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# Isotropic Viscosity of Solid Spherical Suspensions in crystalline States

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## ABSTRACT

A geometric description of particle stresses in a mixture determines isotropic viscosity of solid spherical suspensions at arbitrary concentrations based on the suspensions' microstructural crystallinity. For an equal-sized spherical suspension system, the resulting geometric expression predicts well the classical experimental and numerical viscosity data at different particle concentrations, when the crystalline states are considered as simple cubic, random distribution, face-centered cubic or body-centered cubic. It is concluded that some observed non-Newtonian behaviors of a suspension system can be construed as micro-structural transitions. This geometric model agrees remarkably well with light-scattering experimental observations on structural transitioning in colloidal and non-colloidal mixtures. Effects of particle inertia and particle Brownian fluctuations on the viscosity are taken to be dependent on solutions' crystalline states. For colloidal suspensions, flow-induced particle pressure and collective self-diffusion of particles mitigate a mixture to transitioning to a less dissipative, higher-order jamming symmetry at higher volume fractions or higher shear rates.

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## 1. Introduction

This paper deals with the classical problem for determining the effective linear viscosity coefficient of an incompressible base fluid containing solid spherical particles. Solutions of solid particles are ubiquitous in nature and practice; and a simple water-sugar mixture is one of its most common forms (Mezzenga, Schurtenberger, Burbidge & Michel, 2005). In the most general case, viscosity of a mixture is dependent on the suspended particles' geometries, concentrations, and the mixture's flow. Determination of a mixture viscosity with prescribed particle and flow characterizations has remained an important and open question of rheology (Mewis & Wagner, 2009), as well as theoretical and statistical physics (Dhont, 2003; Ramaswamy, 1997). To approach this problem, an understanding of spherical particle effects on the mixtures' properties can be key to analyze and approach a more general mixture composed of solid particles of arbitrary-geometry and arbitrary concentration. For spherical particles, to arrive at a simple and physical form, in addition to the simplifying assumption on particle geometry, this work treats all particles to be of equal size and diameter while interactively possessing a flow-induced effective crystalline state during shearing. Reviews and guides on available mixture mechanics and their possible applications have been provided previously (Klika, 2014; Ranjagopal and Tao, 1995).

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### 1.1. Crystalline states

Early pioneering works of Hoffman (1972) and Ackerson (1990) and his collaborators (Liu, Weitz & Ackerson, 1993) have been followed by a great progress in observing and categorizing the crystalline state of spherical suspensions, particularly in near jamming states in the flow-vorticity and the flow-gradient planes of shear flows (Vermant & Solomon, 2005). Existence of these structures in less concentrated states and well into the “dilute” limit have also been reported in a few instances (Verhaegh, van Duijneveldt, van Blaaderen & Lekkerkerker, 1995). However, for a large concentration spectrum, formation of crystalline structures has been only recently verified for suspensions of non-spherical geometries (O’Brien, Martin, Hai-Xin, Millana, de la Cruz, Leed & Mirkina, 2016; Briand & Dauchot, 2017).

Individual particles in a sheared mixture have unsteady motion due to thermal motion and changing and/or shifting stress fields induced by neighboring moving particles. Unsteady particle motion can give rise to instantaneous microstructural formation with adjacent particles. Such local ordered states are fragile formations in non-equilibrium states with continuous local formations and breakups of such crystalline structures. Point-wise sequential light-scattering observations in sheared mixture and computer simulations have depicted transitory nature of structures (Ackerson, 1990; Butler & Harrowell, 1996). Accordingly, if one considers existence of a microstructure in a mixture under shear, such structures are hydrodynamic quasi-equilibrium states that are prevalent in the mixture and can characterize macroscopic properties of a mixture by an effective crystalline microstructure.

Ackerson (1990) studied and detailed transitional microstructural changes for both colloidal and non-colloidal particles in an isotropic fluid. In a suspension with a given uniform microstructure, a new microstructural zone can emerge beyond a threshold shear on the mixture. Distinct microstructural phase boundaries exist between the two microstructural zones, each zone with a uniform microstructural symmetry. Microstructural phase boundaries, sharp or fuzzy, are likewise transient structures that can be observed in spot formations (Butler & Harrowell, 1996), wall-boundary-driven layers (Ackerson, 1990) and planar progression of one zone over the other with changing the shear stress on the mixture (Shereda, Larson, Solomon 2010). A change in the applied shear can change the shifting hydrodynamics between surrounding particles giving rise to a new structure in coexistence and equilibrium within an existing microstructure.

Existence and emergence of hybrid microstructural states of the suspension particles in solutions has two analytical drawbacks. First, such states won’t be isotropic locally throughout the mixture and no analytical solution yet exist for these hybrid microstructural mixtures. However, if both these microstructures are individually isotropic within each zone, an isotropic analytic solution can be applied separately for each zone and approximate the overall effective viscosity of hybrid structural spherical mixtures. Second, with two different coexisting microstructures in a suspension, particle average distances will be different for each zone and each zone will have its own particle concentration. It follows that a single concentration parameter can’t characterize macroscale properties of such hybrid mixtures. Theoretical, analytical and experimental studies on mixture viscosity in the literature, irrespective of presence of such hybrid states in their systems, express their results exclusively in terms of the particle volume fraction (Foss & Brady, 2000; Guy, Hermes & Poon, 2015). Accordingly, interpretation of a hybrid structure based on a given volume fraction from the data will be an approximation.

### 1.2. Inertial Effects

In the limit of  $Re_p \ll 1$ , with  $Re_p \equiv \frac{d^2 |\overline{\epsilon}_{ij}|}{\nu_o}$  is the particle Reynolds number,  $d$  the particle diameter,  $|\overline{\epsilon}_{ij}|$  magnitude of the ensemble-average strain rate tensor and  $\nu_o$  the kinematic viscosity of the base fluid, Stokesian Dynamic simulations have provided numerical values for the effective viscosity of spherical suspensions for a large spectrum of particle concentrations and have successfully captured near and far fields hydrodynamic interactions as well as the role that thermal motion and intra-particle forces play in impacting the solution rheology (Brady, 1996; Sierou & Brady, 2002). In a recent related study, Brownian dynamic simulations of solid spherical particles in high concentrations are used to arrest emergence of isotropic ordered structures in the shear plane and anisotropic layered structures in the flow-vorticity plane under oscillatory shearing (Koumdis, Brady & Petekidis, 2016).

Particle Reynolds number  $Re_p$  characterizes inertial effects in analytical and theoretical treatment on this subject (Stickel & Powell, 2005); and while for most suspensions its value is much less than unity, in case of very large shear rates and/or larger particles in the solution,  $Re_p$  can become larger than unit, theoretically rendering inclusion of inertial terms in the analysis. However, when three or more particles hydrodynamically interacting, observations in a linear shear flow have shown that particles interactions are not reversible when  $Re_p > 10^{-5}$  (Karnis, Goldsmith & Masons, 1966), indicating that inertial effects can’t be ignored even for very small particle Reynolds numbers. Inertial effects can also be present when particles are moving next to a solid wall creating a Vand zone and particles in a non-uniform shear rate generating a Segre-Silberberg effect (Takano, Goldsmith & Mason, 1968). In this no-flow analysis, no assumption on  $Re_p$  will be made and its effects on suspension viscosity are discussed in terms of microstructural variations of the particles’ relative positions and assuming that particles have a uniform distribution.

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