



The British Society of Rheology



# Mid-Winter Meeting Programme

16 – 18<sup>th</sup> December 2024

Straining to Reach Beyond Steady Simple Shear -Advanced Techniques for Complex Fluid and Flow Field Characterisation

#### <u>Preface</u>

This year's Mid-Winter Meeting is jointly organised by Richard Hodgkinson of the BSR with support from the NNFM (Non-Newtonian Fluid Mechanics) Special Interest Group, co-led by Rob Poole and Alexander Morozov. With a total of 71 registered inperson attendees, a large proportion of which being specially subsidised rate student/retired BSR members, this 2½ day extended Mid-Winter Meeting at Sheffield promises to be one of the largest in-person Mid-Winter Meetings in recent years.

On the theme of "Straining to reach beyond steady simple shear - advanced techniques for complex fluid and flow field characterisation", the organisers hope that this event will both showcase the cutting edges of the field of rheometry as well as introduce some attendees to the wider nuances and complexities of the field. Our goal is to stimulate discussion on flows that depart from the canonical "steady simple shear" towards more complex flows: highlighting their importance in real world applications, the need for characterisation of materials under such complex flows in the laboratory, and the development and testing of rheological models for and under such flows.

We are grateful for additional funding to support an international plenary speaker courtesy of the EPSRC NFFDy (National Fellowships in Fluid Dynamics) hub. We are additionally grateful to TA Instruments and Anton Paar who have sponsored our morning coffee session pastries; the Journal of Non-Newtonian Fluid Mechanics for sponsoring our poster prize; the School of Chemical, Materials and Biological Engineering here at Sheffield for free use of their Ideas Space and support from Gemma Daniel for local coordination; and PJ Taste, our caterers, for arranging a bespoke package deal for this event.

MWM 2024 organisers

For abstracts, please see the online version of this programme:

https://www.bsr.org.uk/events/ bsr-2024-mid-winter-meeting



### Monday 16<sup>th</sup> December

#### Preliminary theme: "Strain rate measures, flow type classifiers and flow type models"

All events take place in the Ideas Space unless otherwise indicated.

12:00	Registration and lunch opens at 12:00		
13:00	Opening remarks		
Session chair: Richard Hodgkinson			
13:10	Roney Thompson (Universidade Federal do Rio de Janeiro) (presenting		
	remotely)*:		
	The Concept of the Persistence of a Dynamic Quantity and its Relation to		
	Flow Classification		
13:45	Paulo Oliveira (Universidade da Beira Interior)*:		
	Can the triple decomposition method be useful for non-Newtonian fluid		
	mechanics?		
14:15	Christopher Keylock (Loughborough University)*:		
	Schur-based Decomposition of the Velocity Gradient Tensor: Current		
	Approaches for (Incompressible) Turbulence		
14:45	Robert Poole (The University of Liverpool):		
	Predictions of the Generalized Newtonian Fluid model incorporating Flow		
	Type (GNFFTy) in simple and complex flows		
15:10	Coffee break		
Session	chair: Maria Charalambides		
15:35	Alice Woodbridge (The University of Manchester):		
	Spreading dynamics of vibrated yield-stress fluid drops		
16:00	BSR MWM 2024 Annual Award talk		
	Ian Frigaard (University of British Columbia (UBC))*:		
16 50	Simple yield stress fluids: no strain, no pain – what do we gain?		
16:50			
-	Henry Royce Institute "Royce Discovery Centre" (Harry Brearley building)		
17:30	(Assemble in the ideas Space)		
17:00	(DC2 Sominar room just off Idage Street)		
-	(Bod Seminar room, just off ideas Space)		
19:00			

(\* denotes invited speaker)

## Tuesday 17<sup>th</sup> December

Preliminary theme: "Non-linear, non-steady, oscillatory and superposition flows"

All events take place in the Ideas Space unless otherwise indicated.

Session chair: Richard Hodgkinson		
08:50	BSR MWM 2024 Plenary talk	
	Randy Ewoldt (University of Illinois Urbana-Champaign)*:	
	Nonlinear emergence in oscillatory and superposition flows	
09:40	Dan Curtis (Swansea University)*:	
	Mastering Optimally Windowed Chirps: Practical Implementation for Your	
	Laboratory	
10:10	Manlio Tassieri (The University of Glasgow)*:	
	i-Rheo: One Small Step (Strain) in Rheology, One Giant Leap for Multiscale	
	Viscoelastic Characterisation	
10:40	Coffee break – pastries sponsored by Anton Paar	
Session	chair: Helen Wilson	
11:05	Pete Laity (The University Of Sheffield):	
	LAOS and the emergence of non-linear flow behaviour in polymeric	
	systems	
11:30	Claudio Fonte (The University of Manchester)*:	
	A comparison between the FENE-P and sPTT constitutive models in large-	
	amplitude oscillatory shear	
12:00	Rishav Agrawal (The University of Liverpool):	
	Features and limitations of recent elastoviscoplastic constitutive models	
	under Large Amplitude Oscillatory Shear (LAOS)	
12:25	Andrew Clarke (Schlumberger Cambridge Research):	
	Parallel superposition and the relaxation time of entangled HPAM	
	solutions in flow	
12:50	Lunch with posters	
14:00	Group photo	
Session	chair: Maria Charalambides	
14:05	BSR MWM 2024 Vernon Harrison Award talk	
	Matthew Smith (The University of Glasgow)*:	
	Broadband computational rheology for material characterisation	
14:50	Chris Ness (The University of Edinburgh):	
	Filled Colloidal Gel Rheology: Strengthening, Softening, and Tuneability	
15:15	Anders Aufderhorst-Roberts (The University of Liverpool):	
	An Adventure in Time and Space – New Approaches for Probing the	
	Spatiotemporal Nature of Gelation in Biopolymer Networks	
15:40	Coffee break	

Session chair: Peter Cooper		
16:05	Ahmad Boroumand (University of Leeds):	
	Scaling behaviour of the mechanics and mesoscale structure of folded	
	protein hydrogels	
16:30	Thomas Wallis (The University Of Sheffield):	
	Scaling-up the encapsulation of microorganisms within hydrogels for	
	agricultural applications	
16:55	Emily Cook (University College London (UCL)):	
	A guide for interpreting simple thixotropic models	
17:20	Stephen Wilson (University of Bath):	
	Flow manipulation of a nematic liquid crystal in a Hele-Shaw cell with an	
	electrically controlled viscous obstruction	
18:30	Evening meal	
-	Piccolino Sheffield	
21:30	(4 Millennium Sq, Sheffield City Centre, Sheffield S1 2JJ)	

(\* denotes invited speaker)

#### Wednesday 18<sup>th</sup> December

#### Preliminary theme: "Extensional flows"

All events take place in the Ideas Space unless otherwise indicated.

Session chair: Randy Ewoldt		
08:50	Simon Haward (Okinawa Institute of Science and Technology (OIST))*:	
	Large Amplitude Oscillatory Extension of Polymer Solutions	
09:25	Phil Threlfall-Holmes (TH Collaborative Innovation)*:	
	Preliminary Results from ROJER-X	
09:55	Henry Ng (The University of Liverpool)*:	
	Highlighting the need for high-speed imaging in capillary breakup	
	extensional rheometry measurements	
10:25	Thomas Abadie (University of Birmingham)*:	
	Fluid dynamics of viscoelastic jets and fluid dripping onto a substrate	
10:55	Coffee break – pastries sponsored by TA Instruments	
Session chair: Esther Garcia-Tunon Blanca		
11:20	Richard Hodgkinson (The University Of Sheffield):	
	Towards understanding shear rheology under kinematically mixed flows	
11:45	Calum Mallorie (The University of Edinburgh):	
	Mechanistic investigation of white blood cell dynamics in cross-slot	
	extensional vortices for early disease detection	
12:10	Joerg Laeuger (Anton Paar Germany):	
	Advanced extensional rheometry on a rotational rheometer platform	

12:35	Lunch	
Session chair: Jesse Taylor-West		
13:25	Helen Wilson (University College London (UCL))*:	
	Probing the rheology of a suspension of flat elastic particles through	
	simulations	
13:55	Olivia Pickup (University of Leeds):	
	Yielding mechanisms in Mg(OH) <sub>2</sub> suspensions with spherical nanoparticle	
	additives	
14:20	Mahdi Davoodi (Schlumberger Cambridge Research):	
	Role of Elasticity and Inertia in Particle Migration: A Theoretical and	
	Experimental Study in Complex Taylor Vortices	
14:45	Coffee break	
Session	chair: Richard Hodgkinson and Robert Poole	
15:10	James Richards (The University of Edinburgh):	
	Mixing concentrated suspensions: Anomalous particle transfer between	
	immiscible liquids	
15:35	Oliver Harlen (University of Leeds)*:	
	Modelling Polymer Flow Induced Crystallisation in Complex Flows	
16:05	Poster prize presentation and closing remarks	
16:15	Planned meeting close at 16:15	

(\* denotes invited speaker)

#### **Useful information**

#### Conference venue, dinner, and area map

The conference venue is the "Ideas Space" in the Sir Robert Hadfield building of the Faculty of Engineering at the University of Sheffield. Access is from Portobello street (as opposed to Mappin street which may be indicated on online maps). If driving, the "Q-Park" parking is immediately opposite (watch out for one way streets).



Note: this room can be slightly chilly, and we recommend e.g. jumpers.

Signs will be visible on railings near the venue. Enter through the gate shown above, following the signs, and make a right into the building. Proceed to your left once through the two sets of double doors to reach the Ideas Space.

The conference venue and Leopold hotel are marked on the map below, and are both easily reachable on the Sheffield supertram from Sheffield station via the West street and City Hall tram stops, respectively, with the blue route towards Malin Bridge being the simplest. The (multiple) tram routes are marked by the red line on the map. Buy tickets on-board from the conductor, cash or card.

Walking from the station may only take as long as waiting for and taking a tram (circa 25mins/1 mile). For taxis, use Uber or Veezu Sheffield (0114 239 3939).

An indicative walking route to our evening meal venue, Piccolino, just off the "Peace Gardens" (as shown in the cover photo) is suggested (15min/0.6mile).



We recommend this route as shortly after leaving the Ideas Space there is a designated crossing point over West Street towards Devonshire street. Parts of Devonshire street are pedestrianised and it currently features festive illuminations. You will pass our city hall and war memorial. As you approach the Peace Gardens you will notice our Sheffield Christmas market (shown overleaf) is in full swing currently, complete with an Alpine bar and illuminated "Sheffield" sign (photo opportunity!). There are also various pubs and bars along the general area of the route indicated. The "Winter Garden" (right photo) is also open currently (8am – 8pm) just off the Peace Gardens. Piccolino's entrance is situated opposite one of the Winter Garden entrances (shown overleaf).





#### Housekeeping

There are no fire alarm tests planned during the Mid-Winter Meeting. Please assume any alarm is real. In the event of the alarm sounding, please proceed out the way you entered, using an exit door to the right of a fire curtain that will descend. The assembly point is St George's churchyard, up the hill in front of the Sir Fredrick Mappin building – follow the conference organisation team and fire marshals.

If you discover a fire, please operate a fire call point, call 0114 222 4444 (not 999) stating the nature of the fire, and leave by the nearest available exit, closing any doors and windows. Do not use lifts.

For first aid, please seek the assistance of the local conference organisers (Richard Hodgkinson or Gemma Daniels) or the assistance of the porters in the porter's lodge which you passed on your way in.

For emergency calls use 0114 222 4444 (not 999) as these go through our central team. For non-emergency calls please call 0114 222 4085 (first aid, access etc).

For internet access, use the "eduroam" or "WifiGuest" networks.

#### Suggestions for Monday evening meal:

There are an array of pubs and restaurants in the general area, many more than apparent with a quick google maps search. Just a few suggestions include:

- Red Deer (Pitt street) a traditional UK pub and a favourite of academics in the Faculty of Engineering, very close to the Ideas Space
- Efes (West street) Turkish restaurant
- Frog and Parrot (Devonshire street), The Museum (Orchard Square) two further traditional pubs serving food
- The Botanist (Leopold square) Cocktail bar and restaurant
- Akbar (Fitzwilliam street) Indian restaurant

# Abstracts

Abstracts are arranged in presentation order for oral talks, and alphabetically by surname of the presenter for posters.

Oral talks, Monday 16<sup>th</sup>: page 10 - 12

Oral talks, Tuesday 17<sup>th</sup>: page 13 - 19

Oral talks, Wednesday 18<sup>th</sup>: page 20 - 25

Posters: page 26 - 32

#### The Concept of the Persistence of a Dynamic Quantity and its Relation to Flow Classification

Roney Leon Thompson, COPPE - Federal University of Rio de Janeiro – Brazil

Flow classification is an open problem. Classical flow classifications examine aspects like laminar × turbulent, steady-state × transient, incompressible × compressible. Here, we discuss flow type. A motivation for this study is the observation that most non-Newtonian fluids behave differently when subjected to a viscometric flow or an extension flow. Although these classical motions are well-defined, it is hard to determine the flow type when it is neither viscometric nor extensional. In addition, extensional flows are a broader class than viscometric flows since the latter has a single set of material functions, while the former exhibits an infinite number of sets of material functions. We discuss the challenges for comparisons between viscometric and extensional material functions. Even in the Newtonian context, flow classification is fundamental due to its connections with other physical phenomena like heat transfer, flow mixing, chemical reactions, etc. Some studies associated with the definition of a vortex can also bring insights into the current subject of analysis. In this regard, a key concept is the persistence of straining. We also explore dynamic flow classifications by considering the persistence of quantities like the conformation tensor or the stress tensor. From the perspective of yield stress materials, we notice that load type needs to replace flow type in order to address pre-yielding conditions. Emphasis is given to the decomposition of a symmetric tensor with respect to a second where one part of the first is coaxial while the other part is orthogonal to this second tensor.

#### Can the triple decomposition method be useful for non-Newtonian fluid mechanics?

Paulo J. Oliveira CMAST, UBI - Universidade da Beira Interior Portugal

The triple decomposition method was devised by Kolar (2007) and consists in the separation of the velocity gradient tensor into shear, elongational, and rotational components. It thus differs from the classical decomposition of the velocity gradient into a symmetric rate-of-strain part and an anti-symmetric rate-of-rotation or vorticity tensor part. Some initial criticisms, that the vorticity tensor cannot be used for the formulation of "constitutive equations", are easily surpassed by using a "relative" vorticity tensor thus giving a decomposition invariant with respect to rotations of the observer. In 2D the method is so straightforward that it may be used inside a flow algorithm; in 3D that is not so at present (although the Schur form might make it viable) but it may still be routinely applied as a post-processing technique. We show that Kolar's method is indeed very useful for non-Newtonian flow analysis, especially for problems involving mixed kinematics, since it allows one to easily obtain the true local rates of elongation, shearing and effective rotation (discounting rotation associated with shear). With such method we can: 1- Obtain those rates while following a fluid element along a path line; 2-Evaluate the true "flow type" locally which may facilitate flow assessment; 3- Incorporate into a flow code and look, for example, for the effects of a strain-hardening elongational viscosity in a flow with mixed kinematics. Kolar, 2007, Int. J. Heat Fluid Flow, 28, 638-652

#### Schur-based Decomposition of the Velocity Gradient Tensor: Current Approaches for (Incompressible) Turbulence

Chris Keylock, Loughborough University

Since about 2018 there has been an upsurge in using the Schur decomposition as an alternative to the Eigenvalue decomposition for studying the velocity gradient tensor in (incompressible) turbulent flows. This has certain advantages both in terms of isolating coherent flow structures and in terms of examining the effects of the pressure and viscous terms on the velocity gradients. The Schur form may be real or complex, with the latter easier to work with mathematically but the former of more obvious physical application. This paper will review some of the recent work in this area, with a view to considering the potential of this approach for non-Newtonian flows.

#### Predictions of the Generalized Newtonian Fluid model incorporating Flow Type (GNFFTy) in simple and complex flows

R J Poole, University of Liverpool

A two-dimensional steady and homogeneous flowfield in which the flow-type can be continuously varied from solid-body rotation to simple shear to planar extension was probed for various viscoelastic models including the upper convected Maxwell, the simplified Phan-Thien and Tanner (sPTT) and the FENE-P. By defining a frame-invariant coordinate system, 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. A functional form of a viscosity equation which incorporates flow-type, but is otherwise inelastic, the so-called GNFFTy (Generalized Newtonian Fluid model incorporating Flow Type pronounced "nifty"), is proposed. We also test this model in a complex two-dimensional flow, of a 4:1 contraction under inertialess conditions and compare to benchmark data and experimental results.

#### Spreading dynamics of vibrated yield-stress fluid drops

Alice Woodbridge<sup>(1)</sup>, Anne Juel<sup>(1)</sup>, Claudio Fonte<sup>(1)</sup> (1) The University of Manchester

Yield-stress fluids are ubiquitous in daily life; however, our understanding of their behaviour remains limited, particularly at and below the yield point. In this work, we use experiments and simulations to investigate the effects of rheology and geometry on the spreading dynamics of a vibrated drop of yield-stress fluid. In experiments, we employed fluids with a range of microstructures (a microgel, emulsion, and suspension) to examine how material microstructure influences spreading dynamics. To study the effect of drop geometry, we developed three distinct deposition methods creating different drop shapes with unique shearing histories. In simulations we varied the Bingham and Weissenberg numbers as well as the initial drop shape to complement the experiments. When vertical vibrations are applied to a yield-stress drop, two distinct regimes are observed. Initially, there is a 'creep' region where the drop exhibits <10% reduction in height. Beyond a critical acceleration, proportional to the yield stress, the drop yields and spreads significantly with increasing acceleration. We propose two scaling arguments which show that both the extent of creep and the critical acceleration are independent of the material

microstructure but depend on the drop height, as well as elasticity and yield stress, respectively. Furthermore, simulations revealed the physical mechanisms governing the spreading dynamics and allowed us to explore the effect of parameters beyond the experimental range.

#### Simple yield stress fluids: no strain, no pain – what do we gain?

Ian Frigaard, University of British Columbia

Yield stress fluids abound: from food products through polymer gels to industrial and natural slurries. The key mechanical feature of such fluids is a threshold stress that must be exceeded in order for the fluids to deform and flow. The most simple models of these materials (Bingham and Herschel-Bulkley fluids), are a century old and in wide usage. Here we first explore some of the theoretical challenges that arise in studying typical flows of these fluids, i.e. as a result of the yield stress. These range from unique and interesting flow features through to effects on the hydrodynamic stability of flows. In the second part of the talk we move from theory to application. These are of two types. (i) New and novel flows that can be achieved because of the yield stress; examples are given. (ii) Industrial processes where the yield stress has a fundamental effect on the flow. For the latter we outline two long standing research areas of relevance to greenhouse gas emissions, where surprisingly the yield stress can either help or hinder.

#### Nonlinear emergence in oscillatory and superposition flows

Randy Ewoldt, University of Illinois Urbana-Champaign

The Pipkin Map is a foundational tool to understand how rheological complexity emerges from both timescale and forcing strength effects. It has the potential to help us organize the landscape beyond steady simple shear, but its typical embodiment has several major limitations. This talk will extend the premise of the Pipkin Map into a larger paradigm to understand complex flows involving shear, extension, and superposed flows where the forcing may be characterized by strain, strain rate, or stress. The dimensionless forcing strength may be represented with the Weissenberg number, Plasticity number, Capillary number, Mnemosyne number, or inverse Bingham number - a unifying concept of a dimensionless forcing strength will be described, for which the above concepts are but subsets associated with specific microstructural physics. The dimensionless forcing strength causes the emergence of nonlinearity. This edge of linearity now has a rigorous definition independent of the subjective threshold of deviation, which is most clearly revealed from oscillatory deformation known as medium-amplitude oscillatory shear (MAOS) that independently controls the forcing timescale and amplitude. Recent work with stresscontrolled MAOS has revealed that stress is a more fundamental measure of nonlinearity strength across a wide range of Deborah number, challenging prior perspectives which have generally fixated on using strain or strain rate.

#### Mastering Optimally Windowed Chirps: Practical Implementation for Your Laboratory

R. E. Hudson<sup>(1)</sup>, M. Das<sup>(2)</sup>, G. H. McKinley<sup>(2)</sup>, D. J. Curtis<sup>(1)</sup> (1) Swansea University, (2) Massachusetts Institute of Technology

Whilst Linear Viscoelastic Characterisation of complex fluids and soft solids using Optimally Windowed Chirp (OWCh) rheometry is gaining popularity, the technique is yet to be implemented as a standard test procedure in the software that runs rheometric devices. In this talk, we will demonstrate the use of SUMIT-OWCh, a freely available standalone executable that can be installed alongside the rheometer software to support the implementation and analysis of OWCh experiments. Focussing on Combined Motor Transducer systems operating in stress-controlled mode, we will present guidelines for waveform design and analysis that mitigate issues associated with instrument inertia, strain offset, and waveform sampling before the use of SUMIT is demonstrated using a range of stable and mutating materials for two widely available rheometers.

#### i-Rheo: One Small Step (Strain) in Rheology, One Giant Leap for Multiscale Viscoelastic Characterisation

#### Manlio Tassieri, University of Glasgow

The linear viscoelastic (LVE) properties of materials, expressed through the complex shear modulus, are key to understanding their structural dynamics across scales. Traditionally, these properties are measured via oscillatory shear experiments, which, while effective, are often time-consuming and limited in frequency range. Recent advances in rheology have addressed these limitations with innovations like i-Rheo for bulk rheology [J. Rheol., 60, 649 (2016)], i-Rheo-GT for molecular simulations [Macromolecules, 51, 5055 (2018)], microrheology using optical tweezers

(MOT) [New J. Phys., 14, 115032 (2012)], and atomic force microscopy (AFM2) [Sci. Rep., 8, 14462 (2018)]. These techniques analyze time-domain data without predefined models, utilizing inverse Fourier transforms to deliver broad-spectrum viscoelastic data. New tools have also expanded rheological characterization in biological systems. OptoRheo integrates light-sheet microscopy with particle-tracking microrheology to study cellular interactions in the extracellular matrix [Commun. Biol., 6, 463 (2023)]. The i-Rheo-optical assay enables viscoelastic analysis of multicellular spheroids, helping distinguish healthy from diseased tissues [Mater. Today Bio, 26, 101066 (2024)]. Optical Halo, using a Bessel beam from optical tweezers, extends microrheology into low-frequency ranges [Micromachines, 15, 889 (2024)]. These innovations open new frontiers in material and biological tissue analysis.

#### LAOS and the emergence of non-linear flow behaviour in polymeric systems

Peter Laity<sup>(1)</sup>, Chris Holland<sup>(1)</sup> (1) The University of Sheffield

The rheology of polymer systems attracts considerable attention, for several reasons. The complexity of the flow behaviour and its interpretation in terms of mechanisms at the molecular scale has been the subject of much academic work. At the same time, reliable descriptions of flow behaviour are of great importance in practical situations, including the characteristics of foodstuffs, healthcare products and the industrial processing of polymeric materials. Two key issues are stress overshoot during the start-up of steady flow and the emergence of non-linear behaviour during oscillatory characterisation. Conventionally, the latter has been evaluated by fitting Fourier series to the experimental data. Whilst providing a good fit, however, questions remain regarding the physical meanings of the second and subsequent terms in the Fourier series. The present work considers a different mathematical function which, in spite of obvious limitations, may provide a clearer interpretation. Hence, the present work examines stress overshoot and non-linear LAOS behaviour for an exemplar polymer system and suggests a link between them.

#### A comparison between the FENE-P and sPTT constitutive models in large-amplitude oscillatory shear

T.P. John<sup>(1)</sup>, R.J. Poole<sup>(2)</sup>, A.J. Kowalski<sup>(3)</sup>, C.P. Fonte<sup>(1)</sup> (1) The University of Manchester, (2) The University of Liverpool, (3) Port Sunlight Laboratory, Unilever R&D

The simplified Phan-Thien-Tanner (sPTT) and FENE-P models are widely used viscoelastic constitutive models for a wide range of materials. Originating from distinct microstructural theories, they become mathematically identical in steady, homogeneous flows. Consequently, fitting either model to experimental data from steady simple shear flows (SSSF) does not determine which is most appropriate for a given material. In complex flows, however, even if Eulerian steady, the two models' behaviour can diverge significantly due to Lagrangian transients, highlighting the importance of understanding their rheological differences in unsteady flows. Large Amplitude Oscillatory Shear (LAOS) rheometry provides an ideal framework for such investigations. Here, we numerically investigate the sPTT and FENE-P models under LAOS across a range of Deborah (De) and Weissenberg (Wi) numbers. We find that the FENE-P model exhibits 14

strong stress overshoots, unlike the sPTT model., which is explained using the conformation tensor evolution equations. Additionally, the sPTT model's LAOS response scales with Wi  $\epsilon$ ^0.5, where  $\epsilon$  is the extensibility parameter. However, the FENE-P LAOS response only scales with its SSSF scaling parameter for large enough values of the extensibility parameter L^2. Using the Sequence of Physical Processes framework (SPP), we show that FENE-type models exhibit viscous backflow, characterised by a negative instantaneous viscous modulus preceding stress overshoots.

#### Features and limitations of recent elastoviscoplastic constitutive models under Large Amplitude Oscillatory Shear (LAOS)

Rishav Agrawal<sup>(1)</sup>, Esther García-Tuñón<sup>(1)</sup>, Robert J Poole<sup>(1)</sup>, Claudio P Fonte<sup>(2)</sup> (1) University of Liverpool, (2) University of Manchester

Elastoviscoplastic (EVP) models are becoming more widely adopted to investigate the deformation and flow of yield stress materials for various applications. In this work, we investigate EVP models, primarily the Saramito model [1] but also the recently developed Kamani-Donley-Rogers (KDR) model [2], under LAOS tests using Fourier Transform rheology, plastic dissipation ratio ( $\phi$ ) analysis [3] and the Sequence of Physical Processes framework [4]. A detailed parametric study has been conducted for the Saramito model for a wide range of values of the relevant non-dimensional parameters. We also compare the Saramito and KDR models for a particular set of conditions and compare them with experimental data for Pluronic F127 hydrogel, a yield stress fluid commonly used in 3D printing. The parametric study of the Saramito model reveals a universal scaling for the onset of purely elastic behaviour, which shows dependence on both Bingham and Weissenberg numbers. Moreover, we demonstrate that although both EVP models can predict the 'yielding' process, the KDR model provides a better agreement with rheological data for Pluronic F127. The KDR also performs better in predicting associated EVP behaviour than the Saramito model, e.g. gradual change in the first-harmonic moduli and the  $\phi$  values near the onset of yielding. [1] Saramito, P. Journal of Non-Newtonian Fluid Mechanics 158.1-3 (2009): 154-161. [2] Kamani, K., et al. Physical review letters 126.21 (2021): 218002. [3] Ewoldt R.H., et al. Rheologica Acta 49 (2010): 191-212 [4] Rogers S.A. Rheologica Acta 56 (2017): 501-525.

#### Parallel superposition and the relaxation time of entangled HPAM solutions in flow

Enny Tran<sup>(1)</sup>, Andrew Clarke<sup>(1)</sup> (1) SLB

It is generally understood that for entangled viscoelastic polymer solutions entanglement effects are significantly weakened under strong flow. In recent years, it has been observed that, despite quiescently being entangled at use concentrations, high molecular weight HPAM solutions are characterised by their Rouse time when transitioning to elastic turbulence in porous flow. Whereas this has been inferred, for these practically important solution polymers direct measurement of the apparent characteristic relaxation time under shear has not been reported. Here we use controlled-stress parallel-superposition background shear to assess the apparent characteristic relaxation time concide with a recently reported calculation using Rolie-poly and Rolie-double-poly models.

#### Broadband computational rheology for material characterisation

Matthew Smith<sup>(1)</sup>, Manlio Tassieri<sup>(1)</sup> (1) The University of Glasgow

This thesis advances microrheology through new computational tools, machine learning integration, and insights into complex fluids under varying shear conditions. It focuses on the viscoelastic properties of materials like polymers, tissues, and complex fluids, relevant to biophysics and material science. A key contribution is "i-RheoFT," a MATLAB tool introduced in Chapter 2. It accurately calculates the complex shear modulus from unevenly spaced, noisy experimental data using advanced interpolation. Demonstrated on hydrogels and biological samples, i-RheoFT shows promise for broader applications and refinement. Chapter 3 enhances microrheology with optical tweezers (MOT) by incorporating a machine learning model, reducing measurement times from tens of minutes to one second with an error margin of just 0.3%. This breakthrough allows real-time fluid analysis, enabling MOT to study more dynamic systems. Chapter 4 explores shear-thinning fluids, revealing that increasing shear rates lower viscosity, improving flow and particle alignment. However, prolonged exposure can disrupt internal structures, highlighting implications for optimizing drug delivery and industrial processes. Overall, this thesis provides tools and methods for understanding complex materials, with wide applications in material science, biophysics, and engineering, paving the way for future studies to refine these techniques and broaden their impact.

#### Filled Colloidal Gel Rheology: Strengthening, Softening, and Tuneability

Yujie Jiang<sup>(1)</sup>, Yang Cui<sup>(1)</sup>, Yankai Li<sup>(1)</sup>, Zhiwei Liu<sup>(1)</sup>, Ryohei Seto<sup>(1)</sup>, Christopher Ness<sup>(2)</sup> (1) Wenzhou Institute, (2) University of Edinburgh

Filler-induced strengthening is ubiquitous in material science and is particularly well-established in polymeric nanocomposites. Despite having similar constituents, colloidal gels with solid filling exhibit distinct rheology, which is poorly understood. We show using experiments and simulations that filling monotonically enhances the yield stress of colloidal gels, while the elastic modulus first increases before decreasing. The latter effect results from a disturbed gel matrix at dense filling, evidenced by a growing inter-phase stress. This structural frustration is, however, not detrimental to yielding resistance. Instead, fillers offer additional mechanical support to the gel backbone via percolating force chains, at the same time decreasing the yield strain. We develop a mechanistic picture of this phenomenology that leads us to a novel `filler-removal protocol,' making possible individual control over the strength and brittleness of a composite gel.

#### An Adventure in Time and Space – New Approaches for Probing the Spatiotemporal Nature of Gelation in Biopolymer Networks

Qandeel Saleem<sup>(1)</sup>, William Barker<sup>(1)</sup>, Timea Feller<sup>(2)</sup>, Natasha Shirshova<sup>(1)</sup>, Anders Aufderhorst-Roberts<sup>(1,3)</sup> (1) Durham University, (2) University of Leeds, (3) University of Liverpool

Many soft and biological materials undergo changes in their rheological response at boundaries and interfaces, leading to non-uniform material properties. Examples include biodegradable polymeric materials and the fibrin blood clots that prevent bleeding when we cut ourselves. Each of these encompasses a common theme – the rheological response has a temporal \*and\* spatial dependence. While rheological techniques exist to accurately probe time dependence, probing rheology in time and space remains experimentally challenging. In this presentation I will discuss a technique known as rheodialysis, which extends the capabilities of a parallel plate rheometer by incorporating a customised flow cell with a porous membrane. Rheodialysis was applied to a model system of alginate biopolymer hydrogels assembled through diffusion of calcium ions. Continuous multiple frequency oscillatory tests allowed the identification the critical gel point as a function of plate separation. This reveals a viscoelastic gradient, owing to the diffusion path of calcium ions through the incipient gel network, which is quantified through the fractal dimension and supported by reaction diffusion modelling. I will also discuss how this technique is being adapted to biological materials; specifically model blood clots constructed from fibrin fibres. We anticipate that rheodialysis will enable us to recreate artificial blood clots to accurately probe fibrin network mechanics under physiological conditions.

#### Scaling behaviour of the mechanics and mesoscale structure of folded protein hydrogels

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Folded protein hydrogels are generating a lot of current interest offering a hierarchical structure with large design space. Yet, a detailed understanding of the dependency of the mechanics and structure of such gels on the protein volume fraction ( $\varphi$ ) to map out their design principles remains unexplored. Inspired by theories from soft matter physics, we have investigated the scaling behaviour of the  $\varphi$  of such hydrogels and their relationship to the underlying structure and mechanics through a combination of rheology and small-angle neutron scattering (SANS). Using the globular protein bovine serum albumin as a model system, we have identified a two-regime behaviour for storage modulus as a function of  $\varphi$  reminiscent of the strong link and weak link regimes defined in the colloidal flocculated gels. Additionally, SANS reveals the fractal dimension (D) of clusters in hydrogels remains constant (~2.35) up to  $\varphi$ =4.4% beyond which the crowded network does not allow for further growth of mature clusters, resulting in a progressive decay in fractal dimension reaching D=2.06 at  $\varphi$ =7.4%. Further, network parameters such as number of proteins in an average cluster and cluster size scale with  $\varphi$  similar to the predictions of the blob and flocculated models. The insight gained from our integrated structural and mechanical approach will enable the design of novel biomaterials for storage and release of drug molecules.

#### Scaling-up the encapsulation of microorganisms within hydrogels for agricultural applications

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Food is a bedrock of human civilisation, which every person is reliant on. Unfortunately, there is a large disparity in food access across the world meaning up to 783 million people despite the UN sustainable development goal of eliminating world hunger by 2030. Resource constraints on the manufacturing of agrochemicals has driven the pursuit of alternative crop products, namely biologicals. These products are highly scalable, exhibit extended availability in the soil, foster a more diverse biological community (improving crop productivity) whilst preserving soil structure and tend not to leach outside of their area of application. At present the transportation and application of living microbes remains challenging; hydrogel encapsulation has been proposed as a method for protecting the living payload. This project aims to develop a scalable method of encapsulating soil microbes in micron-sized hydrogel beads towards delivering a more sustainable agricultural product. A range of biopolymers, including chitosan, alginate and cellulose are crosslinked with a range of crosslinking agents to form biodegradable gels. The mechanical properties of the gels and the kinetics of crosslinking are measured by rheology. In parallel, various microbes are cultured to form a broth. The broth is then loaded with a crosslinkable biopolymer and controllably crosslinked to measure viability for different biopolymers and different crosslinking densities. The loaded biopolymer precursor is then sprayed into the crosslinking agent to form microparticles. The shear stress required causes a tradeoff between particle size and viability of the microbe. Identifying and tuning the operational parameters on a pilot scale rig enables fast screening for different microbe/hydrogel/crosslinker combinations. This project develops an inexpensive, scalable product that supports sustainable farming and economic growth.

#### A guide for interpreting simple thixotropic models

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Thixotropic materials exhibit a viscosity that evolves over time and in response to forcing. Broadly speaking, thixotropy is a phenomenological characterisation that captures, at a continuum scale, the transient evolution of internal micro-scale structure in a fluid. This can manifest in different ways: while some thixotropic fluids only display transient shear thinning, other materials exhibit yield stresses, yield stress ageing, and viscosity bifurcations, in addition to phenomena such as viscoelasticity. There is an overwhelming array of thixotropic models proposed in the literature, ranging in construction and application, many of which are coupled with other non-Newtonian behaviours. Such models provide a powerful tool to capture complex and realistic physics, but they can be difficult to interpret and unpack, especially when the underlying thixotropic construction is not fully understood. This talk will discuss ideally thixotropic models, and establish a systematic approach for choosing and understanding the right thixotropic rheology. We will discuss model requirements for capturing the thixotropic behaviours mentioned above, and outline two approaches to capture thixotropic yielding. As we will see, the sorts of behaviours predicted by these models are often not immediately obvious from their construction: seemingly simple models can generate surprisingly complex behaviour, and models that seem almost equivalent can predict qualitatively different rheological phenomena.

# Flow manipulation of a nematic liquid crystal in a Hele-Shaw cell with an electrically controlled viscous obstruction

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The flow of a nematic liquid crystal in a Hele-Shaw cell with an electrically controlled viscous obstruction is investigated using both a theoretical model and physical experiments. The viscous obstruction is created by temporarily electrically altering the viscosity of the nematic in a region of the cell across which an electric field is applied. The theoretical model is validated experimentally for a circular cylindrical obstruction, demonstrating user-controlled flow manipulation of an anisotropic liquid within a heterogeneous single-phase microfluidic device.

#### Large Amplitude Oscillatory Extension of Polymer Solutions

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In practical applications, complex fluids such as polymer melts and solutions, biological fluids, and food products often undergo repeated large and rapid deformations involving mixed shear and extensional kinematics. Large Amplitude Oscillatory Shear (LAOS) tests are pivotal in providing rheological fingerprints that detail the behavior of such soft materials under nonlinear shear deformations. However, the use oscillatory flows for elucidating the extensional properties of complex fluids remains largely unexplored. We introduce a new experimental method to examine the fluid response to Large Amplitude Oscillatory Extension (LAOE). We employ high-precision programmable syringe pumps to drive a sinusoidal flow through the microfluidic Optimized Shape Cross-slot Extensional Rheometer (OSCER) device that generates an almost homogeneous planar extension. We analyze the time-dependent flow field inside the OSCER by means of  $\mu$ -particle image velocimetry and measure the simultaneous pressure drop to evaluate the elastic stress response of the fluid. We examine the flow of viscoelastic dilute polymeric solutions during LAOE, covering a broad range of Weissenberg and Deborah numbers. This investigation advances our understanding of non-Newtonian fluid dynamics and extensional rheometry, and leads to a potentially promising new methodology for characterizing complex fluids under extremely nonlinear flow conditions.

#### Preliminary Results from ROJER-X

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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.

# Highlighting the need for high-speed imaging in capillary breakup extensional rheometry measurements

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The diameter evolution of a liquid bridge undergoing capillary thinning and breakup in a uniaxial extensional flow can be analysed to extract material properties of complex fluids. The capillary breakup extensional rheometer (CaBER) is a commonly used instrument for this purpose and in the case of the commercial HAAKE<sup>™</sup> CaBER1<sup>™</sup> (sold by Thermo Scientific<sup>™</sup>) the filament diameter evolution is tracked at the mid-plane between the two end-plates using a laser micrometer. We show using high-speed imaging that while this arrangement is satisfactory in flows where the filament is long, slender and approximately cylindrical, errors can be significant when the filaments are short and curved such as encountered when using the so-called slow-retraction-method and 'Dripping-onto-Substrate' rheometry. We will further highlight the need for high-speed imaging in CaBER experiments by presenting a case study where the laser micrometer is misaligned with the location of filament breakup such as could easily be encountered for many so-called 'yield-stress' fluids where the location of filament breakup is not typically known a priori.

#### Fluid dynamics of viscoelastic jets and fluid dripping onto a substrate

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Spraying and jetting of formulated products, which involve strong elongational flow, are encountered in many domestic, industrial and medical applications. The fluid viscoelasticity and its relaxation time play an important role in the flow topology and it is therefore crucial to measure accurately extensional properties. The balance between inertial, capillary and elastic forces are investigated through numerical simulations in two configurations: (i) dripping-on-substrate (DoS), which is a conceptually simple extensional rheometer as it consists in measuring the thinning rate of a single drop dispensed onto a solid substrate; (ii) a viscoelastic jet, where a complex fluid is impulsively injected through a nozzle and the thinning rate of multiple threads are followed. Axisymmetric numerical simulations are performed with the Basilisk finite volume solver, using a Volume of Fluid representation of the interface, a FENE-P constitutive equation to describe the fluid viscoelasticity and the log-conformation method to capture the evolution of the conformation tensor. The role of the finite extensibility of the fluid on the thinning dynamics in both cases is investigated together with the influence of perturbative effects - wettability and gravity in DoS; inertia for the pulsed jets-. A simple 1D model that captures the initial rate of thinning, as well as the structure of the transition region to the nonlinear elastocapillary regime is also compared to DoS simulations.

#### Towards understanding shear rheology under kinematically mixed flows

Richard Hodgkinson<sup>(1)</sup>, Steven Reynolds<sup>(1)</sup> (1) The University of Sheffield

This talk details the background of and progress on the "SECRET" fellowship project (shear extension combined rheology experimental techniques) towards the measurement of shear rheology under "kinematically mixed" - combined shearing and extension - flows for a variety of test systems. It is well known that extensional flows align particles and molecules, and in the case of polymers, can significantly extend them, and we know from the field of liquid crystals that even orientation affects viscosity. It was shown previously using particle image velocimetry (PIV) and a two phase stratified flow experiment that extensional flow does indeed affect shear viscosity[1], however optical PIV places specific restrictions on the nature of the fluid under test. Here, data showing the feasibility of magnetic resonance imaging (MRI) towards performing measurements of shear viscosity under extensional flow using the two phase flow technique previously developed is presented. Discussion highlights some of the nuances of MRI flow imaging and work underway to develop imaging strategies and address artefacts and distortions. This talk also connects to the first and second sessions of this Mid-Winter Meeting, prompting the question "what is the correct measure of shear and extension rate in kinematically mixed experimental flow fields?", and in that the shear experienced by the fluid is highly time transient, presenting a challenge for control shear experiments. [1] Hodgkinson, R., et al., Journal of Rheology (2022) 66 (4), 79.

# Mechanistic investigation of white blood cell dynamics in cross-slot extensional vortices for early disease detection

Calum Mallorie<sup>(1)</sup>, Timm Krüger<sup>(1)</sup> (1) The University of Edinburgh

Early detection of diseases can improve patient survival and recovery, but is especially challenging for fast-progressing conditions such as sepsis. By eliminating the need for slow methods like cell culturing or DNA amplification, detecting pathological changes in white blood cell mechanical properties can reduce diagnosis times. Microfluidic cross-slot devices can quickly probe the mechanical properties of large cell populations by applying high stresses through an extensional vortex, then measuring how the deforming cells transiently interact with the fluid flow. However, the mechanisms by which the mechanical properties of individual white blood cells influence their behaviour in this flow are not well understood, limiting the development of improved diagnostic devices. Here, we use viscoelastic capsules as simplified models of white blood cells and employ coupled lattice-Boltzmann finite element simulations to show that capsule deformation disrupts the vortex core, altering the forces acting on the capsules. This change in forces leads to experimentally measurable changes in capsule trajectories that reflect their mechanical properties. This new mechanistic understanding will aid experimental cross-slot users, allowing them to incorporate physical insights into their design decisions and analysis, ultimately contributing to better diagnostic tools for fast-progressing diseases.

#### Advanced extensional rheometry on a rotational rheometer platform

José Alberto Rodríguez Agudo<sup>(1)</sup>, Jan Haeberle<sup>(1)</sup>, Joerg Laeuger<sup>(1)</sup> (1) Anton Paar Germany

Performing extensional measurements with a rotational rheometer has proven successful for polymer melts with counter-rotating drums geometries. We describe new techniques to extend the limits of existing devices at low extensional stresses and large Hencky strains, and show the importance of simultaneous sample visualization to ensure truly uniform extensional deformation. By using a second motor when measuring with two counter-rotating drums, torque sensitivity is significantly improved and the measuring range is extended to lower extensional rates and smaller sample sizes. By moving one of the drums in vertical direction during extensional deformation, the sample is tilted slightly and overlapping after one revolution is avoided. Compared to standard operation, where the maximum Hencky strain is limited to 4, Hencky strains of over 8 were achieved. The visualization of the sample and image analyses reveal that overshoots in the extensional viscosity of the measured polymer melts are attributable to necking. By equipping the rheometer with a lower linear drive and combining the setup with a high-speed camera, capillary breakup experiments can be carried out on liquids with low viscosity on the same rheometer platform. The use of polarized imaging, which visualizes the stress distribution of the samples during elongation by measuring the local birefringence over the entire sample area, provides further insight into the various applications discussed.

#### Probing the rheology of a suspension of flat elastic particles through simulations

Luke Debono<sup>(1)</sup>, Helen Wilson<sup>(1)</sup> (1) University College London (UCL)

Recent (2023) theoretical work by Eggers, Liverpool & Mietke provided a minimal physical construction for a particle which, in suspension, is predicted to behave as an Oldroyd-A fluid. This construction is a flat elastic particle built from three beads connected by springs and slip-links. We present numerical simulations of a suspension of three-bead particles which are a close analogy to the idealised theoretical particle, and show how the particles align in flow and what we can deduce about the rheology of the whole suspension.

#### Yielding mechanisms in Mg(OH)<sub>2</sub> suspensions with spherical nanoparticle additives

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Addition of spherical nanoparticles into a concentrated particulate suspension of magnesium hydroxide has shown to significantly reduce the yield stress. The mechanism of which is difficult to investigate due to the complex nature of the Mg(OH)<sub>2</sub> particle shape, size and surface charge. Blending SiO<sub>2</sub> nanoparticles of varying sizes (100 nm, 250 nm, 500 nm and 800 nm) at different concentrations (Mg(OH)<sub>2</sub>:SiO<sub>2</sub> – 27:3 vol%, 28:2 vol%, 29:1 vol%) caused the yielding process to transition from an abrupt, brittle yield in Mg(OH)<sub>2</sub> suspensions to a more gradual, softer yield with the SiO<sub>2</sub> particle sizes between 100 – 500 nm. The basis of this yield stress reduction is via a ball bearing mechanism of lubrication between the irregularly shaped Mg(OH)<sub>2</sub> particles and the

spherical nano-silica. This mechanism was shown to be driven by two main factors; a sufficient number of SiO<sub>2</sub> particles and sufficient dispersion of those particles throughout the suspension. Creep rheology showed multiple yielding steps with the addition of smaller SiO<sub>2</sub> particles and large amplitude oscillatory shear (LAOS) rheology showed a greater reduction in friction with the optimum concentration of small, well dispersed  $SiO_2$ . This study demonstrates the use of nanoparticles as yield stress modifiers in concentrated suspensions and investigates the physical mechanism between the two particle types, generating a greater understanding of a ball bearing type lubrication present in the system.

#### Role of Elasticity and Inertia in Particle Migration: A Theoretical and **Experimental Study in Complex Taylor Vortices**

M. Davoodi<sup>(1)</sup>, A. Clarke<sup>(1)</sup> (1) Schlumberger Cambridge Research

The flow between concentric cylinders, driven by wall motion, is widely studied in fluid mechanics due to the ability to create diverse flow conditions. These range from simple shear flows to complex steady state and to time-dependent behaviours characterised by Taylor vortices. While often considered in academic research, this geometry is also found in practical applications, such as in the oil and gas industry, where drilling fluids are used to transport cuttings along an annulus to the surface. Understanding the behaviour of these fluids, especially when they exhibit non-Newtonian and viscoelastic properties, is crucial for practical purposes; that is, improving particle transport and well-bore cleaning efficacy. In this study we aim to clarify the roles of different physical forces, particularly elasticity and inertia, in particle migration within annular flow with center-body rotation. We develop a comprehensive approach that combines theoretical analysis, numerical simulations, and experimental observations. Our Eulerian-Eulerian model solves separate momentum equations for both the fluid and particle phases, allowing us to determine distinct velocity fields for each. This approach highlights the relative motion between the particles and fluid, framing the problem as a "resistance" rather than a "mobility" issue. By examining how inertial and elastic forces influence the drag and lift on particles, we demonstrate their combined effects on particle migration. Our results reveal that inertia causes particles to move outward within the vortices, forming ring-like structures, while elasticity tends to pull them toward the vortex center. The interplay between these forces is captured effectively by our model and aligns well with experimental observations, offering new insights into suspension flow dynamics in complex conditions.

#### Mixing concentrated suspensions: Anomalous particle transfer between immiscible liquids

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Mixing a concentrated suspension with an immiscible liquid is integral to using formulated products, e.g. chocolate mixing with saliva, and processing them, such as diluting oil-dispersion agrochemicals. Interfacial adsorption of particles is usually only considered at dilute concentrations. Here particles wetted by both liquids adsorb with a stress-scale set by surface tension over particle size,  $\gtrsim$  1000 Pa, that seemingly restricts transfer between liquids. Here, we show using rheo-confocal microscopy on a glass-in-oil suspension mixed with an aqueous phase

that particles transfer at stress-scales three orders of magnitude below that predicted from the single particle picture. These microscopic measurements are supported by a quantitative model for macroscopic particle transport and the observed drop in viscosity for rheological measurements while immersed in an immiscible liquid. This necessitates a multi-particle picture for interfaces with concentrated suspensions that will control the structures formed in mixing from energy materials to eating chocolate.

#### Modelling Polymer Flow Induced Crystallisation in Complex Flows

William Grant<sup>(1)</sup>, Oliver Harlen<sup>(1)</sup>, Daniel Read<sup>(1)</sup> (1) University of Leeds

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. In a complex geometry, such as flow through a constriction, the different strain histories experienced by the polymer chains produce highly localised regions of crystallisation. In this work we combine the Rolie Double Poly (RDP) [1] constitutive model with the polySTRAND [2] model for predicting the flow enhanced nucleation rate of a polydisperse blend of linear polymers, implemented using rheoTool [3] platform within openFOAM. This allows us to study flow induced nucleation and growth within the contraction-expansion geometry [4]. This geometry contains regions of both shear and extensional flow. However, we find that it is the high shear region near the wall of the constriction that is responsible for the crystal formation, which is localised to narrow "fang-like" structures, as seen in experiments [5]. We also show that by holding the wall at different temperature from the melt we can vary the position in the flow at which crystallisation occurs. [1] V.A. Boudara, et al., J. Rheol, 63, 71 (2019). [2] D.J. Read, et al., Physical Review Letters, 124, 147802 (2020). [3] F. Pimenta and M. A. Alves, J. Non-Newt. Fluid Mechanics, 239, 85 (2017). [4] L. Scelsi, et al., J. Rheol, 53, 859 (2009).

#### Parallel Superposition Rheometry of Yield Stress Materials

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The elasto-visco-plastic behavior of Yield Stress Materials (YSMs) has received significant interest in recent years with measurements in different flows and a range of models proposed. Here, two of the most promising models, Saramito1 and KDR2, are compared with experimentally obtained data for the case of sub-yielding parallel superposition of steady and oscillatory shear for a variety of YSMs (Carbopol and Pluronic microgels, and a commercially available emulsion). The deformation amplitude and rate of the material was measured in response to a stress-controlled sinusoidal signal with a constant offset. As the maximum applied stress (i.e., the sum of steady and oscillatory terms) has been kept well below the yielding range, the long-time response was observed to be a bounded periodic oscillation with an offset, as expected for a stress insufficient for yielding. The analytical solutions of the Saramito model, with fit parameters from rheological flow curves and SAOS measurements, capture this behavior qualitatively, whereas the predictions of KDR model, obtained numerically, exhibit an unbounded deformation growth that is not observed experimentally for the tested fluid systems, even for inputs well below yield stress, i.e.,  $\tau(t)max = O(0.1 \tau y)$ .

#### Chemorheology of vitrimer thermosetting system

Rebecca Cass<sup>(1)</sup>, Matthew Unthank<sup>(1)</sup> (1) Northumbria University

Materials with reversible covalent bonds are called 'vitrimers'. Vitrimers are capable of being reshaped or dissolved under specific conditions, due to an exchange crosslinking bonds, while otherwise behaving as a thermoset. A new polymeric material has been developed which undertakes two different crosslinking mechanisms; with the mechanism followed being determined by the temperature the material is cured at. The first mechanism forms reversible covalent bonds to produce a vitrimer, while the second forms permanent covalent bonds, creating a thermoset material. The complete isothermal curing of this material was studied by chemorheological analysis, across a range of temperatures. Below a threshold temperature, only one point of structural evolution is evident, indicated by a point at which the tan  $\delta$  is independent of frequency, whereas above this temperature there are two points of structural evolution. Analysis of the material structure, after both transitions has been completed, with the crosslinking density calculated for both transitions.

#### The Icing on the Doughnut: Using Rheology to Deconstruct Icing for Improvement of Low-Fat, Low Sugar Alternatives

Peter Cooper<sup>(1)</sup>, Anthony Fletcher<sup>(2)</sup>, Thalis Bussiere<sup>(2)</sup>, Chris Holland<sup>(1)</sup> (1) The University of Sheffield, (2) Believe in Science Ltd

Low viscosity, pourable sugar-fat icings are used in the food industry as coatings for confectionery which impart pleasant taste and mouthfeel while improving shelf-life and aesthetic quality. However these icings are typically high in saturated fats and sugar, especially in commercial and

mass-produced confectioneries and as a result there is a shift towards the reduction of fat content and improvement of non-caloric sugar-replacements for many businesses in order to meet consumer and health policy demands. A notable difficulty in recipes using fat or sugar replacements is in maintaining the ideal rheological properties of industry standard icings. We report the rheological properties of both industry standard (high fat, high sugar) and reduced fatsugar icings using shear rheology. Oscillatory frequency sweeps were performed to probe icing structural properties and a Heat-Cool-Heat procedure was used to assess stiffness temperature dependence. Results have provided insight into the previously unknown state of the gelling agent, agar, in icings; reduced fat icings were seen to produce stiffer, more brittle icings; and alternative sugar icings were found to produce less stiff icings with poor thermal stability. These results will provide a foundation for fat replacement standards, further sugar alternative tests, and innovative icing recipes.

#### Creating a DEM based model for the simulation of non-Brownian suspensions of rods

Alex Dixon<sup>(1)</sup>, Chris Ness<sup>(1)</sup>, John Hone<sup>(2)</sup> (1) The University of Edinburgh, (2) Syngenta

Creating a DEM based model for the simulation of non-Brownian suspensions of rods: Rods are common in both industrial and natural processes, such as agriculture and pharmaceuticals, yet far less research exists on them compared to spheres. Rods with their high aspect ratios introduce additional complexities due to their shape anisotropy and hence orientation dependence in flow. As a result the fundamental knowledge established for spheres does not easily extend to rods, requiring new insights to understand the rheology in systems. The industrial sponsor for this project, Syngenta, have observed operational challenges. involving flow arrest and blockage of their mixing vessels in their processes that involve rod suspensions and are unsure of the cause. These issues demonstrate the need for a robust simulation model that can accurately predict the behaviour observed in these systems. This project develops a Discrete Element Method (DEM) framework that represents rods as spherocylinders, overcoming limitations of current particle-based models that approximate rods using spheres. This model includes contact mechanics as well as hydrodynamic forces, providing a more complete physical description than many existing models. By addressing these gaps, the simulation aims to advance the understanding of rod suspension rheology and provide a practical tool for solving industrial challenges.

#### Using Machine Learning to Deduce Molecular Weight Distribution from Rheology

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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 capacity' of this alternate data type when optimised for machine learning. We finally assess the performance of the models with experimental polystyrene data, in order to validate the use of computer-generated datasets.

#### Measuring Fluid Rheology using High Frequency Resonator

Guohong Gao, University of Leeds

The safe and reliable recovery of nuclear waste from legacy facilities in the UK is a crucial issue, especially given the large amount of waste in the form of suspended solids caused by corroded fuel and fuel cladding. Complex storage ponds at Sellafield retain hydroxide-based sludge where the rheological properties are largely unknown. The sludge is a corroded form of magnesium-based fuel cladding used in first generation Magnox reactors. Long term storage and open-air aqueous conditions are major contributors to its corrosion. To design effective recovery processes, a better understanding of the suspension rheology is needed. Obtaining reliable rheological data in the storage environment is essential to eliminate the influence of sample handling and history. However, traditional rheometer techniques have limitations, so this project will explore the use of a simple plate resonator to measure the viscoelasticity and yield stress of complex nuclear waste suspensions.

#### Exploring multi-stability in three-dimensional viscoelastic flow around a free stagnation point

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Fluid elements passing near a stagnation point experience finite strain rates over long persistence times, and thus accumulate large strains. By the numerical optimization of a microfluidic 6-arm cross-slot geometry, recent works have harnessed this flow type as a tool for performing uniaxial and biaxial extensional rheometry [Haward et al J. Rheol. 67 (2023) 995-1009; Haward et al J. Rheol. 67 (2023) 1011-1030]. Here we use the microfluidic 'Optimized-shape Uniaxial and Biaxial Extensional Rheometer' (OUBER) geometry to probe an elastic flow instability which is sensitive to the alignment of the extensional flow. A three-dimensional symmetry-breaking instability occurring for flow of a dilute polymer solution in the OUBER geometry is studied experimentally by leveraging tomographic particle image velocimetry. Above a critical Weissenberg number, flow in uniaxial extension undergoes a supercritical pitchfork bifurcation to a multi-stable state. However, for biaxial extension (which is simply the kinematic inverse of uniaxial extension) the instability is strongly suppressed. In uniaxial extension, the multiple stable states align in an apparently random orientation as flow joining from four neighboring inlet channels passes to one of the two opposing outlets; thus forming a mirrored asymmetry about the stagnation point. We relate the suppression of the instability in biaxial extension to the kinematic history of flow under the context of breaking the time-reversibility assumption.

# Towards phase contrast MRI of stratified multiphase flows: a probe for rheology under combined shear and extension

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A fluid packet may be strained via shearing or extension, and rheology studies a fluid's stress response to such strains. Behaviour can be very different under extensional vs. shear flow for complex fluids, making it critical to understand for situations such as polymer processing. Since changes to underlying microstructure generate rheological responses, it is a cross-coupling factor in kinematically mixed flows, flows with simultaneous shear and extension. Until now there was no method to directly and simultaneously probe specific stress responses of fluids under combined deformation: changes in full-field data such as velocity/flow pattern[1] and birefringence[2-4] could potentially be attributed to either extensional[5], shear[6], or time dependant rheology[7]. A previous study by us developed an optical PIV (particle image velocimetry) two-phase flow technique, using an immiscible Newtonian oil acting as a shear stress sensor above a non-Newtonian aqueous phase[8], allowing measurement of shear rheology under simultaneous combination of shear and extensional deformation. However, optical techniques limit the variety of materials which may be accessed. Flow velocimetry by Phase-Contrast (PC) MRI can examine a wider range of fluids, e.g. opaque suspensions, and chemically resolve these. The aim of this project is to establish a MRI method for measuring velocity profiles for two contacting, stratified, flowing immiscible fluids, one Newtonian, the other non-Newtonian. Here we outline the preliminary develops undertaken in realising this.

[1] Alves, M. A., et al., Journal of Rheology (2008) 52 (6), 1347; [2] Lee, K., et al., Journal of Rheology (2001) 45 (6), 1261; [3] Liu, Q., et al., Rheologica Acta (2022) 61 (2), 139; [4] Olley, P., et al., Plastics, Rubber and Composites (2021) 50 (1), 18; [5] Haward, S. J., et al., Physical Review Letters (2012) 109 (12), 128301; [6] Trebbin, M., et al., Proceedings of the National Academy of Sciences (2013) 110 (17), 6706; [7] Varchanis, S., et al., Journal of Non-Newtonian Fluid Mechanics (2022) 300, 104698; [8] Hodgkinson, R., et al., Journal of Rheology (2022) 66 (4), 79.

#### Structure and Dynamics of Reversible Polyurethane Adhesives

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In 2022, almost 1 million tons of polyurethane adhesives and sealants were consumed, being used primarily in the automotive, packaging, and construction industries. As these adhesives are irreversibly bonded, recycling of the adhesives and their substrates is not economically viable and as a result their use generates landfill waste. Alternative thermoplastics with dissociative covalent bonds have been prepared to address this problem, making strong adhesives that melt readily upon heating. This project seeks to test and characterise the structures and properties of these new, thermally reversible polyurethane adhesives. Methylene diisocyanate/polyester semicrystalline polyurethanes with reversible Diels-Alder chain extenders were studied. Rheological measurements were performed on the polyurethane melts. Frequency sweeps were taken over a range of temperatures and used to construct Time Temperature Superposition (TTS) master curves for the polymers' terminal and transition to flow regions. Interestingly, the TTS principle could be applied to these materials suggesting they are thermorheologically simple up to the reversible bond dissociation temperature. To correlate viscoelastic properties with structural

morphology, Small-Angle X-ray Scattering (SAXS) was employed and indicated the polyurethanes exhibit a lamellar structure with d-spacings in the range of 103-158 Å. Simultaneous SAXS/WAXS/DSC melting studies were applied to elucidate morphological changes as the materials melt and dissociate, allowing the crystalline melting point and retro Diels-Alder reaction to be followed online.

#### Impact of Protein-Protein Interactions on Viscosity and Non-Linear Rheological Behaviour of Concentrated Antibody Solutions

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Monoclonal antibody (mAb) solutions have become popular in threating a large amount of diseases, including respiratory deficiency, arthritis and various forms of cancer[1]. At concentrations above 100 mg/mL, mAbs often become too viscous for subcutaneous injection, and are also prone to liquid-liquid phase separation (LLPS)[2]. Under such conditions, short-range attractions cause mAb self-association, often resulting in high (>100 cP) zero-shear viscosity and non-Newtonian behaviour, including weak shear thinning[3-5] and elasticity during capillary thinning[6], while also displaying poor storage stability. It is unclear how the morphology of mAb clusters influences solution rheology and/or LLPS[7]. Here, we adopt a systematic approach, where the protein-protein interactions (PPIs) measured via Static and Dynamic Light Scattering[8], X-Ray Scattering[9] and Fluorescence Correlation Spectroscopy[10] are used to rationalise the non-linear rheological behaviour observed at shar rates above 10<sup>4</sup> s<sup>-1</sup> via a proprietary Rheo-chip instrument[11]. By interpreting the low concentration PPIs via a simple Baxter-Yukawa model, we surprisingly demonstrate that the shear thinning behaviour observed at high concentration is understood in terms of the percolation threshold predicted to occur for "sticky sphere" systems[12]. Such methodology is then extended to high concentration anisotropic PPIs, which are extracted from SAXS measurements via critical comparison with a 12-bead model[5, 9, 13]. Here we are able to develop a predictive framework that relates the shear viscosity and longest relaxation time with the cluster size distribution extracted from SAXS, while we show that even small changes in the hydrophobicity of single amino acids, occurring when pH and ionic strength are varied, result in a strong variation of the shear viscosity. We envision that our results will provide guideline into the design of biopharmaceutical drugs that are both easily injectable and stable while stored for long periods of time.

[1] Shukla, A.A. et al, Trends in biotechnology (2010) 28(5), 253; [2] Kingsbury, J.S., et al., Science Advances (2020) 6(32), eabb0372; [3] Zarraga, I.E., et al., Journal of pharmaceutical sciences (2013) 102(8), 2538; [4] Lanzaro, A., et al., Molecular Pharmaceutics (2021) 18(7), 2669; [5] Chowdhury, A.A., et al., Molecular Pharmaceutics (2023) 20(11), 5563; [6] Lauser, K.T., et al., Soft matter (2021) 17(42), 9624; [7] Sibanda, N., et al., Molecular Pharmaceutics (2023) 20(5), 2662 [8] Roberts, D., et al., Molecular pharmaceutics (2014) 11(7), 2475 [9] Dear, B.J., et al., The Journal of Physical Chemistry B (2019) 123(25), 5274 [10] Shah, M., et al., European Journal of Pharmaceutics and Biopharmaceutics (2020) 153, 211 [11] Lanzaro, A. et al., Journal of non-newtonian fluid mechanics (2011) 166(17-18), 1064 [12] Chiew, Y. et al., Journal of Physics A: Mathematical and General (1983) 16(11), 2599 [13] Chowdhury, A.A., et al., Molecular Pharmaceutics (2023) 20(6), 2991.

#### Investigation into mechanical behaviour of sustainable chocolate materials

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With increasing health and environmental consciousness from consumers, healthier and more sustainable food ingredients are becoming important. Chocolate is commonly made from cocoa liquor, cocoa butter, sugar, fat, milk powder and milk fat. As healthier and more sustainable ingredients are incorporated, they risk off-setting the delicate balance that creates the well-loved taste and sensory experience of chocolate. Young's modulus and yield stress have been shown to be related to textural attributes of hardness and crumbliness, properties associated with the consumers' first bite, while viscosity has been related to the 'melting mouthfeel' which is correlated to consumer preference. Three chocolate materials have been manufactured to quantify the effect a specific ingredient has on their mechanical and textural behaviour. Uni-axial compression tests were performed to determine the Young's modulus and yield stress. Vickers hardness experiments determined the relative hardness of the chocolates. Viscosity flow curves have been generated at 37°C to observe the change in relative viscosity with increasing shear rate and simulate the bolus formed during mastication. Preliminary findings in rheological experiments have revealed that, as particle size mode increased, sample viscosity decreased. Sample Vickers hardness decreased as milk fat content increased. From the compression experiments, sample Young's modulus and compressive strength decreased with increasing milk fat content.

#### Non-Newtonian fluid flow in porous medium with transverse permeability discontinuity

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The flow of shear-thinning fluids was investigated in a model porous medium with a transverse permeability discontinuity using MRI velocimetry. The fluids studied (0.25% and 0.5% Xanthan gum aqueous solutions) were also characterized rheologically. The model porous medium was a reticulated polyurethane foam with a 5 pores-per-inch (PPI) structure. A transverse permeability discontinuity was introduced by creating a cylindrical channel, with a diameter equal to the average pore length, coaxial with the bulk porous sample. MRI velocimetry experiments were conducted under varying bed thicknesses of the porous medium, fluid compositions, and flow rates. The resulting axial velocity profiles revealed significant deviations from those expected for Newtonian fluids. Within the open channel, power-law behavior was observed, whereas the porous medium exhibited preferential flow through tortuous pathways defined by its structure. Notably, the velocity profile within individual flow channels differed substantially from the primary channel flow. At the interface, velocities increased with rising bulk flow rates for both fluid compositions. These results provide a robust experimental framework for refining theoretical models of power-law fluid flow in porous media with permeability discontinuities. This study highlights the complex interplay of rheological properties, porous medium structure, and flow behavior. References: 1. Callaghan P.T. Oxford Science 1995.

#### Unsteady rheology of inertial granular flows

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Dense granular flows are commonly observed in nature and industry. The steady-state rheology of their inertial behaviour is well described by the so-called \$\mu\$(\$I\$) rheology, which defines the shear-stress-to-pressure ratio (\$\mu\$) and the solid volume fraction as functions of inertial number (\$I\$). A direct application of \$\mu\$(\$I\$) rheology to unsteady flows would imply the stresses respond instantaneously to changes in \$I\$ and vice versa – the history effect is not taken into account. To examine the importance of the history effect in dense inertial flows, unsteady flows with a varying shear rate are studied. The results clearly demonstrate that there is significant stress evolution following the rate change and that the microstructure evolves similarly to \$\mu\$ and \$I\$ using the topology of the nearest neighbours within a contact distance of 1.01 diameter.

#### Evaluation of dynamic elastomer-filler network reversibility via multiscale rheology

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The introduction of self-healing and reprocessability to conventional vulcanised rubbers has been recognised as a promising strategy to promote elastomer circularity. However, there is no solid strategy to assess the reversibility and recovery of properties in crosslinked polymer networks. The most often used static mechanical testing is unable to illustrate the self-healing mechanism at the microscale. We have explored large amplitude oscillation shear (LAOS) technology coupled with Fourier transform analysis to study the network break and recovery across linear and non-linear regimes (0.01~500%). The self-healing process of the rubber composite networks is monitored by using programmed time-temperature oscillation shear measurement. Two distinct non-linear enhancement behaviours beyond the linear viscoelastic regime were detected in the rubber nanocomposites, which were ascribed to the filler network disruption followed by the polymer network deformation. The relationship of the nonlinearity parameter  $I_{3/1}$  as a function of strain amplitude was selected to quantify the non-linear rheological responses as a complementary tool to Lissajous-Bowditch mapping, where the role of the filler and polymer on the network recovery can be differentiated. This work provides an efficient method to evaluate the self-healing and reprocessability of crosslinked rubbers and offers a fast-screening route for formulation development and sustainable rubber composite design.