One-dimensional analysis of a continuum model for structure in electrorheological fluids

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Abstract

A continuum model is used to describe the evolution of nonuniform particle concentration distributions in electrorheological and magnetorheological fluids. Particles are shown to migrate relative to the bulk mixture motion as the result of the particle-phase contribution to the suspension stress. The particle stress results from both electrostatic and hydrodynamic contributions; the ratio of hydrodynamic to electrostatic stress is characterized by the Mason number, Mn. The model predicts structures which are in agreement with those observed experimentally for both unsheared and sheared conditions. The emphasis here is on the sheared condition, where particle-rich stripes with normal along the vorticity direction of a simple shear flow are observed when a field is applied in the direction of the velocity gradient. The linear stability and steady-state structures are analyzed for the one spatial dimensional model with variation in the vorticity direction. The linear stability analysis predicts a critical Mason number below which the uniformly distributed suspension demixes to a striped structure. The particle concentration in the steady state stripe structures is found to depend on Mn, with concentration decreasing with increasing Mn.

1 Introduction

This work addresses the bulk flow of electrorheological and magnetorheological (ER and MR) fluids. These fluids, composed of solid particles in liquid, have rheological properties which can be dramatically altered by the application of an external electric or magnetic field, depending upon whether we consider the ER
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or MR case, respectively. Considerable practical interest in these materials has resulted from the tunability of the rheology and potential applications include various damping devices [1]. In such applications, the numbers of particles present is very large, and description of the mixture flow using discrete-particle simulations is impractical. For practical design purposes, a continuum description is of value, and we have in earlier work [2, 3] presented a framework for the description of the bulk flow of mixtures subjected to external electromagnetic fields. This approach captures several phenomena observed experimentally in shear flows of ER and MR fluids, including the “stripe” structures upon which we will focus here.

The application of a field to an ER or MR fluid results in the formation of chains of particles which over time tend to grow as chains of single particle scale join to create thicker columns. Upon application of a shear flow to electrified ER fluid, the stripe structures appear: these have particle-rich and essentially pure liquid zones alternating as one progresses along the vorticity axis, with the electric field in the direction of the velocity gradient; for a simple shear flow $u_x = \gamma z$ with $\gamma$ the shear rate, $z$ is the vorticity axis and we consider cases with the electric field along $z$. The structures have been described in more detail [4], analyzed by other approaches [5], and also have been seen in simulations [6].

The goal of this work is to illustrate that a mixture flow model which includes the particle-induced rheology predicts these structures. The basic idea which the model utilizes is that the stress state in a mixture flow depends upon the shear rate, the composition (here given by the particle volume fraction, $\phi$), and external electric fields if the particles are polarizable. From the mass and momentum conservation equations of the mixture, we develop a predictive model for the particle volume fraction which incorporates the electrostatic and hydrodynamic contributions to the stress. Linear stability and steady state analyses of the model equations yield predictions for the conditions of onset of the banding structure and the concentration in the steady stripes, respectively.

The following section outlines the model and reduces it to the one-dimensional form. In §3, linear stability and steady state predictions of the model are described.

2 Two-fluid model description

Electrorheological fluids are suspensions of solid particles in liquid, with the particles and fluid having a dielectric (or polarizability) mismatch. The application of an electric field across the suspension induces the formation of electrical dipoles in the particles, and these dipoles then generate interparticle forces and contribute to an electrostatic bulk stress. Detailed discussions and analysis of this topic are found elsewhere [7, 8]. Because ER fluids are suspensions typically flowing at small Reynolds number (vanishing inertia), we find it convenient to use a flow model developed for description of such suspensions [9, 10]. The key influence of the electric field is in the rheology of the mixture, and the balance between the electrostatic and hydrodynamic stresses is of particular interest here.
2.1 Particle stress contributions

The particle stress is written

$$\sigma^p = \sigma^{(p,E)} + \sigma^{(p,H)},$$  \hspace{1cm} (1)

where $\sigma^{(p,E)}$ and $\sigma^{(p,H)}$ are the electrostatic and hydrodynamic particle stress contributions, respectively. These are described below.

2.1.1 Electrostatic contribution

We consider a suspension composed of dielectric spheres in a liquid of different dielectric constant. Prior to application of the field, we assume a random isotropic structure. For the flowing system, this neglects the anisotropy in pair correlation due to shearing [11, 12, 13]; consideration of the coupled structure due to electromagnetic fields and shear flow is deferred to future work. Upon application of a field, $E$, the particulate contribution to the electrostatic stress is

$$\sigma^{(p,E)} = \sigma^E - \sigma^E(\phi = 0) = \epsilon_0 \left( (\epsilon - \epsilon_c) EE - \frac{1}{2} (\epsilon - \epsilon_c + \alpha_2) E^2 \delta \right),$$  \hspace{1cm} (2)

where $\sigma^E$ is the electrostatic stress of the bulk mixture, and $\epsilon_0 = 8.8542 \times 10^{-12}$ F/m is the permittivity of free space. The suspension dielectric constant, given self-consistently by mean-field analysis in the point-dipole limit [8], is

$$\epsilon(\phi) = \epsilon_c \frac{1 + 2\beta\phi}{1 - \beta\phi},$$  \hspace{1cm} (3)

where $\epsilon_c$ is the dielectric constant of the suspending fluid, $\epsilon_p$ is that of the particle material, and $\beta = (\epsilon_p - \epsilon_c)/(\epsilon_p + 2\epsilon_c)$. Typically for good performance, $\beta \approx 1$. We assume that neither the dispersed nor the continuous phase is individually electrostrictive, under which condition the electrostriction coefficient is [8]

$$\alpha_2(\phi) = \frac{(\epsilon(\phi) - \epsilon_c)(\epsilon(\phi) + 2\epsilon_c)}{3\epsilon_c}. $$  \hspace{1cm} (4)

The total electrostatic stress includes contributions due to the continuous fluid [2, 3] which are not relevant to the present analysis.

2.1.2 Hydrodynamic contribution

Addition of small particles to a liquid is well known to result in increased viscosity, and also results in shear thinning and thickening under appropriate conditions [14]. A more striking rheological feature of suspensions, particularly when the suspensions are composed of spherical particles, is the presence of normal stresses arising from anisotropy in the microstructure [11, 12, 15]. We consider suspensions of particles sufficiently large that Brownian motion may be neglected, and
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also do not consider shear thinning and thickening. For this condition, a model for the particle contribution to the hydrodynamic stress has been proposed [10],

$$\sigma^{(p,H)} = -\eta_n \dot{\gamma} \hat{Q} + 2\eta_p e,$$

(5)

where the first term models the shear-induced normal stresses, the rate of strain is given by $\dot{e}$ with $e_{ij} = 1/2(\nabla_i (u)_j + \nabla_j (u)_i)$, and we define $\dot{\gamma} \equiv \sqrt{2e} :e$. The $\phi$ dependence of the stress is captured by two functions, $\eta_n(\phi)$ which is termed the “normal stress viscosity,” and $\eta_p(\phi)$ which is the particle contribution to the shear viscosity. The $x$, $y$, and $z$ directions (or 1, 2, 3) are here oriented with the flow, vorticity, and gradient directions, respectively, and the normal stresses are specified by

$$\hat{Q} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \lambda_2 & 0 \\ 0 & 0 & \lambda_3 \end{pmatrix},$$

(6)

where $\lambda_2 = 0.5$ and $\lambda_3 = 0.8$ provide best fits of various suspension flows; note that this work differs from that of the cited reference [10] in its coordinate labeling, as in that work the $y$ and $z$ directions are the gradient and vorticity directions. For present purposes, the anisotropy of the normal stress yielding normal stress differences is not of much relevance, but the fact these are compressive (negative) stresses, and thus tend to be dispersive in a simple shear flow, is critical.

The normal stress viscosity was treated empirically [10], by writing

$$\eta_n = K_n \eta_c \left( \frac{\phi}{\phi_{\text{max}}} \right)^2 \left( 1 - \frac{\phi}{\phi_{\text{max}}} \right)^{-2},$$

(7)

where $\eta_c$ is the pure fluid (continuous phase) viscosity, $\phi_{\text{max}}$ is the maximum packing fraction and is taken as 0.68, and $K_n = 0.75$ provided reasonable agreement with experimental data. Because the shear rate will be assumed fixed in this work, conclusions of this work are unchanged by modeling of the shear viscosity, $\eta_s = \eta_p + \eta_c$, and any of several similar forms generate qualitatively similar results in cases where the shear rate may vary.

2.2 Mass and momentum conservation

We begin from the continuum statements of mass and momentum conservation for both the bulk suspension and the particle phase. The bulk balances are

$$\nabla \cdot \mathbf{u} = 0 \quad \text{and} \quad \nabla \cdot \mathbf{\Sigma} = 0,$$

(8)

which express incompressibility and the inertia-free condition of the entire mixture, respectively. Averaging the general mass conservation equation over the particle phase, we obtain the equation for conservation of particle mass,

$$\frac{\partial \phi}{\partial \bar{\theta}} + \langle \mathbf{u} \rangle \cdot \nabla \phi = -\nabla \cdot \mathbf{j},$$

(9)
where $\phi$ is the particle volume fraction, $\langle u \rangle$ is the suspension average velocity, $j = \phi(U - \langle u \rangle)$ is the particle flux relative to the mean suspension motion, and $U$ is the local average velocity of the particulate phase. A similar procedure applied to the general momentum conservation equation gives (for $Re = 0$)

$$0 = \nabla \cdot \sigma^p + n F^H + \phi(\rho_p - \langle \rho \rangle) g. \quad (10)$$

For the hydrodynamic drag force, $F^H$, we use a generalization of Faxén’s drag law to higher concentrations, which amounts to a sedimentation model, $F^H = -6\pi \eta_c a f^{-1}(\phi)(U - \langle u \rangle)$; $f(\phi) = (1 - \phi)^\alpha$ is the sedimentation function with $\alpha = 2 - 5$ the recommended values, with $\alpha = 4$ used here [10]. With this substitution for $F^H$, we find in the case of neutrally buoyant particles that the particle flux is

$$j = \phi(U - \langle u \rangle) = \frac{2a^2}{9\eta_c} f(\phi) \nabla \cdot \sigma^p. \quad (11)$$

Inserting (11) to (9), and noting that the particle stress is composed of both electrostatic and hydrodynamic contributions yields an evolution equation relating variation of $\phi$ to the particle-induced rheology:

$$\frac{\partial \phi}{\partial t} + \langle u \rangle \cdot \nabla \phi = -\frac{2a^2}{9\eta_c} \nabla \cdot \left[ f(\phi) \nabla \cdot \left( \sigma^{(E)} + \sigma^{(H)} \right) \right]. \quad (12)$$

To provide a complete description of the concentration field, constitutive relations for the stresses, e.g. (2) and (5), must be introduced, and (12) must be coupled both with the Maxwell’s equations for the electrostatic part of the problem, and with the bulk flow conservation statements (8). In the present instance, we will consider a flow which remains a simple shear flow, $u_x = \dot{\gamma} z$, with $z$ also the direction of the electric field.

### 2.3 Reduction to a one-dimensional model

It has been demonstrated in our prior work [2, 3] that for the quiescent system a stability analysis of (12) with linearized $\phi$ dependence leads to predictions of columnar structures in the plane perpendicular to the field, with the fastest-growing structures being the thinnest. Nonlocal electrostatics [3] sets a lower limit of the scale of the particle size on the wavelength, so that the fastest-growing structures are predicted to be essentially single-particle width chains. In a shear flow, fluctuations with wavevector oriented along the flow direction are rotated such that they have a component in the field direction, and these field-directed fluctuations decay. Here, with field along the gradient direction, we study the evolution equation in the one spatial dimension of the vorticity direction, which is consistent with observations of the stripe formation [4, 5].
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Assuming invariance in the flow and field directions, $x$ and $z$ respectively, the evolution equation (12) for $\phi$ is simplified to

$$\frac{\partial \phi}{\partial t} = -\frac{2a^2}{9\eta_c} \frac{\partial}{\partial y} \left( f(\phi) \sigma_{yy}^p \right).$$

(13)

The total (electrostatic plus hydrodynamic) particle normal stress is written

$$\sigma_{yy}^p = 2\varepsilon_0\varepsilon_c \beta^2 E^2 \phi^2 \left[ \frac{3}{4(1-\beta\phi)^2} - \frac{\lambda_2 K_n M_n}{(\phi_{\text{max}} - \phi)^2} \right],$$

(14)

in which the entire square-bracketed term is dimensionless and the ratio of characteristic hydrodynamic and electrostatic stresses is given by the Mason number,

$$M_n = \frac{\eta_c \gamma}{2\varepsilon_0 \varepsilon_c \beta^2 E^2}.$$  

(15)

The stress scaled as $\sigma_{yy}^p / 2\varepsilon_0\varepsilon_c \beta^2 E^2 \phi^2$ is plotted as a function of $\phi$ for several $M_n$ in figure 1; in this and the following figures, data is plotted for the parameters $\beta = 1$, $\lambda_2 = 0.5$, $K_n = 0.75$, and $\phi = 0.68$. A positive stress indicates a tension while a negative stress is compressive (or dispersive). For the three smaller values of $M_n$, the two zeros of $\sigma_{yy}^p(\phi)$ are at $\phi = 0$ and some finite volume fraction: these will be shown in the steady state analysis below to correspond to the particle-free zones and stripes, respectively. The maximum $M_n$ for which two zeroes exist is $M_n \approx 0.92$.

3 Analytical results

3.1 Linear stability

Using standard methods [16], we write $\phi(y, t) = \phi_0 + \phi'(y, t)$ and linearize the $\phi$ dependence in the one-dimensional evolution equation about the mean $\phi_0$ to obtain

$$\frac{\partial \phi'}{\partial t} = -MA_2 \frac{\partial^2 \phi'}{\partial y^2},$$

(16)

in which

$$A_2 = 1 - \frac{2\lambda_2\eta_n'(\phi_0)(1-\beta\phi_0)^3}{3\eta_c\phi_0} M_n,$$

(17)

and

$$M = -\frac{a^2 \varepsilon_0 E_0^2 f(\phi_0)}{9\eta_c} \left( \frac{dc}{d\phi} + \frac{d\alpha}{d\phi} \right)_{\phi_0} - \frac{2a^2 \varepsilon_0 \varepsilon_c \beta^2 E_0^2 f(\phi_0) \phi_0}{3\eta_c(1-\beta\phi_0)^3}.$$  

(18)

Seeking solutions to (16) in the form $\phi' = e^{st}e^{iky}$ yields the relation $\frac{s}{M} = A_2 k^2$, where $M \geq 0$ is given by (18). This indicates that the temporal growth exponent $s$ is negative, meaning fluctuations are stable, for $A_2 < 0$. Solving for
Figure 1: The normal stress $\sigma^{p}_{yy}$ plotted for varying Mn; $y$ is the vorticity direction with electric field in the velocity gradient direction.

$A_{2} = 0$ from (17), using the explicit form of $\eta_{n}(\phi)$ given by (7), yields the critical Mason number defining this stability boundary:

$$M_{n_c} = \frac{3n_{max}^{2}(1 - \phi_{0}/\phi_{max})^{3}}{4\lambda_{2} K_{n} (1 - \beta \phi_{0})^{3}}.$$  \hspace{1cm} (19)

The dependence of $M_{n_c}$ upon $\phi_{0}$ is illustrated in figure 2.

### 3.2 Steady state

Any steady state must satisfy $\nabla \cdot j = \frac{\partial j_{x}}{\partial y} = 0$. Because impermeable boundaries exist at some point in a real system, the meaningful solution is $j_{y} = 0$. Returning to (11), and noting that the sedimentation function is nonzero at all $\phi$ this implies that $\frac{\partial \sigma^{p}_{yy}}{\partial y} = 0$, or $\sigma^{p}_{yy}$ is constant with respect to $y$. Here, with both shear rate and electric field constant, variation of the stress results from variation in $\phi$, and $j_{y} = (2\phi_{0}^{2}/9n_{c}) f(\phi)(d\sigma^{p}_{yy}/d\phi)(\partial \phi/\partial y)$. Consider the figure 1, we observe that for a fixed Mn there exist two stable points toward which all fractions will be driven, and these are the zeroes of $\sigma^{p}_{yy}$: the system is predicted to move toward a neutral stress state with respect to the $yy$ component of $\sigma^{p}$, in which the dispersive hydrodynamic stresses are balanced by the tensile electrostatic stresses. The particle volume fraction in the two states is
Figure 2: Critical Mason number as a function of concentration from the linear stability analysis.

found by requiring $\sigma_{yy}^P = 0$, which can be satisfied by either $\phi = 0$ or

$$\phi_{\text{stripe}} = \frac{4\beta \lambda_2 K_n M_n - 3\phi_{\text{max}} + 2\sqrt{3(1 - \beta \phi_{\text{max}})^2 \lambda_2 K_n M_n}}{4\beta^2 \lambda_2 K_n M_n - 3}.$$  \hspace{1cm} (20)

The suspension is predicted to segregate to the stripe structure, with $\phi_{\text{stripe}}$ as a function of $M_n$ plotted in figure 3. In principle, the final state should result from coalescence of the stripes into a single band, as this minimizes the electrostatic free energy, but the present modeling does not appear to contain the necessary features to describe this structural coarsening, which we expect to occur on very long timescales.

In figure 4, we combine the primary results of the linear stability and steady state analyses, i.e. the $\phi$-dependence of $M_n$ and the $M_n$ dependence of $\phi_{\text{stripe}}$, respectively, to illustrate that the model predicts a hysteresis in the structural evolution. The plot actually shows that the predicted maximum particle fraction in a suspension, max $\phi$, as a function of $M_n$ depends upon the path taken to $M_n$, as we now describe. The horizontal lines coming from the right of the figure are labeled with the initial bulk fraction, and start at conditions of uniform $\phi$. To illustrate the hysteresis, consider progressing from large to small $M_n$ (right to left) along one of these lines until the $M_n$ curve is just intersected. At this point the uniformly distributed material is predicted to be unstable, and to demix to a banded structure with $\phi_{\text{stripe}}$ found by going vertically in the figure to the upper curve. If we
Figure 3: The stripe volume fraction as a function of $M_n$ from the steady state analysis.

Now increase $M_n$, max $\phi$ remains on the curve describing the stripes: the mixture evolves to a condition having stripes of lower concentrations, rather than returning immediately to the uniformly distributed state.

4 Concluding remarks

The bulk flow model described in this work provides insight to the properties of the mixture which give rise to the nonequilibrium structures observed in ER and MR suspensions. We have focused upon the demixing phenomena, with particular attention here paid to the stripe structure with bands of particles and particle free zones alternating along the vorticity direction of a simple shear flow. These structures, described in detail in [4], have been noted to reduce the electrostatic free energy of the system relative to a uniform distribution. This work takes quite a different approach, as the bulk stress under nonequilibrium conditions is considered and the segregation is related to an instability in $\phi$ driven by the normal stresses resulting from the electrostatic stress. This is predicted by linear analysis of the coupled mass and momentum conservation equations for the particles. This prediction, illustrated in more detail in other of our work [2, 3], in conjunction with the free energy interpretation illustrates that the bulk stress and free energy are related quantities, but the stress may be either compressive or tensile and the segregated behavior observed in ER and MR fluids results from the tensile nature...
of the electrostatic stresses.

While the particle flux model [10] employed here was developed to explain particle migration leading to bulk segregation, hydrodynamic normal stresses apparently tend, in true simple-shear flow, to disperse the particles. Thus, in the steady state of an ER or MR fluid in simple shear at $Mn < Mn_c$, there exists a balance between the tensile electrostatic and compressive hydrodynamic normal stresses within the particle-rich bands. Because the material adjoining a stripe is particle-free, the net particle stress must balance to zero, i.e. the particles are in a neutral stress state.

The ability of the continuum equations to capture the segregation behavior based upon inclusion of an accepted form for the rheological contribution of the particle phase is encouraging. However, the band width is arbitrary within certain constraints on the total particle mass set by the bulk particle fraction, and in reality the bands are found to have some coarsening behavior at least in dilute systems; what we have called a steady state structure is actually an intermediate stage at which the system arrives relatively rapidly, and is subject to further slow dynamics. Among the most prominent unresolved issues is the manner in which this coarsening occurs. As the mechanism is not agreed upon, it is uncertain whether coarsening may be included within the framework of the multiphase flow model for ER and MR fluid behavior described in this work. If through future work it proves possible to model this later stage of the dynamics in continuum fashion,
it would provide a significant improvement for modeling of ER and MR fluids in applications.

References
