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Constant Stress and Pressure Rheology of Dense Colloidal Suspensions

Thesis by
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ABSTRACT

This thesis is a computational investigation on several aspects of the constant stress and pressure rheology of dense polydisperse colloidal suspensions. Using bidisperse suspensions as a model, we first study the influences of size polydispersity on short-time transport properties. The hydrodynamic interactions are calculated using a polydisperse implementation of Stokesian Dynamics (SD) via a Monte-Carlo approach. We carefully compare the SD computations with existing theoretical and numerical results, and critically assess the strengths and weaknesses of the SD algorithm. For suspensions, we find that the Pairwise Additive (PA) approximations with the Percus-Yevick structural input is valid up to volume fraction $\phi = 0.1$. We also develop an semi-analytical approximation scheme to predict the wavenumber-dependent partial hydrodynamic functions based on the $\delta\gamma$-scheme of Beenakker & Mazur [Physica 120A (1983) 388 & 126A (1984) 349], which is shown to be valid up to $\phi = 0.4$.

To meet the computation requirements of dynamic simulations, we then developed the Spectral Ewald Accelerated Stokesian Dynamics (SEASD) based on the framework of SD with extension to compressible solvents. The SEASD uses the Spectral Ewald (SE) method [Lindbo & Tornberg, J. Comput. Phys. 229 (2010) 8994] for mobility computation with flexible error control, a novel block-diagonal preconditioner for the iterative solver, and the Graphic Processing Units (GPU) acceleration. For further speedup, we developed the SEASD-nf, a polydisperse extension of the mean-field Brownian approximation of Banchio & Brady [J. Chem. Phys. 118 (2003) 10323]. The SEASD and SEASD-nf are extensively validated with static and dynamic computations, and are found to scale as $O(N \log N)$ with N the system size. The SEASD and SEASD-nf agree satisfactorily over a wide range of parameters for dynamic simulations.

Next, we investigate the colloidal film drying processes to understand the structural and mechanical implications when the constant pressure constraint is imposed by confining boundaries. The suspension is sandwiched between a stationary substrate and an interface moving either at a constant velocity or with constant imposed stress. Using Brownian Dynamics (BD) simulations without hydrodynamic interactions, we find that both fast and slow interface movement promote crystallization via distinct mechanisms. The most amorphous suspension structures occur when the interface moves at a rate comparable to particle Brownian motion. Imposing constant normal stresses leads to similar suspension behaviors, except that the interface stops moving when the suspension osmotic pressure matches the imposed stress. We also compare the simulation results with a continuum model. This work reveals the critical role of interface movement on the stress and structure of the suspension.

Finally, we study the constant shear stress and pressure rheology of dense colloidal suspensions using both BD and SEASD-nf to identify the role of hydrodynamic interactions. The constant pressure constraint is imposed by introducing a compressible solvent. We focus on the rheological, structural, and dynamical characteristics of flowing suspensions. Although hydrodynamic interactions profoundly affect the suspension structure and dynamics, they only quantitatively influence the behaviors of amorphous suspensions. The suspension becomes glassy, i.e., exhibits flow-arrest transitions, when the imposed pressure is high, and reveals the Shear Arrest Point (SAP) in the non-Brownian limit. From a granular perspective, we find that the suspensions move away from the arrested state in a universal fashion regardless of the imposed pressure, suggesting the critical role of the jamming physics. The hydrodynamic simulations quantitatively agree with the experiments of Boyer et al. [Phys. Rev. Lett. 107 (2011) 188301] with a volume fraction shift. The results at all imposed stresses and pressures reveal a generalized Stokes-Einstein-Sutherland relation with an effective temperature proportional to the pressure. We develop a model that accurately describes the rheology and diffusion of glassy suspensions. Our results show the critical role of pressure on the behaviors of dense colloidal suspensions.

PUBLISHED CONTENT AND CONTRIBUTIONS

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