

## CPP 55: Flow-Induced Structures in Complex Fluids (with DRG, Deutsche Rheologische Gesellschaft, and DY)

Annual Meeting of the German Rheological Society together with the Spring Meeting of the Condensed Matter section of German Physical Society

Time: Wednesday 15:00–18:30

Location: C 264

**Invited Talk** CPP 55.1 Wed 15:00 C 264

**A new perspective of materials processing** — ●KYUNG HYUN AHN — Seoul National University, Seoul, Korea

With recent advances in emerging technologies, materials design encounters new challenges. With more nano particles inside, processing experiences thinner and faster deformations than ever, which should be reflected in materials design. However, little is known about the flow characteristics of such complex fluids and less is known about how to design and control the process. Industrial coating materials such as ink, slurry and paste form heterogeneous microstructure as they contain various components. Therefore, it is necessary to incorporate the concept of heterogeneity into materials processing and to develop the methodology to quantitatively analyze the heterogeneous nature observed in both materials and processing. It will be a big challenge to establish a systematic protocol to characterize the materials and maintain uniform quality during manufacturing. In this talk, I will show illustrative examples that prove the heterogeneous nature in different length scales, covering the length scale from nano, micro to macro. With many illustrative examples of both system and methodology, I will deliver my idea on the perspective and strategy of the researches, which will be a new paradigm of materials processing as well as of materials design.

CPP 55.2 Wed 15:30 C 264

**Slow dynamics in sheared DGEBA/SiO<sub>2</sub> suspensions** — ●RICK DANNERT, ROLAND SANCTUARY, and JÖRG BALLER — University of Luxembourg, Laboratory for the Physics of Advanced Materials, Grand-Duchy of Luxembourg

Investigations of concentrated and semi-diluted colloidal suspensions of spherical silica nanoparticles in Diglycidyl Ether of Bisphenol A (DGEBA) with oscillatory shear rheology have recently shown an anomaly at low frequencies, which was interpreted as Brownian stress relaxation resulting from strain-induced perturbations of the isotropic filler distribution [1]. To complete the study of the concentration dependency we extend the rheological investigation of the low-frequency anomaly to ultra-diluted DGEBA/silica suspensions. We illustrate that the Brownian relaxation process depends in a complex manner on the volume concentration: the relaxation frequency exhibits a maximum at low filler contents. This non-monotonic dependency of the relaxation frequency can no longer be modelled by classical Peclet frequencies. Including a structural, concentration dependent parameter allows for an accurate description of the Brownian relaxation process for all concentrations.

[1] R. Dannert, R. Sanctuary, M. Thomassey, P. Elens, J.K. Krüger, J. Baller, *Rheologica Acta*, 53 (2014) 715-723.

CPP 55.3 Wed 15:45 C 264

**Microstructure and nonlinear signatures of yielding in a heterogeneous colloidal gel under large amplitude oscillatory shear** — JUNTAE KIM<sup>1</sup>, ●DIMITRI MERGER<sup>2</sup>, MANFRED WILHELM<sup>2</sup>, and MATTHEW E. HELGESON<sup>1</sup> — <sup>1</sup>Department of Chemical Engineering, University of California Santa Barbara, Santa Barbara, California 93106 — <sup>2</sup>Institute for Chemical Technology and Polymer Chemistry, Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany

We investigate yielding in a colloidal gel that forms from a nanoemulsion by polymer mediated aggregation. Combining large amplitude oscillatory shear measurements with simultaneous small and ultra-small angle neutron scattering (rheo-SANS/USANS), we characterize both the nonlinear mechanical processes and strain amplitude-dependent microstructure underlying yielding. We observe a broad, three-stage yielding process that evolves over an order of magnitude in strain amplitude between the onset of nonlinearity and flow. Analyzing the intracycle response stress reveals a transition from elastic straining to elastoplastic thinning which eventually leads to yielding and flow. The instantaneous nonlinear parameters associated with yielding are correlated with time-averaged rheo-USANS measurements. This shows how the material passes through a cascade of structural breakdown

from large to progressively smaller length scales. All significant structural changes occur on the micron-scale, suggesting that large-scale rearrangements of hundreds or thousands of particles, rather than the homogeneous rearrangement of particle-particle bonds, dominate the initial yielding of heterogeneous colloidal gels.

CPP 55.4 Wed 16:00 C 264

**Rheological behavior of a highly concentrated colloidal dispersion on different length scales** — ●CLARA WEIS and NORBERT WILLENBACHER — Karlsruhe Institute for Technology (KIT), Karlsruhe, Germany

Multi particle tracking and bulk mechanical rheometry have been used to study rheological properties of concentrated, colloidal suspensions. Using fluorescent tracer particles with particle sizes between 100nm and 1000nm enables MPT even in turbid systems and provides rheological information on the microscale. Following systems have been investigated: 1. Fluid suspensions with short range repulsive interactions at  $\Phi_{\text{eff}} < 0.5$ , in this case perfect agreement between bulk and microrheology is found. 2. Hard sphere type crystallizing dispersions in the liquid/crystalline coexistence regime with and without added non-adsorbing polymer. A large variation in mean square displacement (MSD) of different tracer particles with slopes  $\delta\text{MSD}/\delta\tau$  between 0 and 1 is found. The heterogeneity of the samples can be directly imaged based on this rheological contrast. The broadening of the coexistence region due to weak attractive depletion forces induced by added polymer is directly proven by MPT. 3. Variation of tracer particle mobility is investigated for systems approaching the colloidal glass transition. 4. The change of particle mobility and the variation of sample heterogeneity is analyzed in the so-called re-entry regime at  $\Phi > \Phi_g = 0.58$  where the system transitions from glassy to fluid and from fluid to gel-like when attractive interaction controlled by non-adsorbing polymer increases from 0 to about 10 kT.

CPP 55.5 Wed 16:15 C 264

**Microstructural studies of colloidal glasses using forced probe particles** — ●MARKUS GRUBER<sup>1</sup>, GUSTAVO ABADE<sup>1</sup>, ANTONIO PUERTAS<sup>2</sup>, and MATTHIAS FUCHS<sup>1</sup> — <sup>1</sup>Universität Konstanz, Germany — <sup>2</sup>Universidad de Almería, Spain

Driving a colloidal probe particle through a complex fluid provides unique insights into local viscoelastic properties. For soft solids there is a delocalization transition when the force on the probe particle is large enough to pull it free [1]. We study the spatial probability distribution of a tracer particle as seen by active microrheology in constant force mode. As model system, we consider a bath of hard spheres performing Brownian motion in the glassy state and an actively pulled hard sphere tracer particle. The spatial probability distribution is accessed within mode-coupling theory (MCT) refining the previous model [2] by decomposing the mobility-tensor kernel as suggested by [3] to obtain physical results for even larger forces.

Highly nonlinear effects for example in mean and mean square displacements are seen already below the critical force. One reason is the emergence of an exponential tail of the probability distribution in the direction of the applied force, which can also be found in molecular dynamic computer simulations (MDS). In addition we compare other MCT predictions with results from MDS.

[1] I. Gazuz, et al. *Phys. Rev. Lett.* 102 (2009) 248302.

[2] Ch. J. Harrer, et al. *Z. Phys. Chem.* 226 (2012) 779.

[3] S. Lang et al. *J. Stat. Mech.* P12007 (2013).

CPP 55.6 Wed 16:30 C 264

**Rheological properties of temporarily cross-linked microcapsules** — ●SARAH DEMAND and HEINZ REHAGE — Chair of Physical Chemistry II, TU Dortmund, 44227 Dortmund, Germany

Microcapsules have a broad spectrum of different applications and can be used as simple model systems for understanding the mechanical properties and controlled release processes of biological cells, e. g. erythrocytes. Capsules consist of a tiny fluid droplet which is surrounded by an ultra-thin, flexible membrane. Temporary networks,

which simply emerge from self-organization processes of surfactants, are of special interest. These coherent films exhibit striking viscoelastic properties which are influenced by dynamic fluctuations and the average life-time of cross-linking points. A typical emulsifying compound, showing this special ambivalent behavior, is the polysaccharide surfactant Span 65. In order to measure the kinetics of crosslinking and the stability of Span 65-films, we performed different types of rheological investigations. The shear and dilatational behavior of microcapsules was obtained from deformation studies in external fields. Capsule deformations in centrifugal fields were studied in a spinning-drop tensiometer. The deformation and orientation behavior in simple shear flow was investigated by means of an optical rheoscope. The results of all the studies showed a high ability of temporarily cross-linked surfactant films to stabilize emulsion droplets. Due to their processes of self-organization, this provides new and interesting perspectives for simple and effective micro- and nano-capsule technologies.

CPP 55.7 Wed 16:45 C 264

**Clusters formation in microcirculation** — OTHMANE AOUANE<sup>1,2,3</sup>, MARINE THIEBAUD<sup>2</sup>, CHAOUQI MISBAH<sup>2</sup>, and CHRISTIAN WAGNER<sup>1</sup> — <sup>1</sup>Department of Experimental Physics, Saarland University, 66123 Saarbrücken, Germany — <sup>2</sup>Université Grenoble Alpes, LIPHY, F-38000 Grenoble, France — <sup>3</sup>LMPHE, URAC 12, Faculte des Sciences, Rabat, Morocco

We investigate numerically the flow of deformable objects such as vesicles and red blood cells (rbc) in micro-channels. We focus on understanding the phenomena behind the formation of small train of cells (called clusters) that occurs in the microcirculation. We consider the rbc in 2D as a closed deformable and non-permeable membrane encapsulating an inner fluid and suspended in an outer fluid. The membrane total force is composed from a bending force, a tension force to fulfill the area conservation constraint, and a cell-cell interaction force to reproduce the depletion forces due to the effect of the plasma macromolecules such as fibrinogen. The cells are placed in a confined geometry (two parallel walls) and subjected to a Poiseuille flow. The inner and outer fluids obey to Stokes equations. This equations are solved using the boundary integral formulation. We observe that two kind of clusters exist namely: i) hydrodynamical clusters, and ii) polymer induced clusters. We notice that there is an interplay between the confinement and the formation/destruction of hydrodynamical clusters. The polymer induced clusters are more robust and remain stable independently from the confinement.

15 min. break.

CPP 55.8 Wed 17:15 C 264

**Red blood cells in intimate contact** — ACHIM GUCKENBERGER and STEPHAN GEKLE — Biofluid Simulation and Modeling, University of Bayreuth, Germany

Red blood cells in confined flow exhibit an effective hydrodynamic attraction: At low volume fractions, they tend to form pairs of cells (clusters). Understanding this effect is important for designing microfluidic devices. Furthermore, agglomeration of erythrocytes plays a major role in biological processes ranging from clotting to cardiovascular diseases. We study this phenomenon with the help of a three-dimensional periodic boundary integral method for various parameter sets. Amongst other things, the cell to cell distance is found to depend on the channel geometry and the flow rate. However, it is independent of the initial configuration of the cells.

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**Flow of complex fluids into porous media** — VIVIANE LUTZ BUENO<sup>1</sup>, MARIANNE LIEBI<sup>2</sup>, and PETER FISCHER<sup>1</sup> — <sup>1</sup>ETH, Zürich, Switzerland — <sup>2</sup>Paul Scherrer Institute, Villigen, Switzerland

The dynamic behavior of complex fluids is studied by controlling flow-geometry-fluid interactions. Quantitative information on flow-induced structures (FIS) and in-situ rheological response are reported. Wormlike micelles (WLMs) flowing through porous media lead to a highly transient and localized rheological signature, composed by shear-banding and FIS due to micellar alignment, stretching, and breaking down. The porous media is simplified by a single contraction for initial studies on shear/extensional rates, microstructure organization, and velocity fields. Flow confinement provokes shear banding and viscoelastic instabilities, which dependent on channel's geometry. Micro-designed channels provide extreme fluid confinement and tailored flow-geometry, which are investigate by flow-induced birefringence, micro-

particle image velocimetry, and scanning small-angle x-ray/neutron scattering. High shear/extensional rates in the flow through an array of cylinders cause permanent fluid gelation. By decoupling extensional and shear contributions, some of the critical factors, which influence this FIS formation, are reported.

CPP 55.10 Wed 17:45 C 264

**A thermodynamic study of shear banding in polymeric solutions** — NATALIE GERMANN — Technische Universität München, Freising, Deutschland

Shear banding is an ubiquitous phenomenon occurring in soft matter. The mechanisms behind this type of flow instability is not fully understood. It has been hypothesized (1) that the formation of localized shear bands in polymeric solutions is caused by the diffusion of the polymers. In the first part of this talk, we will introduce a new model for polymeric solutions. A new thermodynamically consistent two-fluid approach (2-3) was employed to account for Fiction diffusion and shear-induced migration effects. In this two-fluid approach, the differential velocity resulting from local variations in concentration and conformation is treated as a state variable. The additional boundary conditions arising from the spatial derivatives of the diffusion terms in the time evolution equations are now directly imposed with respect to that state variable. Hence, it is not anymore necessary to make assumptions about the polymeric microstructure on the boundaries. In the second part of this talk, we will discuss the transient behaviour of the model. The influence of the viscoelasticity of the polymers and the flow geometry on the shear band formation will be examined. The uniqueness of the numerical results and the conditions under which multiple banded states develop will also be elaborated. (1) M. Cromer, G.H. Fredrickson, and L.G. Leal, *Phys. Fluids*, 26, 063101, 2014. (2) N. Germann, L.P. Cook, and A.N. Beris. *JNNFM*, (207):21-31, 2014. (3) N. Germann, L.P. Cook, and A.N. Beris, in preparation.

CPP 55.11 Wed 18:00 C 264

**Evidence for simultaneous appearance of gradient and vorticity shear bands using time-resolved Rheo-SANS and laser light transmittance measurements** — ANNEKATHRIN MÜTZE<sup>1</sup>, PEGGY HEUNEMANN<sup>1</sup>, LIONEL PORCAR<sup>2</sup>, and PETER FISCHER<sup>1</sup> — <sup>1</sup>ETH Zürich, Schmelzbergstr. 9, 8092 Zürich, Switzerland — <sup>2</sup>Institute Laue-Langevin, 6 rue Jules Horowitz, B.P.156, F-38042 Grenoble Cedex 9, France

The flow properties of wormlike micellar surfactant solutions play an important role in applications like drag reduction in turbulent flows, fracturing fluids, and encapsulation agents. Such systems are studied with respect to the applied shear stress, concentration, temperature and composition of the salt counter ions. A combination of rheological measurements, laser-light transmittance, video analysis, and rheo-small angle neutron scattering allow a detailed exploration of number and types of shear bands. Typical flow curves of the solutions show Newtonian, shear-thinning, and shear-thickening flow behavior. In the latter regime, the solutions show vorticity and gradient shear bands simultaneously, in which vorticity shear bands dominate the visual effect, while gradient shear bands always coexist and predominate the rheological response. We show that gradient shear bands change their phases (turbid, clear) with the same frequency as the shear rate oscillates, whereas in-time vorticity shear bands change their phases with half the frequency of the shear rate [1].

[1] A. Mütze, P. Heunemann, P. Fischer. *Journal of Rheology* 58(6): 1647

CPP 55.12 Wed 18:15 C 264

**Soft Solid Rheology Near the Gel Point** — HORST HENNING WINTER — University of Massachusetts Amherst

For most amorphous materials that undergo gelation, the powerlaw relaxation time spectrum,  $H(\tau) \sim \tau^{-n}$  for  $\tau_{\min} < \tau < \tau_{\max}$ , governs the rheology only in a narrow window very close to the gel point. Soon beyond the gel point, the soft solid develops a very rich viscoelastic behavior for the evolving material states with increasing connectivity. Typical rheological features of the evolving soft solid are a growing relaxation modulus and accelerated relaxation processes for the structural components that can still relax. Time-resolved rheology measurements, in combination with time-cure superposition, on two model materials show this soft solid behavior. One model material (self-exfoliating clay-polymer composite) represents physical gelation and the other one (crosslinking polyurethane) represents chemical gelation. The relaxation characteristics near the gel point are different for

the two materials. For the physically gelling material, the modulus growth was found to be inversely proportional to the relaxation time decay. For chemical gelation, the modulus grows only with a factor of

about 0.7. During the next couple of months, more gelling materials are going to be included in the study since it is unclear how widespread the observed viscoelastic pattern occurs.