OIST

Okinawa Institute of Science and Technology Graduate University

Minisymposium

Yielding and Flow of Soft Matter Systems

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Organizers: Amy Shen and Simon Haward

BOOK OF ABSTRACTS

Models, Protocols and Nomenclature for Distinguishing the Responses of Thixotropic Elastoviscoelastoplastic (TEVP) Fluids

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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, an age-dependent yield stress and more. As the lead-off talk in this workshop I hope to set the stage for future discussions by outlining the present state of the field and highlight areas in which there are agreements and disagreements; for example how do we distinguish between aging and thixotropy (or do we even bother to try?), and between viscoelasticity and thixotropy. I shall also outline how modern oscillatory rheometric techniques can be used to help distinguish the different contributions to what we may more broadly or inclusively call a *mutating material* with rheological properties that are typically both time– and shear-rate–dependent. The relative importance of these effects can be quantified by a mutation number, a Weissenberg number and a *Mnemosyne number* [1] that quantifies the importance of thixotropic effects in a fluid.

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 accurately quantify shear-induced rheological changes in materials that are gelling or crystallizing or exhibiting thixotropic effects. I will outline how to apply a *Gabor transform* (a Short Time Fourier Transform (STFT) combined with a Gaussian window) for providing optimal time-frequency resolution of the local time-resolved viscoelastic properties of a mutating material. Using measurements on a Bentonite clay, we show that Gaborheometry [2] enables us to measure more accurately rapid changes in the complex modulus and also extract a characteristic thixotropic/aging time scale for the material. We also analyze a model thixotropic silica suspension first described by Dullaert & Mewis that robustly and repeatably restructures following a strong preshearing (or *letherization*) and illustrate how parallel superposition flows can be used to arrest aging in this colloidal gel and simultaneously monitor the temporal evolution in the viscoelastic response. By varying the relative strengths of the steady shearing component that shear-melts (or *letherizes*) the microstructure compared to the superimposed oscillatory component we show how one can distinguish time-dependent thixotropic responses from evolving viscoelastic responses, and finally separate thixotropy from aging.

[1] Jamali, S., McKinley G.H. The Mnemosyne number and the rheology of remembrance, *J. Rheol.* (2022), **66**, 1027; https://doi.org/10.1122/8.0000432

[2] Rathinaraj J.D.J., McKinley, G.H. Gaborheometry: Applications of the discrete Gabor transform for time-resolved oscillatory rheometry, *J. Rheol.* (2023) **67**, 479-497 https://doi.org/10.1122/8.0000549

Microscopic Rheological Characterization of Concentrated Dispersion Fluids using Nano-Indentation Tests with a Spherical Indenter

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Evaluation of the microscopic-level rheological properties are important for understanding the structure of soft matters. Atomic Force Microscope, AFM, is generally applied to various materials because of their high force and displacement resolutions [1]. Previous studies have mainly focused on polymeric materials and drug-cell interactions, with the aim of quantitatively analyzing adhesion behavior at the nanoscale. However, considering the probe size of the AFM, AFM-based measurements are problematic for the evaluation of microrheological properties of dispersion fluids with yield stress, such as creams and concentrated suspensions.

In this study, a nanoindentation test using a spherical indenter is applied to evaluate microscopic rheological properties of concentrated dispersion fluids. The nanoindentation tester, Ultra Nano Hardness Tester (UNHT) manufactured by Anton Paar, was used. The minimum load and displacement of this instrument are 0.001 μN and 0.006 nm, respectively. A spherical indenter made of ruby with a radius of 500 μm was used. The indenter was applied at a constant loading rate and then separated at the same rate. The O/W emulsions with various multilamellar structures were used as a test sample. The Cryo-SEM image depicting the difference in lamellar structures of the simple 1 and 2 are show in Fig.1(a). The classical JKR model was fitted on the separation process to evaluate the two fitted parameters, the work of adhesion $\Delta\gamma$ and equivalent elastic modulus $E*$. Creep effects were also examined by holding the load constant for a time during the separation process. $E*$ indicates good agreement with the JKR model, except for the behavior immediately after the start of separation, by normalizing data based on Maugis' work [2]. The degree of change in $E*$ represents the characteristics of the formation of multilamellar structures formed in O/W emulsions. Significant adhesive interactions are obtained when the multilamellar bilayers are loosely stratified with respect to the droplet. Since viscosity effects such as mechanical energy dissipation and interpenetration appear as a part of Δγ, the behavior of Δγ clearly shows the microscopic characteristics of the O/W emulsions. The good correlation between experimental results and microscopic changes in the multilamellar structure of the O/W emulsions indicates that the Spherical Nano-Indentation test is a method with high potential for the evaluation of microscopic properties.

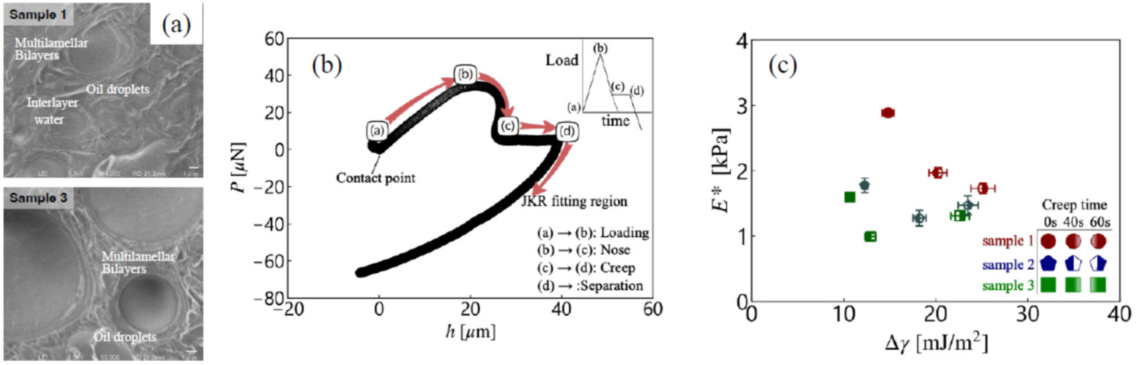

Fig 1: (a) Cryo-SEM image of sample 1 and 3, with median droplet size 2.87 μm and 3.93 μm, respectively. (b) Load P – displacement h chart with experimental sequence. (c) Equivalent elastic modulus E∗ versus work of adhesion Δγ.

[1] M. Chyasnavichyus, *et al.*, *Langmuir*, 30(35), 10566 (2014).
[2] D. Maugis, *J. Colloid and Interface Sci.*, 150(1), 243 (1992).

Geometric Effects on the Dynamics of Viscoelastic Porous Media Flows

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Viscoelastic flows through porous media become unstable and chaotic beyond critical flow conditions, impacting widespread industrial and biological processes such as enhanced oil recovery and drug delivery. Understanding the influence of the pore structure or geometry on the onset of flow instability can lead to fundamental insights into these processes and, potentially, to their optimization. Recently, for viscoelastic flows through porous media modeled by arrays of microscopic posts, it has been demonstrated that geometric disorder greatly suppressed the strength of the chaotic fluctuations that arose as the flow rate was increased [1]. However, in that work, disorder was only applied to one originally ordered configuration of posts. Here, we demonstrate experimentally that, given a slightly modified ordered array of posts, introducing disorder can also promote chaotic fluctuations. We provide a unifying explanation for these contrasting results by considering the effect of disorder on the occurrence of stagnation points exposed to the flow field, which depends on the nature of the originally ordered post array. This work provides a general understanding of how pore geometry affects the stability of viscoelastic porous media flows.

[1] D. M. Walkama, N. Waisbord, J. S. Guasto, Disorder suppresses chaos in viscoelastic flows. *Phys. Rev. Lett*. **124**, 164501 (2020)

Edge Fracture and the Tanner Number

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Edge fracture is a viscoelastic flow instability characterized by the sudden indentation of a fluid's free surface when the fluid is subjected to sufficiently strong shear. In rotational rheometry, the fracture can invade the fluid sample, decreasing its contact area with the rheometer fixture and compromising the measurement validity at high shear rates. Over the years, empirical and theoretical research has unraveled the physics behind edge fracture, enabling rheologists to develop techniques to minimize the adverse effect of fracture in their experiments. In recent years, edge fracture has also been utilized to break up rheologically complex liquid bridges efficiently, showing its potential to be adapted to the design of functional dispensing nozzles and the development of new rheometry techniques. This presentation will introduce a new dimensionless number, the Tanner number (Tn), related to edge fracture. Through simple back-of-the-envelope calculations, Tn can be used to gain physical insights into rheological phenomena involving edge fracture. Examples will be highlighted. The focus will be on the fracture of thixotropic elastoviscoplastic liquid bridges. If time permits, the prevention of fracture using a liquid metal sealant will also be discussed.

Continuum Modeling of Shear Startup in Soft Glassy Materials

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Soft Glassy Materials (SGM) consist in dense amorphous assemblies of colloidal particles of multiple shapes, elasticity, and interactions, which confer upon them solid-like properties at rest. They are ubiquitously encountered in modern engineering, including additive manufacturing, semi-solid flow cells, dip-coating, and adhesive locomotion, where they are subjected to complex mechanical histories. Such processes often include a solid-to-liquid transition induced by large enough shear, which results in complex transient phenomena such as non-monotonic stress responses, i.e., stress overshoot, and spatially heterogeneous flows, e.g., shear-banding or brittle failure. In this presentation, we will propose a pedagogical introduction to a continuum model based on a spatially-resolved fluidity approach that we recently introduced to rationalize shear-induced yielding in SGMs. Our model, which relies upon non-local effects, quantitatively captures salient features associated with such complex flows, including the rate dependence of the stress overshoot, as well as transient shear-banded flows together with nontrivial scaling laws for fluidization times. This approach offers a versatile framework to account for subtle effects, such as avalanche-like phenomena, or the impact of boundary conditions, which we illustrate by including in our model the elastohydrodynamic slippage of soft particles compressed against solid surfaces.

Are Thixotropic Yield Stress Fluids Necessarily Plastic?

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Thixotropy concerns the microstructural evolution of soft material under quiescent or weak flow conditions, causing a time-dependent increase in the viscosity; and destruction of the evolved micro-structure under strong flow conditions leading to a time-dependent decrease in the viscosity. Many thixotropic fluids show yield stress through viscosity bifurcation, in which, when a material is subjected to stress below a threshold value, the resultant shear rate decreases continuously, eventually causing the flow to cease. In the thixotropy literature, this threshold has also been referred to as the yield stress, despite deviating from the conventional definition of yield stress - according to Bingham or Herschel–Bulkley model - which requires only elastic deformation in a material when stress below the threshold is applied. However, structure kinetic models (or λ models) frequently include an independent plastic term in which yield stress is explicitly introduced as either a constant or a variable (dependent on time or shear rate through the structure parameter). In this work we ponder over a question: under what conditions is conventional yield stress as defined by Herschel–Bulkley observed in thixotropic fluids? We also discuss how a structure kinetic model, without a plastic term, intrinsically leads to constant and time dependent yield stress through viscosity bifurcation.

(virtual presentation)

Yielding, Flow, and Memory of Shear in Soft Jammed Materials

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Shearing jammed suspensions of soft particles results in heterogeneous and correlated dynamics leading to stress heterogeneities. We have studied the role of such complex dynamics in long-lasting flow inhomogeneities and in encoding memory of shear, comparing shear cessation simulations and experiments at various shear rates. We have investigated the correlations of the microscopic non-affine displacement direction and fluctuations both during shear and upon shear cessation. We find remarkable similarities between the data obtained in both regimes. This indicates that the spatially heterogeneous dynamics during shear imprints stress-inhomogeneities that reversely drive spatially heterogeneous dynamics with similar features upon shear cessation. Moreover, the final relaxation towards residual stress can be related to the evolution of local stiffness in the yielded material. Our findings suggest that steady-state shear can be thought of as a process through which memory of the imposed shear and of the distribution of local yield stresses can be built into soft jammed materials. This information can then be extracted in a shear cessation test.

Modeling and Simulations of Thixotropic Elastoviscoplastic Fluids

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We present a simple tensorial model that describes the macroscopic rheological behavior of thixotropic elasto-visco-plastic materials (TEVP). We model aggregation and disaggregation phenomena using a scalar variable that describes the level of structure at any instance and governs the transition from the liquid to the solid state. The material viscosity, elastic modulus, and relaxation time depend nonlinearly on the structure parameter, providing accurate predictions with a single-mode constitutive equation. The predictions of the new model are directly compared to experimental data of a soft colloidal glass and a commercial thermal paste in rheometric flows, including small and large amplitude oscillatory shear, simple shear, shear startup, and two-step shear tests. We also compare the model predictions with experimental data in complex flows, such as flow in microfluidic channels (past a cylinder in a straight channel and in the optimized shape cross-slot extensional rheometer) and liquid bridge twisting. In all cases, the proposed model accurately reproduces the experimental data, yielding at least semi-quantitative agreement.

Tuning the Extensional Behavior of Elastoviscoplastic Fluids

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Elastoviscoplastic (EVP) fluids, a type of yield stress fluids with elastic properties, are widely used in various industrial applications such as 3D printing, food processing, and pharmaceuticals. The inherent elasticity of these fluids allows them to store energy, thereby exhibiting partial recovery after deformation and resisting extension. This elastic behavior has been observed to impact the flow of EVP fluids in various ways, such as altering the symmetry of fluid flow and forming negative wakes behind settling spherical particles. In practical terms, the extensional characteristics of EVP can enhance the dispensing process in 3D printing systems. However, a drawback of this is the formation of secondary droplets after liquid filament breakup upon elongation, which can impact the precision of the process. Therefore, tuning the elastic properties of EVPs and understanding their effect on fluid flow is crucial. Here, we modify a well-studied EVP fluid (Pluronic F127 solution) by introducing polymer additives, in order to achieve tunable extensional behavior. We report the impact of polymer additives on the shear and extensional rheology as well as the flow behavior of the modified materials.

Colloidal Polycrystalline Grain Growths and Solid-Solid Transition Under Shear

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Colloids are outstanding model systems to reveal single-particle kinetics in solids. Our experiments measure the defects motions and phase-transition kinetics in colloidal crystals with tunable volume fractions under an oscillatory shear. The single-particle kinetics enables us to confirm the theory of shear-coupled grain boundary migration [1]. The observed rapid melting-recrystallization process may explain the stress decrease observed in the shear-induced abnormal grain growth in some metals [1]. The solid-solid (or crystal-crystal) transition exhibit rich kinetics under different shears. The growth front of the product crystal can form a stable layer of liquid; such virtual melting has been predicted in simulation, and we provide the first experimental test and an alternative explanation [2].

If time permits, I will report our more important simulation work about the yield stress or strength of crystalline-amorphous composite materials. The strength of a polycrystal can decrease or increase with the grain diameter D, i.e., the famous Hall-Petch (HP) and inverse-Hall-Petch (IHP) behaviors, respectively. Here we measure the solids' strength by systematically varing the grain diameter and grain-boundary thickness, thus generalize the HP and IHP behaviors to a function with two variables. The results reveal a way to exceed the maximum strength of normal polycrystals.

[1] W Li, Y Peng, Y Zhang, T Still, AG Yodh, Y Han, Shear-assisted grain coarsening in colloidal polycrystals, *PNAS* **117**: 24055 (2020)

[2] W Li, Y Peng, T Still, A Yodh, Y Han, Nucleation kinetics and virtual melting in shear-induced crystal–crystal transitions, DOI: https://doi.org/10.21203/rs.3.rs-1878428/v1

The Fluid Mechanics of Bacterial Swimming in Elastoviscoplastic Fluids

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Bacterial swimming in complex biological fluids such as mucus contributes to the virulence of pathogenic bacteria by allowing them to reach the plasma membrane of susceptible cells and cause systemic infections. In addition, motile nonpathogenic bacteria or synthetic swimmers may provide a route for drug delivery through mucus. Therefore, a fundamental understanding of how the rheology and properties of biological fluids affect the bacteria swimming could inform therapies by changing the properties of mucus, optimizing the design of synthetic swimmers, or developing gene regulatory programs for bacteria. Viscoelasticity is one of the notable rheological properties of biological fluids and fluid mechanics have taken a keen interest in understanding motility in continuum viscoelastic fluids. However, the scientific community has disregarded two critical facts. First, biological fluids such as mucus exhibit yield stresses, which complicates the constitutive laws by imposing a singularity on the governing equations and determines the feasibility and onset of motion. Second, biological fluids have microstructures on length scales of relevance to the flagellar propulsion. Thus, when a flagellated bacterium swims in such a medium, the cell body and the flagella experience the fluid differently. Investigating the resulting non-continuum effects is extremely challenging and requires integrating across orders of varying length scales (from nm to mm). However, it is essential to include the complexities mentioned above to advance our fluid mechanics understanding of bacterial swimming to a level applicable to biomedicine. In this talk, I will present a mechanistic argument for bacterial motility in mucosal environments, show some preliminary results, and discuss our future research directions.

(pre-recorded presentation & virtual Q/A)

What We Learn About Yielding from Recovery Rheology

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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 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 if which is yielding. New protocols inspired by recovery concepts are also used to show that small amounts of flow occur below the yield stress determined by a Herschel-Bulkley fit to steady-shear flow measurements in a predictable manner as a function of stress amplitude, angular frequency, and applied stress phase angle. Relations are presented that show how these measures can be used to determine the contribution to the loss modulus from unrecoverable plastic deformations, providing some of the same information as the full iteratively performed recovery tests, but in a fraction of the time. Recovery rheology therefore adds nuance to our understanding of yield stress fluids by highlighting the continuous nature of yielding and providing a general set of methods by which reliable and consistent yielding information can be rapidly obtained.

Emergent Functions of Electrically-induced Bubble

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Cell poration technologies offer opportunities not only to understand the activities of biological molecules but also to investigate genetic manipulation possibilities. Unfortunately, transferring large molecules that can carry huge genomic information is challenging. In this presentation, I will introduce electromechanical poration using a core–shell-structured microbubble generator, consisting of a fine microelectrode covered with a dielectric material. By introducing a microcavity at its tip, we could concentrate the electrical field with the application of electric pulses and generate microbubbles for electromechanical stimulation of cells. Specifically, the technology enables transfection with molecules that are thousands of kDa even into osteoblasts and Chlamydomonas, which are generally considered to be difficult to inject. Notably, we found that the transfection efficiency can be enhanced by adjusting the viscosity of the cell suspension, which was presumably achieved by remodeling of the membrane cytoskeleton. The applicability of the approach to a variety of cell types opens up numerous emerging gene engineering applications. In this presentation, I will present the mechanism of electrically-induced bubbles and its wide range of application including electro-mechanical poration.

Buoyancy Driven Rise of Bubbles in Elasto-visco-plastic Fluids

John Tsamopoulos

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We consider the buoyancy-driven rise of a single bubble or of a pair of coaxially placed bubbles of equal or unequal size, that are initially stationary inside an elasto-visco-plastic (EVP) material. We simulate the material using the Saramito/Herschel-Bulkley constitutive model and we fit its properties to a 0.1% aqueous Carbopol solution. The interplay between plasticity, viscoelasticity and inertia is investigated.

We find that in a single bubble the fore-aft symmetry is lost even under limiting conditions that resemble creeping flow, and a negative wake is formed in the trailing edge of the bubble. The agreement of bubble shapes and rise velocities with the corresponding ones in experiments is very satisfactory either for the smaller or the larger bubbles among those examined. The smaller bubbles have an inverted teardrop shape, a shape which is impossible to predict without including elasticity in the constitutive model. The larger ones have either an oblate or a spherical cap shape. A deviation from the experiments is found for a bubble radius for which the transition from the inverted teardrop shape to the oblate shape occurs. This deviation is attributed primarily to insufficient rheological characterization of the material, particularly in terms of its nonlinear elasticity or under extension. We show that elastic and plastic forces are dominant in the smaller bubbles, whereas in the larger ones significant shear and extensional thinning occurs making inertia forces dominant.

In bubble pairs we observe that a “bridge” of shear stresses develops, which connects the leading and the trailing bubble, decreasing the drag force on the latter and initiating their approach. The solid-like behavior of the material preserves stresses generated by the passage of the leading bubble and makes the material “softer” for the trailing bubble. At the same time the normal stresses primarily extend the bubbles, but their finite distance eventually causes the leading bubble to adopt a hydrodynamically less favorable shape that slows it down, further promoting the approach. Moreover, we examine the effect of the geometric characteristics and the material properties, such as bubble radius and distance between them, material elasticity, viscosity, or shear- and extension-thinning, and yield stress. Bubbles of equal size eventually approach each other always, but conditions exist under which bubbles of unequal size retain their initial distance.

The analysis is extended to a single drop or a pair of drops of a Newtonian liquid in an EVP material. We find that drops may extend, form a tail and breakup to a sequence of smaller ones or merge with each other to form larger ones.

Turbulence in Elastoviscoplastic Fluids

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We investigate the turbulent flow characteristics of elastoviscoplastic fluids using high-resolution direct numerical simulations at high Reynolds number, focusing on the effect of plasticity. We find that by increasing the fluid plasticity, the energy content increases at large scales while decreasing at small scales resulting in the emergence of a new intermediate scaling range. Extended self-similarity analysis of structure functions reveals that intermittency grows with fluid plasticity. The increase in intermittency is caused by the intermittent behaviour that the non-Newtonian dissipation exhibits as the fluid becomes more plastic.

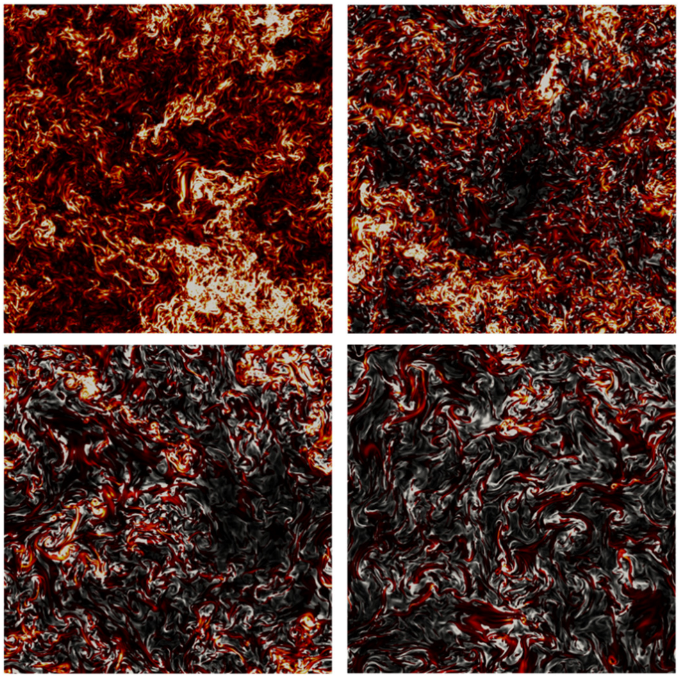

Fig. 1: Instantaneous colormaps of the turbulent fluid dissipation in homogeneous isotropic turbulence of an EVP fluid at different Bingham numbers. Yielded regions are shown with the black-red-yellow colorscale, while unyielded regions with the black-gray-white one.

Shear Localisation during Yielding of Amorphous Materials

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Amorphous materials include soft solids such as colloids, foams, emulsions, granular materials, and gels, as well as harder materials such as molecular and metallic glasses. In contrast to crystalline solids, the internal arrangement of their constituent microstructures (emulsion droplets, sand grains, etc.) lacks order. Understanding the rheological properties of these materials thus poses a major challenge. Typically, they behave elastically at low loads but yield plastically at larger loads. This talk will summarise recent progress in understanding the yielding transition between an initially solid-like state and a finally fluidised one, as a function of time since the application of an imposed strain or load, with a particular focus on the phenomenon of strain localisation during this dynamical process of yielding.

(virtual presentation)