Pattern Formation in Flowing Electrorheological Fluids

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A two-fluid continuum model is developed to describe mass transport in electro- and magnetorheological suspensions. The particle flux is related to the field-induced stresses. Solutions of the resulting mass balance show column formation in the absence of flow, and stripe formation when a suspension is subjected simultaneously to an applied electric field and shear flow.

Electro- and magnetorheological (ER and MR) fluids are particulate suspensions whose rheological properties are dramatically altered by electric and magnetic fields, respectively [1–4]. In shear flow, applied fields can increase the apparent viscosity by several orders of magnitude. This phenomenon is now being exploited in commercial applications [5,6]. Along with the rheological phenomena, the applied fields also induce pattern formation: in quiescent suspensions, the applied field causes the formation of particulate columns oriented in the direction of the field [2,4], while, in flowing suspensions, the field causes the formation of particulate stripes oriented in the flow direction [7–11]. These patterns are intimately connected to the rheological phenomena.

Particle-level models and simulations [4,12] have been valuable for understanding the relationships between particle properties, interactions, and macroscopic behavior. Unfortunately, these techniques are limited to small numbers of particles and hence are not well suited for describing phenomena involving large numbers of particles, or predicting the behavior of a suspension throughout an entire device. These issues are particularly important in situations where the field, temperature, or particle concentration are not uniform.

In this Letter we develop and study a continuum description of ER fluids. This is accomplished by solving a conservation equation for the particle concentration, using an expression for the particle flux obtained from a momentum balance. Using this two-fluid approach, one obtains the relation for the particle flux

\[ j = \frac{2a^2}{9\eta\epsilon} f(\phi) \nabla \cdot \mathbf{\alpha}^{(p)}, \]

where \( a \) is the particle radius, \( \eta \) is the viscosity of the suspending fluid, \( f(\phi) = (1 - \phi)^{\beta} \) is a hindered mobility (also termed a sedimentation function), and \( \mathbf{\alpha}^{(p)} \) is the particle contribution to the stress. Below we obtain the electrostatic part of \( \mathbf{\alpha}^{(p)} \).

We consider the suspension to be composed of dielectric spheres immersed in a dielectric suspending fluid (conducting spheres in a weakly conducting fluid are treated analogously). The suspension structure is assumed to be isotropic and amorphous. Such a structure is reasonable for dilute quiescent or flowing suspensions prior to the application of an electric field, as well as for the initial stages of structure formation following the application of an electric field. For such a suspension, the electrostatic stress is

\[ \mathbf{\alpha}^{(E)} = \epsilon_0 \left[ \frac{\epsilon - \frac{1}{2} a_1}{\epsilon + \frac{1}{2} a_2} EE - \frac{1}{2} (\epsilon + a_2) E^2 \right] \epsilon \phi, \]

where \( \epsilon_0 = 8.8542 \times 10^{-12} \, \text{F/m} \) is the permittivity of free space, and \( \epsilon, a_1, \) and \( a_2 \) are described as follows. A self-consistent mean-field analysis in the point-dipole limit yields a suspension dielectric constant [16]

\[ \epsilon(\phi) = \epsilon_c \frac{1 + 2\beta \phi}{1 - \beta \phi}, \]

where \( \epsilon_c \) is the dielectric constant of the suspending fluid, \( \beta = (\epsilon_p - \epsilon_c)/(\epsilon_p + 2\epsilon_c) \), and \( \epsilon_p \) is the dielectric
constant of the particulate material. Assuming that the disperse and continuous phases themselves are not electrostrictive, the electrostriction coefficients of the suspension, $a_1$ and $a_2$, are [16] $a_1 = 0$ and

$$a_2(\phi) = -\frac{(\epsilon(\phi) - \epsilon_c)(\epsilon(\phi) + 2\epsilon_c)}{3\epsilon_c}.$$  \hspace{1cm} (5)

Taking the divergence of Eq. (3) and utilizing Maxwell’s equations $\nabla \cdot D = \rho^e (\rho^o$ is the free charge density) and $\nabla \times E = 0$ gives

$$\nabla \cdot \sigma^{(E)} = \rho^e E - \frac{1}{\tau} \epsilon_0 E^2 \nabla \epsilon - \frac{1}{\tau} \epsilon_0 a_2 E^2.$$  \hspace{1cm} (6)

The terms on the right side are body forces. For the present analysis, $\rho^e = 0$, and the remaining terms are body forces exerted on the particles; thus $\nabla \cdot \sigma^{(E)}$ contains only the influence of particle contributions to the stress (i.e., $\nabla \cdot \sigma^{(E)} = \nabla \cdot \sigma^{(p,E)}$).

The particle stress will also contain a hydrodynamic contribution [13]. This contribution is neglected here in the interest of space and to illustrate the effects of the electrostatic driving force. Hydrodynamic contributions will be addressed in a future publication.

Combining Eqs. (1), (2), and (6) yields a conservation equation for the evolution of the volume fraction when a field is applied to a flowing suspension. Here we consider only simple shear flow $[\mathbf{u} = (\dot{\gamma} z, 0, 0)]$ and limit attention to small fluctuations in concentration. For an applied field $E = (0, 0, E_0)$, the conservation equation becomes

$$\frac{\partial \phi'}{\partial t} + \dot{\gamma} z \frac{\partial \phi'}{\partial x} = -M \left( \frac{\partial^2 \phi'}{\partial x^2} + \frac{\partial^2 \phi'}{\partial y^2} - \kappa \frac{\partial^2 \phi'}{\partial z^2} \right),$$  \hspace{1cm} (7)

where $\phi'(x,t) = \phi(x,t) - \phi_0$ is the deviation of the volume fraction from the average value $\phi_0$.

$$M = \frac{a_2^2 \epsilon_0 E_0^2 \phi_0}{9 \eta_c} \left( \frac{d\epsilon}{d\phi} + \frac{da_2}{d\phi} \right) \phi_0,$$  \hspace{1cm} (8)

$$= \frac{2a_2^2 \epsilon_0 \epsilon_c \beta^2 E_0^2 f(\phi_0) \phi_0}{3 \eta_c (1 - \beta \phi_0)^3},$$  \hspace{1cm} (9)

and $\kappa = 2(1 - \beta \phi_0)/(1 + 2\beta \phi_0) > 0$. Equation (7) is similar to a convection-diffusion equation, with anisotropic diffusion and a negative diffusivity in the $x$ and $y$ directions. However, the particle phase motion has nothing to do with Brownian diffusion, but rather is caused by electrostatic forces. In fact, for typical ER suspensions, Brownian motion is negligible: for $a = 1 \times 10^{-5}$ m, $T = 298$ K, and $\eta_c = 0.1$ Pa s, the Brownian diffusivity is $D_0 = kT/6\pi \eta_c a = 2.2 \times 10^{-10}$ m$^2$/s, whereas $M = 1.1 \times 10^{-13}$ m$^2$/s for $\epsilon_c = 2$, $\beta = 1$, $E_0 = 1 \times 10^6$ V/m, and $\phi_0 = 0.1$.

The negative apparent diffusivity is reminiscent of spinodal decomposition in phase separating systems [17]. Indeed, a uniform suspension in an electrostatic field is thermodynamically unstable. Consider the electrostatic contribution to the free energy, $\mathcal{F}^E = -\epsilon_0 \epsilon(\phi) E^2/2$ [18]. Since $\partial^2 \epsilon/\partial \phi^2 > 0$ [e.g., Eq. (4)], $\partial^2 \mathcal{F}^E/\partial \phi^2 < 0$, which implies that the free energy can be reduced by separating the system into dilute and concentrated phases.

Below we show that Eq. (7) predicts that the application of an electric field to a uniform suspension indeed causes the formation of more concentrated “phases.” We show that for a quiescent suspension the field induces the formation of particle-rich columns oriented in the direction of the applied field; for sheared suspensions, the field induces the formation of particle-rich stripes oriented in the flow direction. These structures are consistent with those commonly observed experimentally.

Consider first the application of an electric field to a quiescent suspension. The electrodes are located at $z = 0$ and $L$. We seek a solution of Eq. (7) of the form

$$\phi'(x,t) = f(z) e^{i(k_x x + k_y y)} e^{\nu t},$$  \hspace{1cm} (10)

which represents fluctuations with sinusoidal variation in the $x$ and $y$ directions. The $z$ dependence is yet to be determined. The growth rate of the fluctuations is represented by $\nu$. The full solution to Eq. (7) would be composed of an infinite series of many terms of the form of Eq. (10). Substitution of Eq. (10) into Eq. (7) yields

$$\kappa f''(z) + (k_x^2 + k_y^2 - s/M) f(z) = 0.$$  \hspace{1cm} (11)

Using the no-flux boundary conditions $f'(z) = 0$ at $z = 0$ and $z = L$ yields

$$f(z) = \cos \pi z / L \quad n = 0, 1, \ldots,$$  \hspace{1cm} (12)

and

$$sL^2 / M = (k_x L)^2 + (k_y L)^2 - \kappa n^2 \pi^2.$$  \hspace{1cm} (13)

Fluctuations will grow for all $s > 0$; the fastest growing fluctuations (for any given $k_x$ and $k_y$) will occur for $n = 0$ and grow with rate $s_{\text{max}}/M = k_x^2 + k_y^2 > 0$. This result has several implications for the resulting suspension structures. First, since the fastest growing fluctuations occur for $n = 0$, the resulting concentration profile should not depend on $z$. Second, the maximum growth rate increases with $k_x$ and $k_y$; in fact, the $\nu \to \infty$ as $k_x$ and $k_y \to \infty$. This implies that the fastest growing fluctuations will be thin structures. Third, the growth rate is symmetric with respect to $k_x$ and $k_y$, suggesting that if the initial fluctuations are random and isotropic, the resulting structures will be cylindrical columns; i.e., columns of particles oriented in the $z$ direction, as commonly observed (a nonlinear analysis is required to determine the structures resulting from arbitrary initial conditions [19]).

This continuum analysis will break down at small length scales. The fastest growing wave numbers are certainly limited to at most roughly the inverse of the particle diameter. We also expect nonlocal polarization (i.e., “surface
tension” [20]) to affect high $|k|$ growth rates and thus the thickness of the resulting structures. However, these features will not alter the conclusion that columnar structures are produced.

A finite difference numerical solution of Eq. (7) is illustrated in Fig. 1. No-flux boundary conditions were applied at the electrodes; periodic boundary conditions were applied at $x, y = 0, L$ to represent a system of infinite extent in the $x$ and $y$ directions. The initial concentration consisted of small, random fluctuations about an average volume fraction $\phi_0 = 0.1$. The evolution of the concentration fluctuations is illustrated in Fig. 1, where positive fluctuations are indicated by dark cubes, and negative fluctuations are indicated by light cubes. The initial distribution and a distribution at a later time ($tM/L^2 = 0.05$) are shown. As time progresses, the concentration fluctuations form columnar structures, which are similar to the fibrous aggregates commonly observed in ER and MR suspensions.

The mechanism of column formation is revealed by considering the change in electrostatic free energy associated with concentration fluctuations, given by $\delta F^E = -\epsilon_0 f \left[ \delta \epsilon(\phi) E \cdot dV / 2 \right]$ [18], where $\delta \epsilon$ is the fluctuation in dielectric constant caused by the concentration fluctuation, $E$ is the electric field, and $E_0$ is the electric field prior to the fluctuation. Consider first a parallel concentration fluctuation of the form $\phi(x) = \phi_0 + A_0 \cos \beta_{||} x$ with $A_0 \ll 1$. In this case the field is unaltered, and the change in the electrostatic free energy is $\delta F^E = -\epsilon_0 E_0^2 (d^2 \epsilon / d \phi^2) A_0^2 V / 8 < 0$, implying that such fluctuations are unstable and will continue to grow. Next consider perpendicular fluctuations of the form $\phi(z) = \phi_0 + A_{\perp} \cos \beta_{\perp} z$. In this case the field is altered by the fluctuation, and the resulting change in free energy is $\delta F^E = +\epsilon_0 E_0^2 (d^2 \epsilon / d \phi^2) A_{\perp}^2 V / 4 > 0$, implying that such fluctuations are stable and will decay. Columnar fluctuations parallel to the applied field, which reduce the system free energy, will thus be favored.

Now we consider the evolution of the concentration profile when an electric field is applied to a uniform suspension under shear flow. Again we seek a solution of the form of Eq. (10), which upon substitution into Eq. (7) yields the ordinary differential equation for $f(z)$,

$$\kappa f''(z) + (k_x^2 + k_y^2 - s/M - i\gamma k_z z/M)f = 0. \quad (14)$$

Solution of this differential equation with boundary conditions $f'(z) = 0$ at $z = 0, L$ provides the eigenvalue $s$ as a function of the wave numbers $k_x$ and $k_y$. Again, positive values of $s(k_x, k_y)$ indicate unstable or growing fluctuations, while negative values of $s(k_x, k_y)$ indicate stable or decaying fluctuations.

Equation (14) was solved numerically to determine $s(k_x, k_y)$. The results for $\gamma L^2/M = 10^3$ and $10^4$ are illustrated in Fig. 2, where contour plots of $sL^2/M$ as a function of $k_x$ and $k_y$ are presented. The $s = 0$ contours in Fig. 2(a) and 2(b) represent the stability boundaries. The $s = 0$ contours are roughly semicircles, which grow with increasing shear rate. Thus certain fluctuations tend to be stabilized by shear flow.

As discussed above the present model breaks down at large wave numbers. This arises from neglecting the physical dimensions of the particles, as well as neglecting short range interactions between particles. Including these features will tend to stabilize large $|k|$ fluctuations. Combining this information with that presented in Fig. 2 implies that, at sufficiently large shear rates, the only unstable fluctuations will be those with small $k_x$ and all $k_y$ below the large $|k|$ cutoff (i.e., the region between the $s = 0$ contour and the $k_y$ axis). Within this region, the most unstable fluctuations will be those with $k_x = 0$ and nonzero $k_y$, i.e., the dominant structure formed by applying an electric field to a flowing suspension is predicted to be stripes of particle-rich regions oriented in the flow direction, as observed experimentally [7–11].

Equation (7) has also been solved by the finite difference method to illustrate the stripe formation process. No flux boundary conditions were applied at the electrodes; periodic boundary conditions were applied at $x, y = 0, L$. The evolution of concentration fluctuations is illustrated in Fig. 3 for $\gamma L^2/M = 10^4$ and $\phi_0 = 0.1$. The initially random fluctuations eventually redistribute into stripes oriented in the flow direction, as described above. For the

![FIG. 1. (a) Initial structure and (b) structure after the application of an electric field to a quiescent suspension ($tM/L^2 = 0.05$). Dark: positive concentration fluctuation; light: negative concentration fluctuation.](image)

![FIG. 2. Contour plots of $sL^2/M$ as a function of $k_x L$ and $k_y L$ for (a) $\gamma L^2/M = 10^3$ and (b) $\gamma L^2/M = 10^4$. Stable and unstable regions are denoted by S and U, respectively.](image)
parameter values listed following Eq. (9) along with $L = 10^{-3}$ m, $\gamma L^3/M = 10^4$ corresponds to a shear rate of $\dot{\gamma} = 10$ s$^{-1}$.

The mechanism of stripe formation in shear flow is related to the mechanism of column formation in quiescent suspensions. Fluctuations grow when parallel to the field and decay when perpendicular to the field. In shear flow, fluctuations parallel to the applied field will be rotated (and stretched) toward the flow direction and thus will become stabilized, as illustrated in Fig. 4. The result is a uniform concentration in the plane of shear. However, since fluctuations in the vorticity direction [i.e., $k = (0, k_y, 0)$] are unaffected by shear and will continue to grow, the resulting structure will be sheets of higher concentration in the plane of shear (i.e., stripes oriented in the flow direction).

To summarize, a two-fluid continuum model for mass transport in ER suspensions has been presented. The particle flux is related to the divergence of the particle concentration contribution to the stress, which in turn is related to the suspension dielectric and electrostrictive properties. Solutions of the resulting particle conservation equation capture common observations: column formation in quiescent suspensions and stripe formation in sheared suspensions. Column formation arises because only these structures produce a decrease in the free energy of polarization. Stripe formation in shear flow arises because the flow stabilizes fluctuations in the plane of shear.

Future work will involve refining and improving this continuum approach. Hydrodynamic interactions must be included to probe the behavior when the concentration within the stripes gets large. Nonlocal polarization contributions (i.e., surface tension [20]) to the stress and flux should be included to correctly capture the large $|k|$ behavior. Effects of particle interactions and structural anisotropy should be included to completely capture the structure evolution process in ER and MR suspensions.

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