56047-UNI9
Modeling the shear thickening response of colloidal dispersions
Michael Cromer, Rochester Institute of Technology

1 Model for Shear Thickening Fluids (STFs)

A key challenge of suspension mechanics is the generation of a constitutive model that can be used for complex flows, which are not practical using detailed particle simulations. Such a constitutive model would need to take into account the underlying mechanisms of shear and extensional thickening, as well as the construction and destruction of microstructure by flow [1]. To tackle this challenge, we have created a new constitutive model based upon the one developed by Stickel et al. [2], who defined a structure tensor on the basis of particle mean free path. In the original paper, the equation describing the evolution is simplified to be linear in the deformation rate and the local structure of the particle suspension, and also involves 13 free parameters. To improve the model, while maintaining the underlying physical description, we have greatly reduced the number of parameters while also including terms nonlinear in the structure:

$$\frac{\partial \mathbf{Y}}{\partial t}+\mathbf{v}\cdot\nabla\mathbf{Y}-\mathbf{Y}\omega+\omega\mathbf{Y}=\dot{\gamma}h(Pe,\gamma^{*})[3f(\phi)\mathbf{I}-\mathbf{Y}]+c_3\mathrm{tr}(\mathbf{Y}\dot{\gamma})\mathbf{I}+c_7(\mathbf{Y}\dot{\gamma}+\dot{\gamma}\mathbf{Y}-2/3\mathrm{tr}(\mathbf{Y}\dot{\gamma})\mathbf{I})$$

$$+c_8\mathrm{tr}(\mathbf{Y}\dot{\gamma})\mathbf{Y}^2+c_9(\mathbf{Y}^2\dot{\gamma}+\dot{\gamma}\mathbf{Y}^2)$$

The new model includes the inverse dependence of the function h on the Peclet number ($Pe=S\dot{\gamma}$), which contains information about Brownian diffusion driving the system towards equilibrium. In order to simplify the model further, one major assumption we have made is that the stress in the material is a function solely of the structure, i.e., the stress relaxes instantaneously to the local structure of the suspension:

$$\mathbf{\Pi}=k_1(f(\phi)-\mathrm{tr}\mathbf{Y}/9)\mathbf{I}+k_4(\mathbf{Y}-(\mathrm{tr}\mathbf{Y}/3)\mathbf{I})$$

Ultimately, we have reduced the model to 6 parameters. While several of these are adjustable, some can be defined from experiment, e.g., small amplitude oscillatory shear. In Figure 1, we plot an example prediction of the storage, *G'*, and loss, *G''*, moduli predicted by the new model. The interesting thing to note here is that *G''* is always larger than *G'*, which has been observed in experiment [3], indicating the suspension behaves like a colloidal solution.

$$G'=2k_4c_7\gamma_0\frac{S^2\omega^3}{S^2\omega^2+1},\quad G''=2k_4c_7\gamma_0\frac{S\omega^2}{S^2\omega^2+1}+2\gamma_0\omega$$

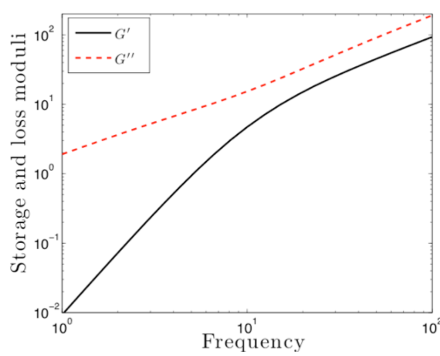

Figure 1: Storage and loss moduli vs. frequency under SAOS.

We have identified parameter spaces in which the model is capable of predicting continuous shear thickening (CST) similar to experiment [4]), and even shear thinning, Figure 2a. The thickening is due the clustering of particles perpendicular to the flow. Simulations of the new model under planar extensional flow capture the extensional thickening observed in experiment, and, furthermore, show that the extensional thickening occurs at lower rates than the shear thickening, similar to experimental results [4] (Figure 2b).

In addition to predicting the correct trends in shear and extensional thickening, which are due to the local structure of the material evolving under flow, the new model can also predict discontinuous shear thickening (DST) (Figure 3). This additional feature of the model will allow us the ability to predict and analyze the differences between CST and DST under different flow conditions, while informing the physical mechanism underlying DST.

2 Complex Flows

We have been extremely successful in simulating flows of polymeric materials in complex geometries using viscoelastic solvers developed within OpenFOAM® [5,6]. With the new model simultaneously predicting the shear

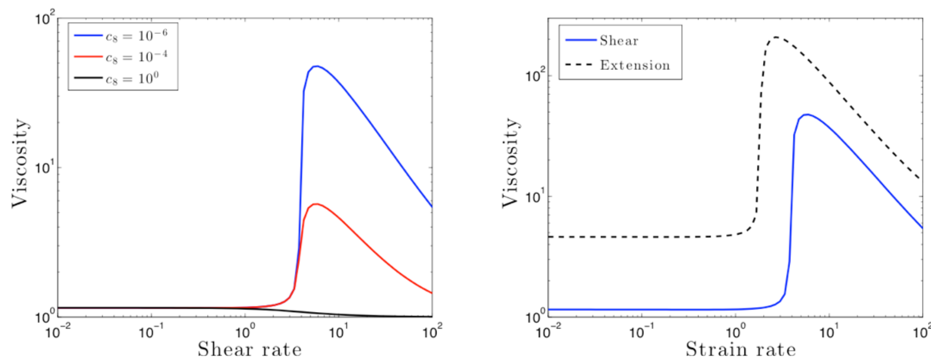

Figure 2: (a) Viscosity vs. shear rate showing the capability of the new model to predict shear thickening with varying degrees of thickening (with c_9.=0). (b) Shear vs extensional viscosity showing the new model's predictions of thickening under both flows, with the onset of thickening occurring at smaller strain rates under extension

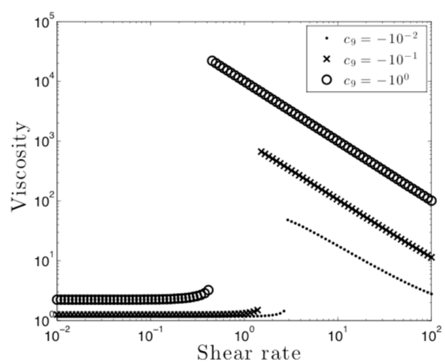

Figure3: Shear viscosity with the c_9 term included, which leads to a prediction of discontinuous shear thickening.

and extensional rheology expected of STFs, and with our success in simulating complex mathematical models and flow phenomena, we are now in a position to provide detailed studies of both the macro and micro behavior of STFs in complex, industrially-relevant flows, for example flow through a contraction [4].

3 Impact

The support from this grant has provided research opportunities for twelve undergraduate students from varying backgrounds (mathematics, chemical engineering, mechanical engineering, and physics). Each of the students gained valuable experience in fluid mechanics and rheology, programming (via Matlab and OpenFOAM), and in differential equations. The students have also presented their work in different venues. Several of the students have gone on to pursue a Ph.D. upon graduation.

The PI has gained significant experience in mentoring student research, and now currently supervises five undergraduate and two graduate research students. The support from this grant has enabled the PI to present at the annual Society of Rheology meeting in Denver, CO, as well as organize a minisymposium and present at the 2018 SIAM Meeting for Mathematical Aspects of Materials Science in Portland, OR.

References

[1] Wilson, *J. Fluid Mech.* 2018, **836**, 1-4.
[2] Stickel et al., *J. Rheol.* 2006, **50**, 379-413.
[3] Passey, 2016. *Study of Pre-Shearing Protocol and Rheological Parameters of Shear Thickening Fluids Containing Nano Particles* (Doctoral dissertation).
[4] Chellamuthu et al., *Soft Matter* 2009, **5**, 2117-2124.
[5] Kalb et al., *Phys Rev Fluids.* 2017, **2**, 071301(R).
[6] Kalb et al., *J. Non-Newtonian Fluid Mech.* 2018, **doi.org/10.1016/j.jnnfm.2018.03.012.**