A Mean Field Model of Layering Instability in Shearing Suspensions

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Abstract

Concentrated suspensions may shear-thin when the suspended particles form planar sheets that slide over one another with less friction than if the particles are randomly distributed. In a naïve model the suspension is described by a mean effective viscosity, and particles that collide with each other redistribute the mean density in the shearing direction. This leads to a diffusion equation for the particle density. If the viscosity in the unthinned state is a steeply increasing function of particle density the effective diffusion coefficient is negative and the diffusion equation, meaningful only on scales larger than the particle separation, is ill-posed. This singularity corresponds to the formation of planar sheets of particles and defines a critical particle density for the onset of shear thinning.

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Shear thinning of suspensions is ubiquitous. It is, at least in part, attributed to the formation of layers of suspended particles in planes normal to the velocity gradient [1], although the relation between layering and shear thinning is complex [2, 3, 4]. Recent theoretical work on this phenomenon [5, 6] has applied thermodynamic methods (dynamic density functional theory) to colloidal suspensions in which Brownian motion is significant. At high Péclet number a wall induces layering that propagates (at a rate not calculated) into the bulk. Yet shear thinning is also observed in non-Brownian suspensions, such as those of corn starch [1, 7], so it must also be explicable by non-thermodynamic methods.

Shear-induced diffusion resulting from particle collisions can make particles migrate through a rheometer and thereby decrease the measured viscosity [8], an effect distinct from shear thinning. Here I present a mechanism by which particle collisions in sheared bulk suspensions induce structure [1] on the scale of the particle size, and to shear thinning, on time scales $\mathcal{O}(1/\dot{\gamma})$, where $\dot{\gamma}$ is the mean (macroscopic) shear rate. This is a much shorter time scale than that of shear-induced transport [8].

Consider a simple model of shear thinning in a suspension of neutrally buoyant particles interacting with only a hard-sphere repulsion. Shear thinning must be distinguished from thixotropy [9, 10] that results from the breakup of clusters of mutually attracting particles, but these do not exist in this hard-sphere model. Unlike earlier models of sheared colloids [5, 6], the model does not assume the presence of a wall. Shear stress may be a boundary condition at a wall, but here it is treated as an initial condition in an unbounded suspension.

Assume a steady plane creeping flow in the $\hat{x}$ direction, with velocity gradient $\dot{\gamma} = \frac{\partial v_x(z)}{\partial z}$ in the $\hat{z}$ direction and a constant shear stress σ_{xz}, and drop subscripts for convenience. Implicitly, v and σ are mean quantities averaged over scales of the size of the particles, taken to be spheres of radius a and mean number density $n(z)$. The suspending fluid has a viscosity η_f, and the suspension is described as having a fluid viscosity η, again averaging over its structure on scales $\mathcal{O}(a)$ in this mean field model.

In a concentrated suspension multi-particle interactions are significant, but they are not calculable or even approximable analytically, so I only consider two-body interactions. The error introduced by this approximation is likely comparable to that introduced by the approximate treatment of two-body interactions. The purpose of this treatment is necessarily only to provide a qualitative explanation of the occurrence of layering and shear thinning, and not a quantitative prediction of the properties of the layered state.

Consider two spheres initially with centers (x_1, z) and $(x_2, z + \Delta z)$, $|\Delta z| < 2a$ and $(x_1 - x_2)\Delta z \frac{\partial v}{\partial z} > 0$. If the spheres were to continue to move at constant z and Δz they would collide. In fact, in this low Reynolds number flow they will interact hydrodynamically (and with many neighboring particles in a concentrated suspension). When they approach closely their separation will shrink exponentially under the influence of the hydrodynamic stress $\sigma = \eta\dot{\gamma}$ with a time constant $\mathcal{O}(\eta_f/\sigma) = \mathcal{O}[\eta_f/(\eta\dot{\gamma})] \ll 1/\dot{\gamma}$, where η is the macroscopic suspension viscosity and the thin film of fluid between the spheres is described by the fluid viscosity η_f. Hence, despite their hydrodynamic interactions, their interaction involves (with any finite, even atomic-scale, surface roughness) a hard sphere collision, at least for small enough $|\Delta z|$ (for larger $|\Delta z|$ hydrodynamic interaction may prevent such a close approach, a possibility we ignore). The hard-sphere repulsion breaks the kinematic reversibility of creeping flow [8, 11] that would otherwise ensure that after interaction the spheres return to the initial values of their z coordinates.

In a two-dimensional approximation, in which the spheres are confined to the $y = 0$ plane (an approximation that simplifies the geometry of their interaction without changing its qualitative properties), the consequence of this collision is that when $x_1 = x_2$ the spheres touch ($|z_2 - z_1| = 2a$) in order that they may pass each other; depending on the initial Δz they may come into contact even before then. Again ignoring their hydrodynamic interaction by assuming that z_1 and z_2 do not change as the spheres move apart in the shear flow, the net displacements are

$$\delta z = \frac{\Delta z}{2} - \mathrm{sgn}(\Delta z)a. \tag{1}$$

This geometry is shown in Fig. 1.

The rate of collisions of a sphere with center (x, z), taking all variables independent of x, is

$$\int_{\Delta z=-2a}^{2a} d\Delta z \left|\frac{\partial v}{\partial z}\Delta z\right| n(z + \Delta z). \tag{2}$$

The mean vertical velocity of this sphere is

$$v_z = \int_{\Delta z=-2a}^{2a} d\Delta z \left|\frac{\partial v}{\partial z}\Delta z\right| n(z + \Delta z)\delta z, \tag{3}$$

similar to a result of [12]. By convention, we take $\partial v/\partial z > 0$.

In order to evaluate this expression we note that the assumption of constant and uniform

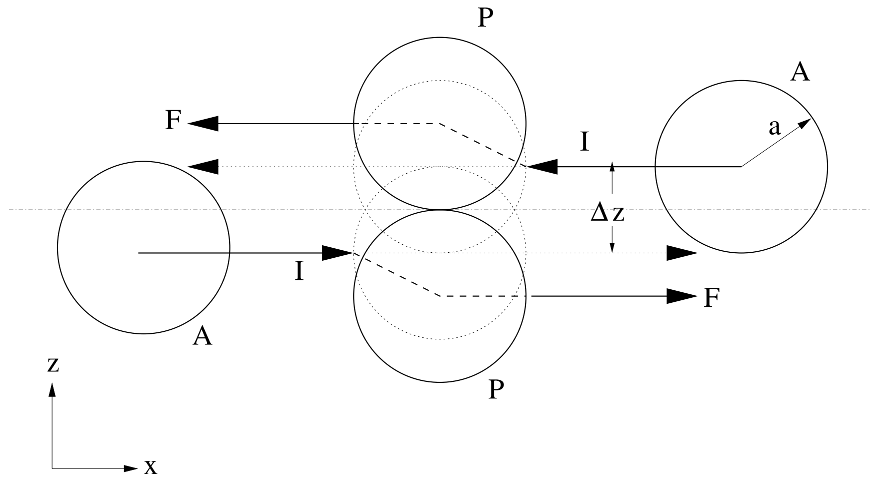

FIG. 1: Toy model of interaction of two spheres of radius a in $y = 0$ plane in a shear flow $v_x = \dot{\gamma} z$. The centers of the spheres are initially on trajectories I separated by $\Delta z < 2a$. A indicates the locations of the spheres before interaction, F indicates the paths of their centers after interaction, and P indicates their nominal positions at closest approach. The dashed lines indicate (but do not represent quantitatively) their paths during interaction. Dotted circles and lines indicate overlapping positions and paths of undeflected (interpenetrating) spheres. The model is only qualitative because it neglects the complex hydrodynamic interaction of the spheres with each other and with other particles in a concentrated suspension.

σ (as found in steady planar flow) relates the velocity gradient at $z + \Delta z$ to the viscosity there

$$\frac{\partial v(z+\Delta z)}{\partial z} \approx \frac{\sigma}{\eta_0 + \frac{d\eta}{dn}\Big|_{n=n_0} \frac{\partial n}{\partial z}\Delta z} \approx \frac{\sigma}{\eta_0}\left(1 - \eta' \frac{\partial n}{\partial z}\frac{\Delta z}{n_0}\right), \tag{4}$$

where $\eta' \equiv \frac{d\ln\eta}{d\ln n}\Big|_{n=n_0}$, n_0 is the mean density of spheres per unit area in the $y = 0$ plane, the viscosity has been expanded around $\eta(n_0) \equiv \eta_0$ and Δz and variables have been expanded to first order around their mean values. Integrating Eq. 3, using Eqs. 1, 4, and $n(z+\Delta z) \approx n_0 + \frac{\partial n}{\partial z}\Delta z$, and taking only the lowest non-vanishing (linear) order in $\frac{\partial n}{\partial z}$, we find

$$\begin{aligned} v_z = &\int_0^{2a} d\Delta z \frac{\sigma}{\eta_0}\left[\Delta z n_0 \left(\frac{\Delta z}{2} - a\right) + \frac{\partial n}{\partial z}\left(1-\eta'\right)\Delta z^2 \left(\frac{\Delta z}{2} - a\right) + \ldots\right] \\ &- \int_{-2a}^{0} d\Delta z \frac{\sigma}{\eta_0}\left[\Delta z n_0 \left(\frac{\Delta z}{2} + a\right) + \frac{\partial n}{\partial z}\left(1-\eta'\right)\Delta z^2 \left(\frac{\Delta z}{2} + a\right) + \ldots\right] \\ \approx &\frac{4a^4\sigma}{3\eta_0}\left(\eta' - 1\right)\frac{\partial n}{\partial z}, \end{aligned} \tag{5}$$

where the minus sign in the second term comes from the absolute value in Eq. 3. This result is comparable to the equally approximate results of [5, 6].

The flow of particles satisfies the one-dimensional continuity equation

$$\frac{\partial n}{\partial t} + n_0 \frac{\partial v_z}{\partial z} = 0, \tag{6}$$

where we have approximated $n \approx n_0$ because v_z is first order in small quantities and we are interested in infinitesimal perturbations from a homogeneous state. Substituting Eq. 5 into Eq. 6 yields a diffusion equation

$$\frac{\partial n}{\partial t} + \frac{4a^4 \sigma n_0}{3\eta_0} \left(\eta' - 1\right) \frac{\partial^2 n}{\partial z^2} = 0. \tag{7}$$

The diffusion coefficient resulting from collisions between particles

$$D_{zz} = \frac{4n_0 \sigma a^4}{3\eta_0} \left(1 - \eta'\right) = \frac{4a^2 \phi \sigma}{3\pi \eta_0} \left(1 - \eta'\right) = \phi \dot{\gamma} \frac{4a^2}{3\pi} \left(1 - \eta'\right), \tag{8}$$

where the (two-dimensional) filling factor in the $y = 0$ plane $\phi = \pi a^2 n_0$ and the stress $\sigma = \eta_0 \dot{\gamma}$. To order of magnitude, $D_{zz} \sim a^2 \dot{\gamma}$, as must be the case because this is the only quantity with dimensions of diffusivity that can be formed from the parameters of the problem.

In general, in a concentrated suspension the viscosity is a steeply increasing function of ϕ [2] so that we expect $\eta' > 1$ and $D_{zz} < 0$. The diffusion equation is ill-posed unless the negative D_{zz} is offset by a positive diffusivity. At finite temperature Brownian diffusion adds a positive $D_{th} \sim (kT/\eta_0 a)$, where in a concentrated suspension the coefficient depends on the environment. However, the total diffusivity $D = D_{th} + D_{zz}$ will still be negative (if $\eta' > 1$) for a greater than some threshold corresponding to large Péclet number, and large enough $\eta_0(\phi)$, corresponding to a sufficiently concentrated suspension. The condition $D < 0$ becomes

$$\eta_0(\phi) \gtrsim \frac{3\pi}{4} \frac{kT}{a^3 \phi \dot{\gamma} (\eta' - 1)}. \tag{9}$$

If the inequality (9) is satisfied the mathematical catastrophe of ill-posedness is avoided because Eq. 8 only describes mean field fluid quantities such as n_0 and η that are not defined on scales $\lesssim a\phi^{-1/2}$. The fastest growing perturbations are those with wavelengths $\sim a\phi^{-1/2}$ that have e-folding times

$$t_{growth} \sim \frac{a^2}{D_{zz} \phi} \sim \frac{1}{\phi \dot{\gamma} (\eta' - 1)}. \tag{10}$$

This instability will saturate at finite amplitude at which Eq. 7 breaks down because higher-order or nonlinear terms become significant. The result is the formation of structure in the $\hat{z}$ direction such as the sheets in $\hat{x}$-$\hat{y}$ planes observed for shear thinning suspensions [1]. The inequality (9) is then the criterion for the onset of layering. If the suspension is non-Brownian (the limit $kT/a^3 \to 0$) then the instability criterion reduces to $D_{zz} < 0$ or $\eta'(\phi) > 1$, which is an implicit criterion for ϕ. In contrast to the results of [5, 6, 12], this layering instability is a bulk phenomenon, and is not dependent on confinement or hydrodynamic interaction with walls.

Allowing for separation of interacting particles in the third (vorticity) dimension $\hat{y}$ would reduce v_z and D_{zz} by factors $\mathcal{O}(1)$ but would not change the conclusion that a layering instability occurs for $\eta' > 1$. However, because there is no momentum flow in the $\hat{y}$ direction ($\sigma_{xy} = 0$) there is no equation for $\frac{\partial v}{\partial y}$ analogous to Eq. 4 and no instability producing structure in that direction.

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