Large, anisotropic structures during steady shear of colloidal gels

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Breakdown of the networked microstructure in colloidal gels subjected external stresses is responsible for failure of these materials in applications essential to industry and of recent scientific interest [1]. For example, colloidal networks built from conductive carbon particles have been utilized as "flowable electrodes" in capacitative deionization processes for desalination and in flow batteries for grid scale energy storage [2]. Flow of these networks through microfluidic electrode geometries can lead to breakdown of the network and formation of vorticity aligned flocs with significantly reduced conductive properties. This instability is widely observed in suspensions of attractive particles including: carbon black and alumina powders, carbon nanotubes, silica and polystyrene spheres, among others. The origins of the instability remain mysterious in spite of its ubiquity. To date, no computational study has been able to reproduce this effect.

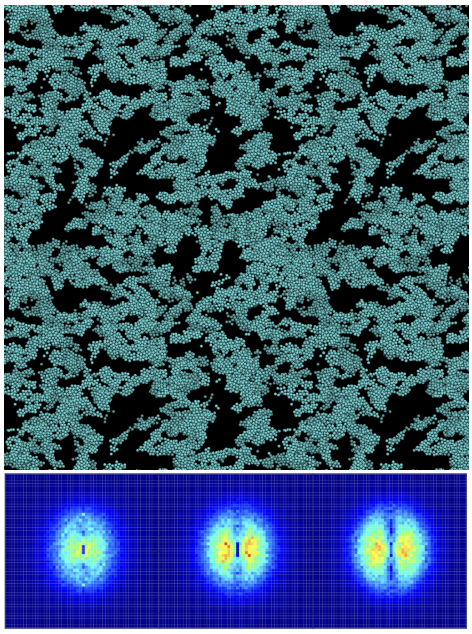

FIG. 1. (top) Snapshot of the flow-vorticity plane in a colloidal gel under shear indicating large-scale microstructural anisotropy. (bottom) Structure factors in the flow-vorticity plane with increasing shear rate applied to a colloidal gel depicting vorticity alignment.

In this talk, I discuss accelerated computational methods for modeling colloidal gels and the application of these methods to study the behavior of gels under steady shear [3]. These methods enable rapid simulations of dispersions of hydrodynamically interacting colloids with up to 10^6 particles on a single GPU. A key feature of this new technique is the ability to generate Brownian noise satisfying the fluctuation-dissipation theorem for fractal microstructures in linear time. Hydrodynamic coupling among suspended particles is shown to be key to the development and stabilization of vorticity aligned flocs in simple shear flow. The presented simulations results are the first to correctly reproduce large-scale anisotropic structures in flowing dispersions of attractive particles (see figure 1, top) [4].

The anisotropic microstructure resulting from the applied shear flow is quantified in terms of a structure factor (see figure 1, bottom) projected onto the flow-vorticity plane, which exhibits the commonly observed "butterfly" scattering pattern reported in light, x-ray, and neutron scattering experiments. A scalar measure of the degree of anisotropy is mapped onto a dimensionless parameter measuring the strength of the shear flow relative to the most probable rupture force for the bonds between the particles. This parameter ensures dynamic similarity of the steady state across a broad range of physical parameters. The characteristic size of the vorticity aligned flocs and the non-hydrodynamic contribution to the relative viscosity of the dispersion can be similarly collapsed as a function of flow rate, particle volume fraction, and interaction parameter using the same scalar parameter. Finally, comparisons with simulations neglecting hydrodynamic flows are used to determine the hydrodynamic mechanics, which stabilizes the vorticity aligned flocs under flow.

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