of steady distance. The pair approaches exclusively when the leading bubble is smaller than the trailing one and when inertia is important for both bubbles. When the trailing bubble is not as large, hence the elastic effects play a key role, the leading one dictates the response. A significantly larger leading bubble results in separation of the pair, while a slightly larger leading bubble results in forming a steady distance of the pair. The steady distance configuration is observed only when the initial separation distance is larger than a critical value and elastic effects are intense. Below this critical value the pair approaches. Above this critical value the pair converges to a constant and common value regardless of the initial separation distance. We construct maps with regions of approach, separation, and constant distance in the plane of trailing bubble radius vs. bubble radii ratio for each initial separation distance we examined.

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[1] Kordalis et al., Phys. Rev. Fluids 8, 083301 (2023)

PO115

# Wednesday 5:00 Exhibition Hall / Poster Session **Particle alignment and bubble deformation in solidifying magma and lava flows** <u>Ceri Allgood</u> and Edward W. Llewellin

#### Department of Earth Sciences, Durham University, Durham, United Kingdom

The textures of crystals and bubbles within solidified volcanic conduits and lava flows are commonly used to infer the flow conditions that existed while they were active. The rotation and collision of particles and the deformation of bubbles within flows have both been studied extensively in order to relate solidified rock textures to flow processes; however, previous theoretical analyses and analogue experiments have assumed that the suspending fluid has a uniform and constant viscosity. This assumption is unlikely to hold true in volcanic settings, where molten rock cools and solidifies progressively inwards from its exterior while its interior continues to flow. Magma develops viscosity gradients, which are likely to impact the rotation and collision of crystals, and the deformation of bubbles, influencing the final texture once it solidifies. The effects of this viscosity gradient will be most pronounced at the fast-cooling flow margins, which are the regions most often sampled in efforts to determine flow directions. Here, we present results of analogue experiments in which particle and bubble suspensions are sheared within progressively solidifying fluids. We use mica flakes as an analogue for suspended crystals, air bubbles as an analogue for volatiles, and molten sugar as an analogue for the melt. 3D x-ray computed tomography scans of our samples reveal that, due to the balance between cooling and deformation rates, vesicles adopt a curved axis rather than remaining ellipsoidal, and crystal textures become more strongly aligned than they would in an isothermal setting. Our experiments demonstrate the importance of the balance between flow rates and cooling rates, which will improve interpretations of textures in volcanic conduits and lava flows, such as using the elongation and orientation of bubbles to estimate strain rates.

# Wednesday 5:00 Exhibition Hall / Poster Session

PO118

PO117

#### **Investigating the influence of magma viscosity on the 3D structure and morphology of volcanic domes** Kendra D. Ní Nualláin and Claire E. Harnett

UCD School of Earth Sciences, Dublin, Dublin D04 V1W8, Ireland

Understanding the dynamics of volcanic dome growth, as well as their internal structure throughout dome growth, is an important tool to determining long-term hazard. Existing models of volcanic dome growth currently lack numerical complexity in that they are primarily 2D, and they do not consider different magma viscosities. We use the most recent cutting-edge technical advances in computational modelling and the latest software to create three-dimensional models of lava dome growth while considering a range of magma viscosities. We use Particle Flow Code 3D software which employs the distinct element methods (DEM) to simulate dome growth.

We modelled three end-member dome morphologies by varying the stiffness of the contacts between particles. This simulates the ductility of the flow with the intention of relating it to an effective shear viscosity. The modelled magma with the lowest stiffness leads to the dome with the greatest width and the lowest aspect ratio. The modelled magma with intermediate stiffness showed a more symmetrical shape with a flatter top. The modelled magma with the highest stiffness produced domes with the greatest height and lowest width. To relate ductility to real-world viscosity, we are adapting a calibration method described to investigate granular flows, whereby the coefficient of friction and inertial number are calculated and used to determine the effective shear viscosity. Following calibration, these values will be used to calculate an effective shear viscosity.

We aim is to use observational data from the 2020-2021 eruption of St. Vincent to help inform modelled viscosity. This eruption was geometrically complex so lends itself well to 3D modelling, and existing studies have made estimates of the viscosity during dome growth. We aim to combine our 3D dome growth models with the pre-eruptive digital elevation model from St. Vincent and iteratively change our model viscosity parameters until the observed dome growth is matched with our calibration models.

# Wednesday 5:00 Exhibition Hall / Poster Session

# PO119

# Simulating the rheology of polydisperse wet granular materials using pairwise formulation of contact, lubrication and Brownian forces

Christopher Ness<sup>1</sup>, Xuan Li<sup>1</sup>, and John Royer<sup>2</sup>

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Dense suspensions of solid particles in viscous liquid are ubiquitous in nature, and there is a clear need for efficient numerical routines to simulate their rheology and microstructure. Particles of micron size present a particular challenge: at low shear rates colloidal interactions control their dynamics while at high rates granular-like contacts dominate. While there are established particle-based simulation schemes for large-scale non-Brownian suspensions using only pairwise lubrication and contact forces, common schemes for colloidal suspensions generally are more computationally costly and thus restricted to relatively small system sizes. Here we present a minimal particle-based numerical model which incorporates Brownian forces in pairwise form alongside contact and lubrication forces. We show that this scheme reproduces key features of dense suspension rheology near the collodial-to-granular transition, particularly relevant for mixtures of particles of broad size range. This scheme is implemented in LAMMPS, a widely-used open source code for parallelized particle-based simulations, with a runtime that scales linearly with the number of particles making it amenable for large-scale simulations.

#### Wednesday 5:00 Exhibition Hall / Poster Session **The Rheological Feasibility of Magma Mixing at the Drumadoon Sill, Isle of Arran** Frankie Butler

#### Volcanology, University Of Bristol, Bristol, United Kingdom

The rheological feasibility of magma mixing is an under researched area of volcanology. Holistic models of emplacement are impossible to reconstruct without considering the rheology of the magmatic components involved. These models are integral for effective hazard management of polymodal volcanic systems e.g., Eyjafjallajökull, Iceland and Campo de Flegri, Italy. Magma mixing influences the explosivity of eruptions, knowing the rheological feasibility of mixing can contribute towards mitigation of hazards. Paleo-igneous systems such as the Drumadoon Sill Complex on the Isle of Arran have often only been considered from a chemical petrological background to describe emplacement theories and the feasibility of magma mixing. This dissertation used the Giodarno et al., 2008 viscosity calculator to assess the rheological feasibility of magma mixing between significant Basalt and Rhyolite samples from the Drumadoon Sill Complex under varying % wt H2O. The results of this study show that there is a theoretical zone where the viscosity of magmas intersect and the viscosity ratio is the same; which this study termed, 'Goldilocks zone'. Dry basalt and wet rhyolite of 2.91-5.21 % wt H2O were found to have the same viscosity at over a temperature range of 850-1000oC and therefore it is rheologically feasible for these magmas to mix at Drumadoon. The model further shows that it is rheologically infeasible at specific compositions and temperatures, in part explaining the presence of mingled and not mixed samples at Drumadoon. These calculations combined with Meade et al., 2009 petrological analysis, hand sample and thin section data allowed for the construction of a holistic schematic for magma emplacement at the Drumadoon Sill Complex.

# Wednesday 5:00 Exhibition Hall / Poster Session

# Shedding (laser) light on fluid flow in a porous medium

#### Caitlin M. Chalk

Earth, Ocean and Ecological Sciences, University of Liverpool, Liverpool L69, United Kingdom

The ease at which fluid can pass through a porous bed controls the transport of solutes, heat, and momentum, which has significant implications for a wide range of industrial and natural processes, notably regarding climate change and future energy. For example, enhanced flow of meltwater through glaciers leads to increased melting and global warming; in geothermal systems, renewable energy is harnessed from the localised flow of fluids through hot, porous rocks in sub-surface reservoirs. Characterising different regimes of porous flow is vital for understanding flow mobility in porous systems, yet necessary quantitative flow data are lacking. Small-scale laboratory experiments using idealised, representative materials can provide valuable insight. Previous experimental studies have measured velocities of solid particles that compose the bed, but measurements of fluid velocity are lacking due to the solid obstructing fluid visualisation. An experimental pilot study was conducted to shed light on the fluid dynamics of porous flow, using an advanced state-of-the-art laser imaging system. An idealised, porous bed was created using glass spheres submerged in glycerol, where the refractive index of the spheres and interstitial fluid was matched as closely as possible - allowing for visualisation of the internal fluid dynamics. Fluid - seeded with tracer particles that fluoresce in laser light - was injected into the bed at different rates, and Particle Image Velocimetry was used to measure internal fluid velocities and quantify flow regimes. These experiments have the potential to provide a better understanding of fluid transport through a porous medium.

# Wednesday 5:00 Exhibition Hall / Poster Session

#### PO122

# **Understanding the physical properties of commercial snack products and their influence on digestion dynamics** Alexandros Makrypidis<sup>1</sup>, Zisheng Liao<sup>1</sup>, Maria M. Papathanasiou<sup>2</sup>, Maria N. Charalambides<sup>1</sup>, and Stacie Tibos<sup>3</sup>

<sup>1</sup>Department of Mechanical Engineering, Imperial College London, London SW7 2AZ, United Kingdom; <sup>2</sup>Department of Chemical Engineering, Imperial College London, London SW7 2AZ, United Kingdom; <sup>3</sup>R&D Fellow, PepsiCo, Leicester, United Kingdom

As the consumption of industrially produced food products constantly increases, the food industry shifts towards diversifying their food portfolio to promote a more nutritionally diverse diet. Nevertheless, there is still no clear consensus on how the physico-mechanical properties of native food components influence digestion, and hence how they affect the physiological post-prandial response with respect to nutrient uptake, bloodstream glucose absorption, and satiety sensation. Herein we present, a holistic approach towards understanding how food properties affect digestion via the intertwining of experimental characterization techniques with predictive models which capture the digestion process. In this regard, we create artificial food boluses by varying the moisture content to capture the effect of different mastication residence times and test them at varying strain rates which encapsulate the shear rate range, 0.27/s-0.45/s, to occur in typical Newtonian fluidic meals at the stomach region. Experimental observations, our research shifts towards the derivation of predictive models which predict the effect of moisture content and strain/shear rate on food flow and breakdown within the gastric compartment. The ultimate goal of our research is to integrate experimentally validated models into computational physiological frameworks, to discover the effects of specific food properties on digestion efficiency and nutrient bioavailability. This holistic approach not only advances our understanding of the intricate relationship between food structure and digestion dynamics but also unveils opportunities for tailoring food formulations to optimize nutritional outcomes.

#### PO120

# Wednesday 5:00 Exhibition Hall / Poster Session Simple 2D texture mapping of characteristic food samples from rheological data Fabian Meyer

#### Material Characterization, Thermo Fisher Scientific, Karlsruhe 76185, Germany

Rheological testing stands as a powerful and well-established tool for mechanically characterizing a range of food samples, including liquids, semisolids, and solids. The utilization of rotational and oscillatory measurement techniques yields numerical data, enabling the classification of foods into distinct clusters. For instance, the apparent viscosity at a shear rate of 50 s-1 is used in the National Dysphagia Diet (NDD) framework from 2002 for categorizing thickened drinks for dysphagia patients into various classes including thin, nectar-like, honey-like, and spoon-thick. However, relying solely on a single number from rheological experiments proves inadequate for fully describing a material's textural properties. In food sensory testing, panels engage in blind tastings of different formulations, ranking them, among others, based on textural features like firmness, hardness, cohesiveness, chewiness, or springiness. Establishing connections between these attributes and rheological measurement results facilitates the development of new food formulations, potentially reducing the necessity for extensive sensory panel testing. Complex mapping, incorporating multiple parameters, can be accomplished through advanced analytical techniques such as Principal Component Analysis (PCA). Nevertheless, this contribution focuses on the simplicity of 2D plots to map characteristic food samples, creating meaningful landscapes that group them according to their distinctive textural attributes.

# Wednesday 5:00 Exhibition Hall / Poster Session

#### PO125

# How do cooling die temperature and shear rate influence the texture development of pea protein-based meat analogues

#### Maria G. Gräfenhahn

Institute of Life Technologies, University of Applied Sciences and Arts Western Switzerland, Sion, Switzerland

Meat analogues are usually produced via high-moisture extrusion processing. The combination of heat and shear during extrusion processing causes protein unfolding, rearrangement, and aggregation into a cross-linked structure. Heat transfer from the heated barrel, conversion of mechanical energy into thermal energy, and shearing from the rotation of the screws result in thermal and mechanical stresses acting simultaneously on the protein matrix. During the cooling down of the protein matrix in the die, a fibrous or layered structure develops which intends to mimic meat structure. One theory used to explain the texture development of meat analogues is related to the flow conditions in the cooling die which is often a slit-die. This is due to the fact that a fibrous layered structure is observed when extrudates are teared-up showing a parabolic shape similar to Hagen-Poseuille flow. From this hypothesis, it was concluded that shear would be favorable in the texture development. Consequently, a rotating die was developed and it could be shown that the apparent shear rate in the cooling die influences the texture development. However, we hypothesize that this effect is temperature-dependent, and that the mechanical treatment can enhance it, but it can also negatively influence it. This contribution therefore aims to present and discuss the effect of cooling die temperature and shear rate on the texture development of pea proteins during extrusion processing. To cope with the complexity of the process, rheological analysis of the proteins under extrusion-like conditions were performed. In this way, not only the dynamic rheological properties, but also the non-linear viscoelastic behavior of the proteins as a function of cooling temperature and shear rate could be determined. In this article, the influence of the rheological properties on the resulting product structure and texture is presented in a model system and the effects on the design of meat substitute products by means of extrusion are discussed in detail.

#### Wednesday 5:00 Exhibition Hall / Poster Session

# Modulation of the fibrous structures of high moisture meat substitutes by altering the pH

<u>Felix Ellwanger</u><sup>1</sup>, Melanie Fuhrmann<sup>1</sup>, Ulrike van der Schaaf<sup>1</sup>, Heike P. Karbstein<sup>1</sup>, and Gabriela I. Saavedra Isusi<sup>2</sup> <sup>1</sup>Karlsruhe Institute of Technology, Karlsruhe 76131, Germany; <sup>2</sup>Thermo Fisher Scientific, Karlsruhe 76227, Germany

With the global population steadily rising, the demand for protein is increasing. To meet this demand, plant proteins, precision fermented proteins, and single cell proteins are becoming increasingly important. The extrusion process can be used to produce tasty foods from these proteins, such as meat substitutes and vegan cheese substitutes. To improve the taste and shelf life of these products, salts and acids are often added in the food industry. However, these additives alter the pH and ion concentration, which can affect the structure of proteins and thus influence network formation. This conference contribution focuses onmeat substitute products from two different vegetable proteins: soy protein isolate and vital wheat gluten. These proteins differ in their network formation mechanism. Soy protein isolate forms a physical network, while gluten forms a chemical network stabilized by disulfide bonding. To investigate the influence of altering the pH, materials and parameters are screened using a closed cavity rheometer, followed by extrusion trials at pH 7.36 - 4.14 for SPI and pH 5.83 - 3.37 for gluten. The resulting extrudates are analyzed optically and mechanically, and protein solubility is tested in a reducing buffer. At decreasing pH, the complex viscosity of SPI is increasing, the generated network is becoming stronger, and the cross-linking density is growing. For gluten, a decreasing pH results in altered reaction kinetics, the resulting network is more deformable, and the cross-linking density is decreasing. Solubility tests show that no additional covalent bonds are formed with SPI. With gluten, however, the polymerization reaction is inhibited and fewer disulfide bonds are formed. For both protein systems, the addition of acid results in less pronounced meat-like structures. These new findings are helping to develop plant-based extrudates with improved taste and texture.

# Wednesday 5:00 Exhibition Hall / Poster Session PO127 Structure, Rheological Behavior, and Foam-Stabilizing Potential of Kidney Bean Proteins at the Air-Water Interface

Wanting Yin<sup>1</sup> and Sagis Leonard<sup>2</sup>

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Red kidney beans (Phaseolus vulgaris L.) are globally one of the most consumed legume crops, and an important component of human nutrition, especially because of their high protein content (20 - 30%). There is limited information available on the processing and properties of red kidney bean protein extracts. Therefore, we conducted a comprehensive study on the interfacial properties of red kidney bean protein isolate, as well as its two primary components, globulins and albumins, to elucidate their foaming characteristics. Because of their smaller size, albumins exhibited faster adsorption at the air-water interface and produced foam with higher overrun compared to globulins. We used large amplitude dilatational rheology in combination with the general stress decomposition method to characterize the rheological properties of the air-water interfaces, and observed that albumins formed solid-like interfacial layers with higher stiffness and network density, and stronger in-plane interactions. The globulins created much weaker and more stretchable interfaces, and yielded foam with superior stability compared to the albumin fraction, possibly attributed to the thicker interfacial structure. The behavior of the whole protein mixture was dominated by the globulins. This interface was also weaker and more stretchable in comparison to the albumin interface, resulting in foam with lower overrun yet superior stability relative to albumin-based foam. Understanding these properties is crucial for improving the use of red kidney bean proteins in various food and industrial applications.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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# Large amplitude oscillatory shear (LAOS) for nonlinear rheological behavior of fish myofibrillar protein in paste

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The nonlinear and linear rheological behaviors of fish myofibrillar protein (surimi) paste with different moisture content (75, 82, and 90%) were evaluated using small amplitude oscillatory shear (SAOS) test and large amplitude oscillatory shear (LAOS) test. The linear rheological property (G', G", and viscosity) of surimi paste measured using SAOS indicated a decreased with increasing moisture content. The frequency exponent (n') from power-law equation reflects a relatively low value (0.24 to 0.36) indicating that the imposed deformation rate during SAOS had no discernible effect on the viscoelastic response of the surimi paste. Therefore, SAOS, may not be perfect in describing the shear strain effect during the surimi paste application process such as high shear extrusion process or near failure estimation. The nonlinear rheological response was analyzed using normalized Lissajous-Bowditch curves. The moisture content of the sample has a strong influence on the microstructure of the surimi pastes. The observed nonlinear response with increasing strain during LAOS was attributed to the microstructural changes of surimi paste during intracycle deformations, which displayed elastic straining (0.01 to 5% strain), followed by yielding, flow, and recovery of paste structure at higher strains (>5 to 50% strain).

Wednesday 5:00 Exhibition Hall / Poster Session

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# Investigating rheological characteristics of protein-polysaccharide crosslinked hydrogels for enhanced spinnability of fibres

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The increasing demand for sustainable, protein-rich foods necessitates the advancement of innovative technologies for producing plant- and fermentation-based meat substitutes. These alternatives must not only replicate the texture of conventional meat, which consumers perceive as superior, but also provide well-balanced nutritional profiles. The unique texture of meat results from the fibrous arrangement of collagen fibres or myofibrils in muscle tissues. However, creating similar fibrous structures in plant-based protein substitutes is challenging due to the absence of such organization in most commercial plant proteins like soy or pea. In this work, we propose an innovative use of polysaccharides as structuring aids. Polysaccharide-protein assemblies are designed to achieve structural effects that facilitate the creation of protein-loaded fibrous materials, mimicking the characteristics of meat fibres. To evaluate the fundamental principles that govern spinnability in polysaccharide-protein systems, a model system has been developed based on carboxymethyl cellulose (CMC) as a model polysaccharide and sodium caseinate as a model protein. The binary hydrocolloid system was transformed into a hydrogel using EDC (1-Ethyl-3-[3-dimethylaminopropyl] carbodiimide hydrochloride) as a cross-linking agent. Rheological methods, including steady shear rotational rheometry, small amplitude oscillatory shear rheometry and extensional rheometry (CABER), were used to establish the link between spinnability and viscoelastic properties of the hydrogels. By tuning the conditions of the cross-linking reaction, it is possible to formulate a hydrogel system with optimum gel-setting properties for enabling formation of consistent protein-polysaccharide fibres.

# Influence of biopolymer-water interactions on selected rheological properties of ionic polysaccharide solutions

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Polysaccharides are biopolymers with complex molecular structures that are responsible for their unique properties in aqueous solutions, including rheological properties. These are shaped by, among other things, biopolymer-solvent and biopolymer-biopolymer interactions, with the length of the main (linear) chain, the presence and nature of branching or chain dispersion playing a key role. The second factor is the biopolymer concentration which, together with the affinity for the solvent, determines the polymer-polymer interactions. Due to the nature of the interactions in the physico-chemistry of (bio)polymers, three concentration ranges are distinguished - dilute, semidilute and concentrated - separated by two critical concentrations (c\* and c\*\*). Knowledge of the interactions between polysaccharides and water as a solvent is used in practice to shape selected functional properties (e.g. rheological, textural and water-binding capacity) of many food products. Polysaccharides are used as thickeners, gelling agents, emulsifiers and anti-syneresis agents. The group of polysaccharides commonly used as food additives includes the ionic biopolymers: xanthan gum (E415), konjac gum (E425) and a cellulose derivative: cellulose gum (E466). Shear and extensional rheological properties were measured for aqueous solutions of the above hydrocolloids at different concentrations. The shear rate dependence of apparent viscosity and N1 and the elongational rate dependence of apparent extensional viscosity were determined. The modified DeKee and Papanastasiou model was fitted to these data and the values of the characteristic time constants were estimated. The dependence of N1 on shear rate was used to interpret the structural behaviour of the studied fluids during shearing. The influence of the concentration and molecular structure of the polysaccharide on the mechanism shaping the rheological properties of the model solutions was discussed.

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# Wednesday 5:00 Exhibition Hall / Poster Session

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**Influence of extraction temperature on the rheological properties of Plantago Ovata husk mucilage solutions** <u>Anna Ptaszek</u>, Marta Liszka-Skoczylas, Magdalena Witek, and Urszula Goik

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Polysaccharides extracted from plant seeds constitute a broad group of hydrocolloids used in the food industry due to their strong antioxidant properties, high prebiotic potential, high water absorption and gelling capacity. From a technological point of view, these properties make them valuable food additives that modify the rheological properties of products, stabilise multiphase gas-liquid and liquid-liquid systems and counteract syneresis through their ability to permanently retain water in the food structure. This group of hydrocolloids is obtained by extraction from the endosperm of seeds (galactomannans and hemicelluloses) and from the husk (polysaccharide-mucilage fraction). As indicated in the literature, the use of several stages of extraction of Plantago ovata seed hulls, under conditions of increasing temperature, results in the extraction of mucilage fractions with increasing average molecular weights and increasing arabinose/xylose ratios. Therefore, the aim of this study was to determine the effect of increasing average mass fraction of polysaccharides on selected rheological properties of the extracts. The dependence of apparent viscosity and N1 on shear rate was determined. The modified DeKee and Papanastasiou model was fitted to these data and the values of the characteristic time constants were estimated. The dependence of N1 on shear rate was used to interpret the structural behaviour of the studied fluids during shear flow. The influence of the concentration and average molecular weight of the polysaccharides on the mechanism shaping the rheological properties of the extracts was discussed.

# Wednesday 5:00 Exhibition Hall / Poster Session

# **Rheomicroscopy of starch gelatinisation**

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Rheomicroscopy offers simultaneous monitoring of microstructure through microscopy and rheology, in large deformation and small deformation oscillating shear as well as in compression. The method was used to visualise and explain starch gelatinisation and how it is affected by shear, granule architecture and botanical origin. Shear had a strong influence on paste viscosity and was explained by granule disruption following swelling. Gelatinisation occurred during narrower temperature range for potato starch than for maize starch, which in turn both had a different behaviour from wheat starch. A-type wheat starch was also shown to have different gelatinisation behaviour than B-type wheat starch.

# Wednesday 5:00 Exhibition Hall / Poster Session

# Ascorbic acid mediated hydrolysis of galactomannans

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Ascorbic acid (AA), also known as vitamin C, is an antioxidant widely used in the food industry to prevent colour fade and spoilage. This study assesses the effect of AA on the rheology of common food thickeners - galactomannans (GM). 0.02 wt% AA was added, a typical level for exerting its antioxidant activity. GMs immediately exhibit a significant reduction in the viscosity of their solutions upon the addition of AA: guar gum (- $69\% \pm 7\%$ ), locust bean gum (- $47\% \pm 5\%$ ), and cassia gum (- $58\% \pm 4\%$ ). This held true when GMs were embedded in a gelling matrix of xanthan gum (XG) +/- low acyl gellan gum (LAG). AA's impact was assessed using small amplitude oscillatory shear rheology. As the temperature decreased, the storage modulus decreased in the presence of AA compared to without. To understand the mechanism behind these rheological

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observations, the molecular weight (MW) of the GMs +/- AA, was assessed using size exclusion chromatography - multi angle light scattering. This showed a marked reduction in MW, between 6-8 times for each GM, and was confirmed with analytical ultracentrifugation. This established the hydrolytic decomposition of GMs in the presence of AA, leading to a decrease in function due to a reduction in MW. This hydrolytic effect occurred at pH  $3.5 \pm 0.1$  and pH  $6.7 \pm 0.3$ , showing that acid hydrolysis is not the primary mechanism. The behaviour of GMs with other food acids at pH  $3.5 \pm 0.1$  and pH  $6.7 \pm 0.3$ , showing that acid hydrolysis is not the primary mechanism. The behaviour of GMs with other food acids at pH 3.5 showed no decline in viscosity or MW. Potassium iodide, another reducing agent caused no hydrolytic effect, indicating it isn't caused by all reducing agents. When AA was added to XG or LAG at both pH values, no major changes were observed in viscosity or the storage modulus, respectively. In conclusion, this study uncovers a previously unreported interaction between GMs and AA. These findings are of particular interest, with their potential to impact a wide range of products; influencing their quality and storage stability, as well as affecting the cost-effectiveness and environmental life-cycle of food ingredients.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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# Comparable analysis of rheological and tribo-rheometrical properties of milk and plant-based milk alternatives

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Both, texture, and mouthfeel, are highly related to the microstructure of foods and are therefore crucial terms for consumer choice and acceptability. Consequently, to enhance customer acceptance, both formulation and production processes must be tailored to achieve certain flow properties, texture and mouthfeel accordingly.

Eating and swallowing are a highly dynamic process, and the oral processing undergoes various stages. At first, a bulk dominant stage takes place involving flow and deformation under shear, compression, and elongation. After that, the oral processing transitions to surface property dominant characterized by closed contact interaction and lubrication of oral surfaces. Rheology is already a widely used technique which serves the characterization of the bulk property dominated regime. However, given that the oral cavity comprises of the tongue touching and sliding on the palate in the presence of a lubricating food product, tribological measurement techniques are required to quantify the frictional properties of foods. For this reason, this technique is gaining more popularity as a research tool. However, the evaluation of the obtained data is still a topic for discussion.

In order to showcase the use of tribology as industrial relevant tool to quantify and compare mouthfeel, this contribution focuses on the impact of different types of commercially available plant-based milk alternatives on the rheological as well as tribo-rheometrical properties compared to different cow milk references. For this, a rheometer equipped with a tribo-rheometrical measuring geometry is used and Stribeck curves together with the respective rheological flow properties of each food product will be compared. This research contributes to understanding the differences between plant-based milk alternatives and their animal-based references regarding their flow and lubricating properties. This has the potential to leverage the variety of options available to meet the consumer's expectations.

#### Wednesday 5:00 Exhibition Hall / Poster Session

# Shear rheology and adsorption measurements unveiling the mucoadhesive properties of lactoferrin

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Mucoadhesion refers to the adherence of a material to soft mucosal layers, presenting opportunities for coating and protecting damaged tissues, and facilitating the localized delivery of agents. In the present study, we focus on understanding how the thermal denaturation of proteins can generate such mucoadhesive materials using lactoferrin (LF), a milk protein as a model system. With lactoferrin's tendency to denature at temperatures higher than 65 °C, we explore the impact of thermal processing on its adsorption properties to bovine submaxillary mucin (BSM). A combination of rotational rheology using a double-gap geometry and quartz crystal microbalance with dissipation monitoring (QCM-D) showed how different degrees of thermal denaturation influence lactoferrin's interaction with mucin. Results show that post-denaturation, the viscosity of a diluted lactoferrin sample (1 wt%) remains unchanged, and a shear-thinning behavior was observed from the reduction in viscosity with increasing shear rates. There is an enhancement in the viscosity values of mucin and lactoferrin-BSM complexes unveil the rheological synergism usually associated with mucoadhesive materials, as even at these low concentrations the interactions between BSM and LF lead to the formation of larger structures. Confirmation of BSM interactions with LF comes from LF adsorption onto mucin, using QCM-D sensors. The denatured form of lactoferrin adsorbs to bovine submaxillary mucin at a larger extent, compared to its native form. These latest findings underscore the heightened mucoadhesivity of denatured lactoferrin, attributed to its positive surface charge, surface hydrophobicity, and thiol group presence. This research provides valuable insights into the dynamic interplay between lactoferrin and mucin during thermal denaturation, paving the way for innovative applications across diverse chemical sectors.

# Wednesday5:00Exhibition Hall / Poster SessionPCInvestigation of the interfacial properties of yeast cells (Saccharomyces cerevisiae) and their components in<br/>emulsion-based foodsPC

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#### Yeast cells as an alternative source of protein in food

Proteins are an important part of the human diet. For reasons of climate and animal protection, there is an ever-increasing demand for new protein sources as an alternative to animal-based options. Vegetable raw materials, but also microorganisms such as yeasts, come into consideration for this. Yeasts are a promising raw material because of their nutritional composition, decentralized and weather-independent cultivation and their acceptance by consumers.

#### Interface stabilization by yeast components

To formulate alternative yeast-based food, knowledge about the techno-functional properties of yeasts and their components is necessary. Yeasts from the Saccharomyces cerevisiae strain possess mannoproteins on the outside of the cell wall that are known to have emulsion-stabilizing properties. In addition, there are solved proteins inside the cell that can be released during cell disruption or lysis. The existing knowledge regarding the interface stabilizing properties of these proteins is not yet sufficient.

#### Investigation of the interfacial functionality of yeast protein

The solved proteins were gained by cell disruption using highpressure homogenization and centrifugation of the non-soluble components. To investigate the interfacial functionality at a model interface we use pendant drop tensiometry and carry out dynamic and oscillating experiments. The first one allows conclusions about the interfacial tension, the latter about the elasticity of the interfacial film formed by proteins. This information is relevant for the breakup and stabilization step in the emulsion production. Pendant drop and emulsification experiments are carried out by varying formulation and process parameters as well as different pretreatments of the proteins.

# Wednesday 5:00 Exhibition Hall / Poster Session

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# Characterisation and interfacial properties of foams stabilised by mung bean proteins and pectin

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Mung beans are a rich source of protein with good foaming properties. However, their application in foods is limited due to its low solubility under acidic conditions. Previous studies have reported that the solubility of proteins can be improved by mixing with polysaccharides. Therefore, we aimed to investigate the interfacial properties of mixtures of low methoxyl pectin (LMP) and mung bean protein isolate (MBPI). The electrostatic interactions between MBPI and LMP were characterised at different pH and ratios, to construct the state diagram (Fig. 1a). The pH dependent complex formation was investigated via turbidity measurement. The stability and foamability of MBPI and mixtures were explored. Our results revealed that, the introduction of LMP to MBPI solution could improve its functionality near the isoelectric point. The maximum turbidity of the mixture was lower compared to native MBPI, indicating the addition of LMP could reduce the self-aggregation of MBPI. To understand the interfacial properties of the mixture, a 1:1 ratio of MBPI/LMP solution under pH4 was subjected to large amplitude oscillatory dilatation and shear rheology. The surface microstructure was analysed by imaging with atomic force microscopy. We used Lissajous plots to characterise the non-linear responses and observed that both native MBPI and the mixture stabilised the interface by forming a viscoelastic solid film. The mixtures showed higher surface dilatational moduli, suggesting the interfacial film became stiffer. For the shear behaviour, the presence of LMP resulted in the formation of a stiffer interface in the linear regime, however, a less stiff and more viscous interface was observed at large deformation. The foam stability of the mixtures was improved because of the higher stiffness of the interfaceal behaviour which can be linked to foaming properties.

# Wednesday 5:00 Exhibition Hall / Poster Session

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# Application of the kinetic equation for changes in fluidity (D) to food foams stabilised by selected polysaccharides

# Pawel Ptaszek and Anna Ptaszek

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This paper presents the results of modelling the non-linear rheological properties of food foams with the character of colloidal glasses. The foams were obtained on the basis of ovalbumin and selected non-starch hydrocolloids: carboxymethylcellulose and inulin. Basic physical parameters were determined for the obtained foams: overrun, density and gas phase fraction. Rheological measurements based on large amplitude shear oscillations (LAOS) were carried out. The LAOS measurements were carried out in the range of strain amplitudes ?o from 0.001 to 50, at a fixed frequency of 1 Hz. All tests were performed at a temperature of 23oC. As a result, time histories were obtained and presented in the stress-strain phase plane in the form of Lissajous figures. A mathematical model based on the shear stress equation and the kinetic equation [1] was fitted to the data obtained. The model required the estimation of seven parameters for which the physical interpretation was discussed.

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Reference: [1] Carrier, V., Petekidis, G., Nonlinear rheology of colloidal glasses of soft thermosensitive microgel particles, Journal of Rheology, 2009, 53(2), 245-273, doi:10.1122/1.3045803

#### Wednesday 5:00 Exhibition Hall / Poster Session

# Ionic strength effect in the equilibrium and rheological behavior of an amphiphilic triblock copolymer at the vapor-solution interface

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This study investigates the effect of an inert salt (NaCl) on the equilibrium interfacial tension and dilatational modulus of Pluronic F68 copolymer, a triblock copolymer consisting of two terminal blocks of poly(ethylene oxide) and a less hydrophilic central block of poly(propylene oxide). Interfacial tension measurements were carried out using a surface force balance and a drop shape tensiometer, while rheological measurements were carried out in two different frequency ranges. This involved the use of the oscillatory barrier/droplet method and electrocapillary wave measurements, complemented by an appropriate theoretical framework. This work aimed to elucidate the influence of NaCl on the interfacial behavior of Gibbs monolayers of Pluronic F68. In addition, this study highlights some of the technical and theoretical limitations in obtaining reliable dilatational rheological data at high frequencies (<1 kHz) using electrocapillary wave measurements. The results provide valuable insights into the interplay between salt presence and interfacial properties of Pluronic F68 and highlight the challenges of obtaining accurate dilatational rheological data under specific measurement conditions, stressing on the limitations of the dispersion equation describing the propagation of capillary waves. The experimental work proved that high NaCl content (>1000 mM) produced significant variations in the surface tension (salting-out effect), tunes the bulk viscosity of Pluronic F-68 solutions, and, in certain conditions, changes the surface rheology of the monolayer (improving foam stability). Furthermore, the study shows how in extremely high ionic strength mediums the resonance condition is not satisfied, giving a contribution to understand the limitations of the dispersion equation used to determine the surface dilational modulus.

#### Wednesday 5:00 Exhibition Hall / Poster Session

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# Rheological investigation of chain modified recycled polyethylene terephthalate/polybutylene terephthalate blends and their corresponding foaming behavior

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To address the growing concerns about plastic waste and enhance the sustainability of polymers, recycling stands out as the initial and straightforward step. However, commonly used polymers, such as Polyethylene terephthalate (PET), face challenges in recycling that affect melt and rheological properties. To recover lost properties and improve existing ones, chain modification and blending provide easily implementable and cost-effective solutions. Both epoxy-based (e.g., Joncryl) and anhydride-based (e.g., PMDA) chain extenders have been successfully used. In this study, recycled PET (rPET) and polybutylene terephthalate (PBT) underwent melt compounding using a melt mixer with chain modifiers (i.e., Joncryl ADR 4468 and PMDA). rPET, PBT, and rPET/PBT blends with ratios of 75/25, 50/50, and 25/75 were melt compounded with the addition of 0.25, 0.5, 0.75, and 1.0wt% of each chain modifier. Melt properties were examined using small amplitude oscillatory shear (SAOS) rheological analysis to reveal the effect of chain extender addition in all samples. Furthermore, extensional viscosity and foaming behavior were also examined through sentmanat extensional rheology (SER) and batch-based autoclave foaming, respectively. Both chain extender additions above 0.25 wt% resulted in significant enhancements in complex viscosity and storage modulus values in rPET and blends. While Joncryl showed substantial improvements in the melt properties of all blend systems, PMDA was unreactive with neat PBT. The addition of chain extenders also increased the extensional viscosity of samples and imparted strain-hardening behavior with higher chain extender contents. The foaming behavior of the modified samples was also improved, with clear observations of enhancements in cell stability and foam expansion ratio.

Wednesday 5:00 Exhibition Hall / Poster Session

# How accurate are pendant drop methods in measuring surface rheology?

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Obtaining surface tension values from sessile or pendant drops and bubbles has been a well-established technique for many years, and several methods have been developed to determine the surface and rheological properties of these drop/bubble interfaces. In this work, we explore how these established methods are affected when the liquid surface develops a "skin", which may arise due to the presence of e.g. proteins, polymers, or particles that can laterally interact and form a two-dimensional network with mechanical strength. This skin not only introduces additional complexity in terms of the interface's resistance to changes in area (compressional rheology), but also imparts additional resistance to alterations in shape (shear rheology). Thus, a consideration of the surface stress - which includes both mechanical and thermodynamic contributions - rather than just the surface tension is needed to adequately characterize the response of complex interfaces to deformations encountered in pendant drop techniques. As a result, both the theoretical basis and the practical considerations for identifying valid operating conditions in pendant drop tensiometry have to be revised. In this work, we numerically assess the performance of different methods in pendant drop tensiometry based on their ability to predict strains, stresses, and dilatational moduli following the step compression of drops with perfectly elastic interfaces. The chosen techniques differ on the drop's size and geometry and on the way the surface stress and strain fields are obtained. We find that although

the surface stress can be predicted with a good degree of accuracy, all methods face inherent limitations when it comes to correctly determining surface rheological parameters. Furthermore, we provide recommendations on the preferred experimental configurations, the treatment of acquired data, and the use of the correct analytical and constitutive models.

### Wednesday 5:00 Exhibition Hall / Poster Session

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# Nanoscale mastery: precision control of non-newtonian droplet impacts via surface acoustic wave devices and nature inspired coatings

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This study explores multi-approaches for altering non-Newtonian droplet impacts, leveraging nanoscale surface vibrations and advanced surface coatings. The nanoscale vibrations are induced through thin-film Surface Acoustic Wave (SAW) devices, while hydrophobic and superhydrophobic coatings, inspired by nature, tailor the surface interactions. The integration of these technologies offers unprecedented control over droplet behaviour, unlocking applications across diverse industries. Surface Acoustic Wave (SAW) are nanoscale vibrations on zinc oxide films, harnessed through the piezoelectric effect, presenting an innovative means to manipulate non-Newtonian droplet impact with remarkable precision. The ability to adapt the thin-film zinc oxide to any underlying substrate illustrates the highly adaptable nature of this approach. Complementing the vibrational aspect, nature-inspired hydrophobic and superhydrophobic coatings play a pivotal role in dictating droplet interactions. These coatings, mimicking such examples as the lotus leaf's water-repellent properties, optimize wetting and spreading characteristics. The tailored interplay between surface coatings and nanoscale vibrations showcases a synergistic approach for achieving precise control over non-Newtonian droplets, with potential applications in inkjet printing, pharmaceutical manufacturing, and agricultural systems. The obtained results illustrate the use of superhydrophobic coating to alter the droplet shape after impact. Further investigations illustrate that by mimicking the lotus leaf structure, a reduction of up to ~50% in contact time of the impacting droplet can be achieved. By proposing this synergistic approach of nanoscale hydrophobic coating and nanoscale vibrations, contact times for lower viscosity liquids can be reduced by up to ~25%. For more viscous liquids outcomes of droplet impact regimes can be altered to promote rebound instead of deposition.

# Wednesday 5:00 Exhibition Hall / Poster Session

# Immersed viscoelastic jets in microfluidics

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In this work we investigate the formation of viscoelastic jets in a microchannel in contraction flow. The microfluidic device has three inlets (main inlet and secondary branches), one outlet (exit channel) and a junction where the contraction is set up. As in the classical viscoelastic studies, with the 4:1 contraction, the PEO (poly ethylene oxide) solutions are introduced in the main channel (approx. 400 micrometers width) and they flow to the exit channel (approx. 100 micrometers width). From the secondary branches of the microchannel, positioned at 30 degrees to the exit channel, the Newtonian fluid (mineral oil) is introduced, in this way the interface between the viscoelastic fluid and the Newtonian fluid is formed. In this study, there are two regions of interest: i) the junction and ii) the exit channel. In case of a mismatch in the flow rate ratios, such as a high flow rate on the secondary branches, or a high flow rate on the main channel, the channel is subjected to back flow. In the case of the viscoelastic fluid (I), there are flow instabilities occurring, such as droplet formation, slug flow. However, when a critical flow rate imposed, we obtain the beads-on-a-string phenomenon, with different widths and distances between the beads. At the same critical flow rate imposed, in the case of a Newtonian fluid (II) introduced on the main channel, the obtained interface is stable. All the cases are analyzed in terms of nondimensional numbers and showcase the importance of the viscoelasticity and the viscosity ratio between the two phases.

# Wednesday 5:00 Exhibition Hall / Poster Session

# Numerical simulation of platelet aggregation in microfluidics

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Microfluidics has become an indispensable tool for mimicking complex physiological processes and disease mechanisms, enabling researchers to gain valuable insights into biological systems at the microscale. Platelet aggregation, while representing a critical step in hemostasis, i.e., the physiological response that suppresses bleeding, can also be a serious problem under pathological conditions. Indeed, an altered platelet aggregation can lead to the formation of thrombi, thus playing a pivotal role in vascular health and diseases such as myocardial infarction and deep vein thrombosis. Although numerous experimental and numerical studies have been carried out to understand the mechanism governing these complex phenomena, the picture is not yet clear and many questions remain, especially at the particle level. In this contribution we investigate platelet aggregation and wall deposition within blood on chip devices using Computational Fluid Dynamics coupled with Discrete Element Method (CFD-DEM) simulations. Platelets are modeled using the multi-sphere approach, in which each particle is represented by a set of connected spheres that are treated as a rigid body. The effect of flow conditions, particle volume fraction, aspect ratio, and cohesion forces are investigated.

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# A Hele-Shaw cell consists of a liquid sandwiched between two parallel plates at a fixed separation distance. When one plate is lifted from the other, a pressure differential is created which forces air into the liquid. As the liquid recedes, a Saffman-Taylor instability develops at the liquid-gas interface, which evolves into long fingers, leaving a branched liquid pattern behind [1]. Control over the branching pattern can be achieved by strategically introducing asymmetry to one of the plates and controlling air entry points. Such asymmetry can be introduced by the addition of

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air holes [2]. Harnessing the effect of asymmetry on air flow is important to control pattern formation. This work experimentally investigates the effect of single holes (varying in size) on pattern formation. An air hole introduces another liquid-air interface which initiates air fingers from the centre to the outer liquid-air interface. The two air fingers create a ring of liquid at a radial distance between the outer liquid-air interface and the central hole. The height, width and inner radii of the ring is characterised for different hole sizes, separation distances and lift speeds. By understanding the effect of hole sizes on pattern formation, strategies can be developed to precisely control the air finger formation, producing controlled multiscale features. Such methods could be used to mimic leaf venation patterns, utilizing their efficient nutrient transport properties. Such bio-inspired features can be used for applications in charge, heat and mass transport while providing an aesthetic benefit.

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# Wednesday 5:00 Exhibition Hall / Poster Session

Wednesday 5:00 Exhibition Hall / Poster Session

David K. Roughton-Reay, Agrawal Prashant, and Barrioz Vincent

Impacts of hole size on pattern formation in lifted Hele-Shaw cells

# Application of planar elongational flow-based microfluidic device to the characterization of the morphology of non-spherical graphite utilizing viscoelastic particle focusing

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The production of high-quality and cost-effective batteries becomes a major task as they are used from small portable devices to electrical vehicles (EVs). Graphite is a commercially used anode active material and takes up most of the anode slurry. Due to its non-spherical shape, the rheological properties of the slurry considerably depend on its shape and size. Also, it is evident that the shape and size of the graphite affect the electrical performance of the battery since the lithiation process occurs through the edge plane of the graphite during the battery charging process. Despite the growing importance of controlling and measuring the morphology of the edge plane of the graphite, there were no proper quantificational techniques. In this work, we demonstrated that the size and shape of graphite particles could be successfully characterized with our novel microfluidic method. Spherized natural graphite was used as a proof-of-concept study for our work. Ball-milled natural graphite was prepared to examine the impact of physical milling on shape and size change. We fabricated a four-walled PDMS cross-slot microchannel for the particle-analyzing device. The microfluidic chip can be utilized to align the graphite particle to its edge plane in the compressional force in the elongational flow field at the cross-slot stagnation point. We used polyvinylpyrrolidone (PVP) solution as particle-suspending medium for the viscoelastic particle focusing. The graphite particles are focused along the channel centerline due to the elastic force exerted by the viscoelastic fluid, facilitating the characterization of the individual particles. To investigate the effect of the graphite shape on rheological properties, we prepared anode slurry with natural and ball-milled graphite, respectively. The shear rheological properties of each anode slurry were measured. The effects of the morphological difference of the graphite on the battery's performance was also investigated.

# Wednesday 5:00 Exhibition Hall / Poster Session

Living cells as a biological analog of optical tweezers – a non-invasive microrheology approach <u>William Hardiman</u><sup>1</sup>, Matt Clark<sup>1</sup>, Claire Friel<sup>2</sup>, Alan Huett<sup>2</sup>, Fernando Pérez-Cota<sup>1</sup>, Kerry Setchfield<sup>1</sup>, Amanda J. Wright<sup>1</sup>, and Manlio Tassieri<sup>3</sup>

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Microrheology, the study of fluids on micron length-scales, promises to reveal insights into cellular biology, including mechanical biomarkers of disease and the interplay between biomechanics and cellular function. Here a minimally-invasive passive microrheology technique is applied to individual living cells by chemically binding a bead to the surface of a cell, and observing the mean squared displacement of the bead at timescales ranging from milliseconds to 100s of seconds. Measurements are repeated over the course of hours, and presented alongside analysis to quantify changes in the cells' low-frequency elastic modulus, G'<sub>0</sub>, and the cell's dynamics over the time window  $\sim 10^{-2}$  s to 10 s. An analogy to optical trapping allows verification of the invariant viscosity of HeLa S3 cells under control conditions and after cytoskeletal disruption. Stiffening of the cell is observed during cytoskeletal rearrangement in the control case, and cell softening when the actin cytoskeletal rearrangement. This is, to our

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knowledge, the first time that cell stiffening has been measured during focal adhesion maturation, and the longest time over which such stiffening has been quantified by any means.

#### Wednesday 5:00 Exhibition Hall / Poster Session

Machine learning opens a doorway for microrheology with optical tweezers in living systems

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It has been argued that linear microrheology with optical tweezers (MOT) of living systems "is not an option" because of the wide gap between the observation time required to collect statistically valid data and the mutational times of the organisms under study. Here, we have explored modern machine learning (ML) methods to reduce the duration of MOT measurements from tens of minutes down to one second by focusing on the analysis of computer simulated experiments. For the first time in the literature, we explicate the relationship between the required duration of MOT measurements  $(T_m)$  and the fluid relative viscosity  $(?_r)$  to achieve an uncertainty as low as 1% by means of conventional analytical methods, i.e.,  $T_m \approx 172r^3$  minutes, thus revealing why conventional MOT measurements commonly underestimate the materials' viscoelastic properties, especially in the case of high viscous fluids or soft-solids. Finally, by means of real experimental data, we have developed and corroborated an ML algorithm to determine the viscosity of Newtonian fluids from trajectories of only one second in duration, yet capable of returning viscosity values carrying an error as low as ~0.3% at best, hence opening a doorway for MOT in living systems.

# Wednesday 5:00 Exhibition Hall / Poster Session

# Measuring the viscoelastic properties of microscopic soft particles using time-dependent Capillary Micromechanics

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Capillary Micromechanics is a method that enables measurement of the elastic properties of soft, microscopic objects such as biological cells or hydrogels. The method employs a tapered capillary with a tip diameter smaller than the particle size. Consequently, as a suspension of particles is flown through such a capillary, a single particle becomes trapped, blocking further flow. By measuring the change in volume and shape as a function of the applied pressure, both the compressive and shear elastic modulus of the particle can be extracted. Here we show a semi-automated implementation of Capillary Micromechanics, where, once a particle is trapped in the tapering section of the capillary, the process of pressure regulation and image acquisition is automated. This has improved the repeatability of the measurements and the ease of operation. Moreover, the semi-automation has enabled us to conduct time-dependent stress-controlled tests analogous to a creep test and oscillatory test known from conventional rheometry. For the creep test, we apply an instantaneous increase in pressure. By measuring the resultant time-dependent volumetric strain and shear strain, we can extract the time-dependent response of the particle, which depends on the inherent viscoelastic properties of the particles, but also on poroelastic effects, which are strongly dependent on the particle volume. We validate and test our new approach through experiments on microscopic polyacrylamide (PAAm) hydrogel particles fabricated in-house.

# Wednesday 5:00 Exhibition Hall / Poster Session

# An innovative 3D Printed rheometric tool to study chemorheology

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Many systems with a complex rheological behaviour, such as colloidal solutions and gels, are used in the field of cultural heritage conservation. Some of these products are used as substrates for cleaning surfaces of historical interest. In recent decades, high-viscosity polymer dispersions of PVA cross-linked with Borax have been used as low-impact cleaning tools for painted surfaces. Their growing popularity stems, in large part, from the possibility of exerting better control of the cleaning action by reducing the problems arising from the penetration of aggressive solvents into the pictorial layers. These systems are characterized by a high elasticity that allows their safe removal through an exfoliation action. The addition of different organic solvents to these gels, up to a maximum of 30% by weight, used for the removal of contaminants from artistic surfaces, is possible during the early stages of gelation. To optimize solvent mixing and efficiency of the formed gel, it is fundamental to monitor the gelation kinetics, which is often too fast to be properly controlled. For this purpose, i.e. follow chemorheology of specific systems in the best possible way, a new rheometer tool was specially designed, and 3D printed to monitor very fast kinetics in situ. This tool, applied to an Anton Paar rheometer, was tested and its effectiveness verified not only for restoration products but also for other materials. Transient experiments were conducted to track the gelation dynamics of an 8% aqueous solution of PVA cross-linked with a 4% aqueous solution of sodium tetraborate. Moreover, the conformational transitions due in this case to physical links between molecules of the ionic surfactant Cetylpyridinium Chloride (CPyCl) upon addition of the sodium salt Diclofenac were also monitored and compared to data available in literature. Finally, the acid hydrolysis kinetics of k-Carrageenan was studied and compared to results obtained with the classical double helix Couette cell.

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# First normal stress difference of rubber compounds at high shear rates via capillary rheometer, basis for the estimation of material processability

Masood Khabazian Esfahani and Joachim Sunder

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First normal stress difference of polymeric materials are measured by cone-plate geometry via rotational rheometers in startup shear experiment [1]. In this experiment, it is necessary for the cone-plate geometry to reach to a gap at a specific distance (gap) before the experiment starts. Materials with long relaxation times e.g. rubbers are hard to test in this geometry since they resist the downward force of the rheometer 'go to gap' function. Even if the gap is reached for these materials, during the test, they generate a lot of normal force and torque which limits the measurements only at very low shear rates. With above mentioned reasons, measuring the first normal stress difference for rubber materials is almost not possible even at moderate shear rates around 1 s-1. However, this shear rate is well below the shear rates that the rubber materials experience in the processing operation such as extrusion and injection molding. To overcome this problem, a special die adaptable to capillary rheometer is developed [2]. The die is able to simultaneously measure steady-stare shear viscosity and steady-state first normal stress difference at high shear rates of around 4000 s-1. The correlation between first normal stress difference, entrance pressure loss and material processability was found. [1] Schramm G., A practical approach to rheology and rheometry. Haake 1994. [2] Khabazian Esfahani M., Georgantopoulos C.K., Naue I.F.C., Sunder J., and Wilhelm M., Journal of Applied Polymer Science 139 (2022) 18.

#### Wednesday 5:00 Exhibition Hall / Poster Session

Advanced extensional rheometry experiments on a rotational rheometer platform

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Performing extensional measurements on a rotational rheometer has long been established for solid samples using rectangular fixtures and for films using extensional fixtures incorporating counter-rotating drums like Sentmanat Extensional Rheometer (SER) or Universal Extensional Fixture (UXF)[1]. In this work, we show how the modularity of a rotational rheometer platform can be leveraged to extend the measuring range of existing methods and allow use of techniques that previously required a separate device.

In particular, we show how incorporating a second motor into measurements with a UXF can greatly improve the available torque range, thereby extending the measuring range to lower shear rates/ lower viscosity samples. The same setup can also be used to further enhance the method recently suggested by Parisi et. al [2] where the sample is mounted at an angle in order to achieve increased maximum deformations. We improve upon this method by additionally controlling the vertical movement of one of the UXF drums to actively control the inclination angle during measurement. Having optical access to the measurement setup during the entire experiment allows validation of the inclination angle.

Finally, we show that by equipping the rheometer with a lower linear drive and combining the setup with a high-speed camera, we can perform capillary breakup experiments on low viscosity liquids. The rapid motion enabled by the linear drive and an optimized optical setup are required to perform these measurements at a level that previously required a separate device.

Due to the modularity of the rheometer platform, we can also explore the use of polarized imaging for the presented applications.

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#### Wednesday 5:00 Exhibition Hall / Poster Session

# PO155 Exploring fluids' behavior: Lab-scale capillary breakup extensional rheometer and its applications in polymer processing

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The flow behavior of low-viscosity complex fluids under extensional deformation is of paramount importance to several processes and technologies, such as electrospinning or spray drying. However, its characterization is far from easy and the study of this behavior is still in its infancy. To achieve this rheological characterization, a Capillary Breakup Extensional Rheometer (CaBER) can be used. This instrument observes the evolution in the diameter of a liquid filament obtained by step-motion separating the two plates sandwiching that liquid. By modelling the filament thinning as a function of time, and assuming that the liquid surface tension is known, it is possible to infer its extensional viscosity and relaxation time [1, 2]. The current difficulties to commercially purchase this type of extensional rheometer hampers the progress of the scientific field. This work presents the design and production of a lab-scale CaBER, outlining the specific mechanical, optical and software systems of the machine. The effectiveness of this technique was found to depend on three interrelated parameters: the diameters of the plates and the initial and final gaps between them. The influence of these parameters on the success of the experiments was evaluated. The reliability of lab-scale CaBER rheometer developed in this work was successfuly tested using Newtonian solutions. It was shown that the measured extensional viscosities of these fluids were approximately three times their shear viscosities obtained using a rotational rheometer. Viscoelastic solutions were also tested and their caracteristic extensional relaxation times were determined. These data were used to predict the processability and characteristics of nanofibers obtained by electrospinning.

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# Wednesday 5:00 Exhibition Hall / Poster Session

# Fatigue analysis via Fourier Transform of the stress

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The fatigue behaviour of several polymers (e.g. PS, PS-PI block copolymers, PE) under oscillatory torsion and large shear amplitudes was analysed via the Fourier Transform (FT) of the stress to quantify nonlinear contributions as higher harmonics.<sup>1,2</sup> The tests were conducted in the solid state at room temperature using rectangular and rotational dog-bone shaped specimens and were filmed by a video camera to monitor changes in the sample such as cracks. The time evolution of the linear parameters (G'(t) and G''(t)) and the nonlinear parameters (higher harmonics, i.e.  $I_{2/1}(t)$ ,  $I_3/1(t)$ ) as well as their cycle number dependent derivatives and integrals were analysed to detect and describe specific events (e.g. crack growth and propagation), to predict the failure lifetime and to establish failure criteria. A monotonic decrease was found for the linear parameters (G', G''), while the intensity of I3/1 (relative amplitude of the third harmonic to the fundamental one) steadily increased until failure.<sup>3</sup> Shortly after the beginning of the experiment, the behaviour of these three parameters were found to change linearly with time (number of cycles) until failure occurs. For the undamaged specimens, the nonlinear parameter I2/1 (relative amplitude of the second harmonic to the fundamental one) is within the noise level, however its intensity increases with the appearance of macroscopic cracks.<sup>4</sup>

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Wednesday 5:00 Exhibition Hall / Poster Session

# Measuring the deflection of the interface between a Newtonian and a viscoelastic fluid to determine the second normal stress difference

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In simple shear, the properties of a complex fluid are fully described by the three material functions viscosity ( $\eta$ ), the first and the second normal stress difference (N<sub>1</sub> and N<sub>2</sub>). Among the three, only N<sub>2</sub> is not part of the routine set of parameters to rheologically describe a material. This is because even today N<sub>2</sub> is still neither easy nor accurately measurable with standard techniques, despite the fact that it is considered as the main property controlling effects as the onset of secondary flows and elastic instabilities. Wineman and Pipkin<sup>[1]</sup>, first theoretically, and Tanner<sup>[2]</sup> later experimentally, demonstrated that a viscoelastic fluid flowing through a trough experiences a deformation of the free surface that corresponds to the magnitude and sign of N<sub>2</sub>. Based on this concept, we designed and optimized a novel technique to measure N<sub>2</sub> for dilute and semi-dilute polymer solution. The current setup consists of a closed channel where in addition to the target viscoelastic fluid, a second Newtonian fluid of comparable viscosity boosts the deformation of the interface due to a smaller mismatch in density. Pumped at the same velocity in the laminar regime, this second fluid allows to reach high shear rates while assuring a smooth and stratified flow. In this paper, we introduce the theoretical analysis of extracting N<sub>2</sub> directly from the bulging profile of the interface between the two fluids. Furthermore, we address the challenges of experimentally measuring the interface position in the closed pipe with optical sensors. The N<sub>2</sub> analysis is discussed in detail, and a comparison of results to existing techniques (partitioned-plate and plate-plate/cone-plate) is presented for model dilute and semi-dilute polymer solutions.

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#### Wednesday 5:00 Exhibition Hall / Poster Session

#### **In-line RheoSANS at ISIS Target Station 2 – recent results and developments** James Doutch

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The small-angle neutron scattering instruments at the second target station of ISIS Neutron source (UK) are well optimised for conducting in-line, simultaneous rheo-SANS measurements on a wide variety of systems, due to the time of flight mode of operation and low instrumental background. Here, we will review a variety of recent, published in-line rheological measurements, spanning a number of scientific fields, ranging from physical chemistry through to biophysics. These include measurements obtained with commercially available Couette geometries, and custom flow fields available through user designed pieces of sample environment equipment. We will also reflect on future capabilities in this area, and tie this to planned instrument upgrades.

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