Curvilinear flows of noncolloidal suspensions: 
The role of normal stresses

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Synopsis

The role of normal stresses in causing particle migration and macroscopic spatial variation of the particle volume fraction $\phi$ in a mixture of rigid neutrally buoyant spherical particles suspended in Newtonian fluid is examined for curvilinear shear flows. The problem is studied for monodisperse noncolloidal Stokes-flow suspensions, i.e., for conditions of low-Reynolds-number flow and infinite Péclet number, $\text{Pe} = O(\eta \gamma a^3/kT)$, where $\eta$ is the suspending fluid viscosity, $\gamma$ is the shear rate, $a$ is the particle radius, and $kT$ is the thermal energy. Wide-gap Couette, parallel-plate torsional, and cone-and-plate torsional flows are studied. The entire $\phi$ dependence of the compressive shear-induced normal stresses is captured by a “normal stress viscosity” $\eta_n(\phi)$, which vanishes as $\phi^2 \to a$ and diverges at maximum packing in the same fashion as does the shear viscosity $\eta_s(\phi)$. Anisotropy of the normal stresses arising from the presence of the particles is modeled as independent of $\phi$, so that ratios of any two particle contributions to the bulk normal stress components are constants, $\Sigma^p_{22}/\Sigma^p_{11} = \lambda_2$ and $\Sigma^p_{33}/\Sigma^p_{11} = \lambda_3$; the standard convention of $(1,2,3)$ denoting the (flow, gradient, vorticity) directions is used so that, for example, $\Sigma^p_{11}$ is the normal component of the particle stress $\Sigma_p$ in the flow direction. Predictions for the steady and unsteady flows are presented to demonstrate the influence of variation of the normal stress anisotropy parameters $\lambda_2$ and $\lambda_3$, the rheological functions $\eta_s$ and $\eta_n$, and the sedimentation hindrance function used to represent the resistance to relative motions of the phases during migration. Comparison with available experimental data shows that a single set of parameters for the rheological model is able to describe all qualitative features of the observed migrations in the flows considered. © 1999 The Society of Rheology.

I. INTRODUCTION

This work addresses modeling of curvilinear flows of noncolloidal suspensions. We seek to determine the influence of normal stresses upon the particle fraction and velocity fields for shear flows. We consider a set of curvilinear flows which are locally well approximated as simple shear flows: the wide-gap Couette, parallel-plate torsional, and small angle cone-and-plate torsional flows. The goal in our study of these flows is to demonstrate that shear-induced normal stresses in noncolloidal suspensions will generally cause variation in the particle fraction in shear flows, and thus that normal stress rheology must be considered in flow analyses for these mixtures. Specifically, this work shows that shear-induced migrations which have been observed in several curvilinear shear flows of...
concentrated monodisperse suspensions [Phillips et al. (1992); Abbott et al. (1991); Chow et al. (1994); Chow et al. (1995)] can be explained by inclusion of shear-induced normal stresses in the rheological model.

Shear-induced migration has been observed in pressure-driven rectilinear flows [Koh et al. (1994); Nott and Brady (1994)] as well as curvilinear flow of low-Reynolds-number suspensions. The phenomenon was recognized and described by Leighton and Acrivos (1987), who concluded that a shear-induced migration was necessary to explain a number of observations in suspension flow in a Couette viscometer. Subsequent studies of monodisperse suspension flows relevant to the present study include the circular Couette and viscometric flows in cone-and-plate and parallel-plate devices. In wide-gap Couette flow, in which the gap size relative to the radius of the inner of two concentric cylinders is large enough to yield $O(1)$ variation of the shear rate, particles are found to migrate radially outward [Phillips et al. (1992); Abbott et al. (1991)]; in parallel-plate torsional flow [Chapman (1990); Chow et al. (1994)], particles are found to undergo weak migration if any; in truncated cone-and-plate torsional flow, the particles are found to migrate radially outward, i.e., away from the apex of the cone [Chow et al. (1995)].

Modeling of the migration phenomenon has primarily taken two forms. The approach used here is based upon analysis of the coupling of particle mass and momentum conservation, with particle migration argued to result from the requirements of the momentum balance when normal forces are exerted by the particle phase [Nott and Brady (1994); Mills and Snabre (1995)]. Analysis of these balances for Stokes flow, presented in Sec. II A, leads to an expression for the particle migration flux $j_p$, of the form $j_p \sim \nabla \Sigma_p$, where $\Sigma_p$ is the particle contribution to the bulk stress. Phenomenological arguments, expressed in the original work of Leighton and Acrivos (1987), hold that the migration flux is proportional to the gradient in the shear rate, $\dot{\gamma}$, i.e., $j_p \sim -\nabla \dot{\gamma}$.

There is obvious similarity in the expressions for $j_p$ in the two approaches, and both approaches have been successful in prediction of migrations in pressure-driven and wide-gap Couette flows. However, the nonmigration observed in parallel-plate flow and the outward migration observed in cone-and-plate torsional flow are predicted qualitatively inaccurately by the phenomenological model, which must be augmented with further modeling of the role of curved streamlines to capture the observed behavior. Specifically, the basic phenomenological modeling of $j_p \sim -\nabla \dot{\gamma}$ predicts an inward migration in the parallel-plate flow as the shear rate is linear in distance from the center of rotation; the same model predicts, of course, no migration in the constant $\dot{\gamma}$ cone-and-plate flow. These predictions conflict with the experimental observations noted above. With regard to the particle kinematics, the migration predicted by the phenomenological approach for the parallel-plate flow is radial but is driven by a radial variation in $\dot{\gamma}_r$. This represents a significant difference from the tube and wide-gap Couette geometries, in which radial migration is driven by nonzero $\partial \dot{\gamma}_r / \partial r$ and $\partial \dot{\gamma}_r / \partial r$, respectively; because of this, Phillips et al. (1992) restricted their use of the phenomenological, or diffusive flux, model to shearing flows in which particles migrated across shear planes. This restriction has been relaxed by others to consider use of this modeling approach in more general flows [see, for example, Zhang and Acrivos (1994), Subia et al. (1998)], but the basic difficulties of the approach in even the well-defined cone-and-plate and parallel-plate flows prompts a more general approach to the analysis of the impact of rheology upon particle fraction in suspension flows.

The essential feature of suspension rheology which is not captured by the phenomenological approach is the presence of normal stresses. Because normal stress differences influence the dynamics in curvilinear flows [Bird et al. (1987)], these flows provide an
important test of the basic structure of the rheological modeling employed in our approach to suspension-flow modeling. The earlier work using the approach employed here, termed the “suspension balance model” [Nott and Brady (1994); Morris and Brady (1998)], recognized the importance of viscously generated normal stresses (i.e., normal stresses linear in the shear rate) and modeled the suspension pressure, but did not address flows where normal stress anisotropy was dynamically relevant. Anisotropy of normal stresses implies that the first and second normal stress differences: \( N_1 = \Sigma_{11} - \Sigma_{22} \) and \( N_2 = \Sigma_{22} - \Sigma_{33} \), respectively, are nonzero. This work will demonstrate that the presence of particle-fraction dependent and anisotropic suspension normal stresses is sufficient to explain observed particle migrations in curvilinear flows of monodisperse suspensions. A basic and direct conclusion of the analysis presented in Sec. II is that migration should be expected to be a feature of dispersed two-phase flows in general. For the low-Reynolds-number suspensions addressed here, this conclusion prompts careful consideration of the bulk mixture rheology, and this work presents a simple model of the full rheology of noncolloidal suspensions in steady shear flows.

Although the rheological model used here is based upon assumed functional forms, with parameters determined from comparison with experimental and simulational data, the relationship of rheology to migration is rigorous. Analysis is presented in Sec. II to support this statement. All rheological information necessary to obtain predictions made in this study may, in principle, be measured in homogeneous shear-flow experiments. That Stokes-flow suspensions of small spherical particles in Newtonian fluid exert shear-induced normal stresses has been shown experimentally by Gadala-Maria (1979) [see also Leighton and Rampall (1993)] and Laun (1994), as well as in simulations by Phung et al. (1996) and Phung (1993). The magnitude and form of the normal stresses depends upon the particle volume fraction and the relative strength of shear-driven to thermal motions at the particle scale (a Pécellet number), as well as the form and strength of interparticle forces [Phung et al. (1996); Brady and Morris (1997); Yurkovetsky (1997)]. While experimental data on the normal stress differences in concentrated suspensions exists [Gadala-Maria (1979); Laun (1994)], the data are ambiguous (see the discussion in Sec. III C); no experimental data on the suspension pressure given by \( \Pi = -\frac{1}{2} \text{tr} \Sigma_p \) [Jeffrey et al. (1993)] is available, to our knowledge. Stokesian Dynamics simulations therefore provide a valuable guide to the sign and relative magnitude of \( N_1 \) and \( N_2 \).

In Sec. II, the governing equations for suspension flow are outlined, and we introduce a rheological model for steady shear of noncolloidal suspensions which includes anisotropic normal stresses with a net isotropic contribution (suspension pressure). In Sec. III, predictions for curvilinear flows of a noncolloidal suspension are presented with comparison to both experimental findings and predictions from prior modeling. A discussion and concluding remarks are found in Sec. IV.

II. GOVERNING EQUATIONS AND RHEOLOGICAL MODEL

We consider a suspension of rigid particles immersed in Newtonian fluid of viscosity \( \eta \) and density \( \rho \). Certain results are applicable to general particle geometries and to polydisperse systems, but this work will specifically address only suspensions of mono-
disperse spherical particles of radius $a$. The mixture behavior will be considered in shearing flows at vanishing Reynolds number and infinite Péclet number

$$\text{Re} = \frac{\rho \dot{\gamma} a^2}{\eta} \to 0,$$

and

$$\text{Pe} = \frac{3 \pi \eta \dot{\gamma} a^3}{kT} \to \infty,$$

where $\dot{\gamma}$ is the shear rate of a simple-shear flow and $kT$ is the thermal energy. The particle volume fraction is given by $\phi = \frac{4}{3} \pi a^3 n / 3$, where $n$ is the particle number density.

We assume that microstructural asymmetry necessary to generate anisotropic normal stresses exists at all volume fractions, $0 < \phi < \phi_{\text{max}}$. In so doing, we implicitly assume a short-range interparticle repulsion or roughness whose influence is rate independent for the conditions studied. The suspension is thus a near-hard-sphere system with nonvanishing anisotropy at large shear rate, a system for which the limiting microstructure in the noncolloidal limit of $\text{Pe} \to \infty$ has been studied analytically by Brady and Morris (1997) and numerically by Morris and Brickman (1998). These studies have shown that, from a rheological perspective, the influence of a short-ranged interparticle force or roughness [Rampall et al. (1997)] which maintains a finite surface separation becomes “saturated” at large Pe, meaning that the rheology ceases to depend upon the shear rate aside from the basic scaling of all stresses as $\eta \dot{\gamma}$. Results of these studies of interest here are that such suspensions tend toward a shear-rate independent structure, with normal and shear stresses linear in $\eta \dot{\gamma}$ for $\text{Pe} \to \infty$.

A. Mass and momentum conservation

1. Particle mass conservation

We begin by considering the mass conservation equation

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0, \tag{1}$$

where $\rho(x,t)$ is the mass density and $\mathbf{u}(x,t)$ is the velocity. The equation is valid pointwise in both components of the mixture. The mass conservation equation for the particles is obtained, formally, by an averaging procedure [Drew and Lahey (1993)] which involves multiplying Eq. (1) by the phase indicator function, i.e.,

$$\chi = 1 \text{ in particles, } \chi = 0 \text{ in fluid},$$

multiplying by the $N$-particle configurational probability $P_{\chi}(x_N)$, and integrating over configurations $x_N$. The resulting particle-phase conservation equation is

$$\frac{\partial \phi}{\partial t} + \nabla \cdot (\mathbf{U} \phi) = 0, \tag{2}$$

where $\mathbf{U}$ is the local average velocity of the particle phase. The constant particle density $\rho_p$ has been divided out of the equation.
2. Particle momentum conservation

The basic development in this section is essentially the same as that of Nott and Brady (1994) and Morris and Brady (1998). To determine the particle flux, \( \mathbf{j} = \phi \mathbf{U} \), appearing in Eq. (2) requires consideration of the pointwise-valid momentum conservation equation

\[
\rho \left( \frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = \nabla \cdot \sigma + \mathbf{b},
\]

(3)

where \( \sigma(\mathbf{x},t) \) denotes the microscopic stress tensor and \( \mathbf{b} \) represents both body and interparticle forces. Averaging by the procedure used to obtain Eq. (2) from Eq. (1) yields

\[
\rho_p \frac{\partial \mathbf{U}}{\partial t} + \mathbf{U} \cdot \nabla (\phi \mathbf{U}) + \rho_p \mathbf{u} \cdot (\phi (\mathbf{u}' \mathbf{u}')) = \nabla \cdot (\phi (\sigma) + n(\mathbf{F}^H)_p + (\mathbf{b})_p - \nabla \cdot \langle \mathbf{x}' \mathbf{b}' \rangle_p,
\]

(4)

where \( \langle \cdot \rangle_p \) is a particle-phase average, and a primed quantity denotes a fluctuation from the average. The suspension average of a quantity is denoted by \( \langle \cdot \rangle \). The hydrodynamic or drag force on a particle is \( \mathbf{F}^H \). The drag force arises in the averaging from the fact that \( \chi \) is discontinuous at the phase boundary, and thus has a singular gradient which upon integration over configurations captures the interphase momentum transfer represented by \( \mathbf{F}^H \). The term \( -\langle \mathbf{x}' \mathbf{b}' \rangle_p \) is a stress resulting from the moment of interparticle forces and is familiar from kinetic theory as an \( xF \) stress; this term would exist for particles in vacuum and cannot be derived by continuum methods.

Equation (4) may be written

\[
\rho_p \frac{D_p(\phi \mathbf{U})}{Dt} = \nabla \cdot \Sigma_p + n(\mathbf{F}^H)_p + (\mathbf{b})_p,
\]

(5)

with the particle-phase convective derivative defined

\[
\frac{D_p}{Dt} = \frac{\partial}{\partial t} + \mathbf{U} \cdot \nabla,
\]

and the particle stress given by

\[
\Sigma_p = \phi [-\rho_p(\mathbf{u}' \mathbf{u}')_p + \langle \sigma \rangle_p - \langle \mathbf{x}' \mathbf{b}' \rangle_p].
\]

(6)

The term \( -\rho_p(\mathbf{u}' \mathbf{u}')_p \) is the particulate contribution to the inertial Reynolds stress and is negligible for the \( Re = 0 \) condition considered here.

3. Suspension mass and momentum balances

Averaging Eq. (3) over the entire suspension yields

\[
\langle \rho \rangle \frac{D(\mathbf{u})}{Dt} = \nabla \cdot \left[ -\langle \rho \mathbf{u}' \mathbf{u}' \rangle + \langle \sigma \rangle - \langle \mathbf{x}' \mathbf{b}' \rangle \right] + \langle \mathbf{b} \rangle = \nabla \cdot \Sigma + \langle \mathbf{b} \rangle.
\]

(7)

We consider low Reynolds number and a neutrally buoyant mixture, with both particles and fluid incompressible, and assume only gravitational body forces. For this situation, the body forces have no influence, and the bulk suspension satisfies the respective mass and momentum balances of
\[ \nabla \cdot \langle \mathbf{u} \rangle = 0, \]
and
\[ \nabla \cdot \mathbf{\Sigma} = 0. \]

4. Particle mass-momentum conservation coupling

In dimensionless form, the left hand side (LHS) of the particle-momentum equation (5) and the first term on the right hand side (RHS) of Eq. (6) scale with the particle density-based Reynolds number, \( \text{Re}_p = \rho_p r^2 / \eta \), and are discarded for Stokes flow. For neutrally-buoyant particles, we arrive at the following reduction of Eq. (5):

\[ 0 = \nabla \cdot \mathbf{\Sigma}_p - \frac{9 \eta}{2a^2} \phi f^{-1}(\phi)(\mathbf{U} - \langle \mathbf{u} \rangle). \]

The sedimentation hindrance function \( f \) relates the sedimentation rate of a homogeneous suspension of spheres at volume fraction \( \phi \) to the isolated Stokes settling velocity, i.e., \( f(\phi) = U_{\text{sed}}(\phi)/U_{\text{sed}}(0) \). A standard form proposed by Richardson and Zaki (1954) [see also Davis and Acrivos (1985)] is used in this work, with \( \alpha = 4 \) used in fitting other parameters; the influence of \( \alpha \) upon migration rate is considered in Sec. III D. Note that \( f(\phi_{\text{max}}) \) does not vanish.

The expression, Eq. (9), implies that the particle flux is given by

\[ \mathbf{j} = \phi \mathbf{U} = \phi \langle \mathbf{u} \rangle + \frac{2a^2}{9 \eta} f(\phi) \nabla \cdot \mathbf{\Sigma}_p. \]

It is convenient to describe the flux of particles relative to the mean motion of the suspension, for which purpose we define the migration flux

\[ \mathbf{j}_\perp = \phi \mathbf{U} - \phi \langle \mathbf{u} \rangle = \frac{2a^2}{9 \eta} f(\phi) \nabla \cdot \mathbf{\Sigma}_p. \]

The subscripted \( \perp \) indicates that primary interest is in the cross-stream ("perpendicular") component of the flux. In terms of \( \mathbf{j}_\perp \), the particle mass conservation equation (4) becomes

\[ \frac{\partial \phi}{\partial t} + \langle \mathbf{u} \rangle \cdot \nabla \phi = -\nabla \cdot \mathbf{j}_\perp \]

\[ = -\frac{2a^2}{9 \eta} \nabla \cdot [f(\phi)\nabla \cdot \mathbf{\Sigma}_p]. \]

The expression for the particle flux—and hence the rate of segregation of a mixture—is independent of the fluid viscosity, because \( \eta \) will set the scale of \( \mathbf{\Sigma}_p \) for a given shear rate. The only independent rate in the description of the motion for neutrally buoyant noncolloidal particles is the shear rate of the flow, and time is properly made dimensionless with the inverse of a characteristic shear rate. When \( \nabla \cdot \mathbf{\Sigma}_p = 0 \), either Eq. (12) or (14) indicates that the particles are advected with the bulk motion and migration ceases.
Equation (14) indicates a coupling of the particle mass (or volume) and momentum conservation due to the contribution of suspended particles to the bulk stress. A simple example illustrates that it is normal stress that is responsible for the cross-stream flux of particles. Consider pressure-driven flow of a neutrally buoyant suspension in a channel with invariance in the flow direction \( x \) and average velocity \( u_x(y) \). The particle \( y \) momentum balance, Eq. (9), at steady state for this set of conditions reduces to \( d\Sigma_{yy}/dy = 0 \), or \( \Sigma_{yy} \) is constant with respect to \( y \). The shear rate \( \dot{\gamma} \) varies from a minimum at the channel centerline (\( y = 0 \)) to a maximum at either wall in this flow. As a result, the normal stresses, which scale viscously and may be written \( \Sigma_{ii}(\phi, \dot{\gamma}) = \eta \dot{\gamma} \text{fn}(\phi) \) [\text{fn}(\phi) implies a function of \( \phi \) only; modeling of the particle fraction dependence follows in Sec. II B] with \( i = 1, 2 \) or \( 3 \) (no implied summation on \( i \) here), will vary with respect to \( y \) for uniform \( \phi \). The particles must therefore migrate to a state of nonuniform \( \phi(y) \) to satisfy the constant \( \Sigma_{yy} \) requirement of the \( y \) momentum balance. The net migration toward the channel center line in this flow is well known [Koh et al. (1994); Nott and Brady (1994); Lyon and Leal (1997)]. The basic concept that normal stress drives migration of stress-generating material in fluids has been invoked to explain migration of polymer chains in solution [Doi (1990); Beris and Mavrantzas (1994); MacDonald and Muller (1996)].

This example leads to the expectation of migration but also shows that the balance at the center line, where the bulk average shear rate vanishes, presents a difficulty in the modeling. For \( \text{Pe}^{-1} = 0 \), this can only be resolved within this framework by inclusion of a nonlocal normal stress. To remedy this, stress modeling utilizing the suspension temperature, \( T = (\mathbf{u}' \cdot \mathbf{u}')_T \) was used by Nott and Brady (1994) and Morris and Brady (1998); an alternative nonlocal normal stress approach based upon formation of a particle network was proposed by Mills and Snabre (1995) for elimination of this same problem at the channel center line. Here, we do not employ the suspension temperature and apply a purely local stress law in most of the work. We will, however, introduce in Sec. III A a nonlocal stress description to the framework proposed here.

### B. Noncolloidal suspension rheology

The previous section presented a rheological basis for particle migrations. This approach provides a conceptual simplification in the description of suspension flows, as the driving force for migration is apparent from the governing equations without \textit{ad hoc} terms or introduction of additional (and, in principle, unnecessary) dependent field variables. As a result, the bulk motions can be predicted from the correct suspension rheology.

Experimental measurements of normal stresses in suspensions are, however, incomplete even for the simplest system of a monodisperse suspension of noncolloidal spheres. As noted in the introduction, Stokesian Dynamics simulations [Phung et al. (1996) and Phung (1993) at large finite Pe with no repulsive forces; Yurkovetsky (1997) at infinite Pe with short-ranged repulsive forces] indicate that both \( N_1 \) and \( N_2 \) are negative, and \( |N_2| > |N_1| \); the results have too much scatter to make more definitive statements. Yurkovetsky found, using the hydrodynamic functions for the suspension pressure of Jeffrey et al. (1993) that \( \Pi \approx |N_2| \) from his simulations. Taken together, the results of Yurkovetsky indicate that all components of the normal stress are negative, meaning the stress is compressive, in a concentrated (0.45 < \( \phi < 0.52 \)) near-hard-sphere suspension, with \( |\Sigma_{33}| < |\Sigma_{22}| < |\Sigma_{11}| \). Microstructural and structure–property analyses by Brady and Morris (1997) clearly demonstrate that all stress components should be compressive and scale as \( \eta \dot{\gamma} \) under the conditions of interest here, and also suggest that \( |N_1| \)
should be small relative to $|N_2|$ because $|N_1|$ vanishes in the planar extensional flow associated with simple shear.

It is clear that anisotropic compressive normal stresses are present in sheared suspensions, but there remains much uncertainty regarding their precise form. Hence, it is beneficial for the purpose of providing insight to the influence of normal stresses on suspension behavior to use a rheological model which is complete in the sense of including normal stresses, yet sufficiently simple to allow for analytical steady-state predictions. To this end, the particle contribution to the stress is written

$$\Sigma_p = -\eta \dot{\gamma} Q(\phi) + 2 \eta \eta_p(\phi) E,$$

(15)

where $\eta_p$ is the particle contribution to the shear viscosity made dimensionless with $\eta$, $E$ is the local bulk suspension rate of strain, $\dot{\gamma} = (2E:E)^{1/2}$, and the normal stresses are specified by the dimensionless $Q$ dependent material property tensor $Q$. We take $Q$ as

$$Q(\phi) = \eta_n(\phi) \begin{pmatrix} 1 & 0 & 0 \\ 0 & \lambda_2 & 0 \\ 0 & 0 & \lambda_3 \end{pmatrix} = \eta_n(\phi) \hat{Q}.$$

(16)

The function $\eta_n(\phi)$ is the ‘‘normal stress viscosity’’ made dimensionless with $\eta$, and is given by the ratio $\Sigma_{11}/(-\eta \dot{\gamma})$. This form mirrors the definition of the conventional suspension viscosity in simple-shear flow, $\eta_s(\phi) = \Sigma_{12}/(\eta \dot{\gamma})$, and use of the term viscosity implies a viscous normal stress law, i.e., a form linear in $\dot{\gamma}$. This is appropriate because all stresses are viscously generated under the stated conditions. The material response function is simplified here from the full $Q$ to a single scalar function $\eta_n(\phi)$ and a constant tensor $\hat{Q}$, which describes the anisotropy of the normal stresses. The anisotropy parameters $\lambda_2$ and $\lambda_3$ along with the $\phi$ dependence of $\eta_n$ will be considered below.

The numerical subscripts in Eq. (16) correspond to the directions in a viscometric shear flow [Bird et al. (1987)] with 1, 2, and 3 the flow, gradient, and vorticity directions, respectively. The coefficients $\lambda_2$ and $\lambda_3$ are assumed to be positive. Considering Eq. (15), this implies all normal stresses are negative, and the shear flow results in a positive suspension pressure, $\Pi = -\frac{1}{\eta} \text{tr} \Sigma_p$. The normal stress differences and suspension pressure are given in terms of this modeling by

$$N_1 = -\eta \eta_n \dot{\gamma}(1-\lambda_2), \quad N_2 = -\eta \eta_n \dot{\gamma}(\lambda_2-\lambda_3),$$

and

$$\Pi = \eta \eta_n \dot{\gamma} \left( \frac{1+\lambda_2+\lambda_3}{3} \right).$$

The modeled normal and shear viscosities of the bulk suspension we employ are

$$\eta_s = 1 + 2.5 \phi_{\text{max}}(1-\bar{\phi})^{-1} + K_s \bar{\phi}^2(1-\bar{\phi})^{-2},$$

(18)

and

$$\eta_n = K_n \bar{\phi}^2(1-\bar{\phi})^{-2},$$

(19)

respectively, where $\bar{\phi} = \phi/\phi_{\text{max}}$. We note that $\eta_s = 1 + \eta_p$, although $\eta_p$ will not be needed in the remainder of this work. The functions $\eta_s$ and $\eta_n$ are presented together in Fig. 1(a) for $K_s = 0.1$ and $K_n = 0.75$, the values found to best represent wide-gap
Couette flow data (see Sec. III B). From theory [Brady and Morris (1997)], it is known that the normal stresses for suspensions of near-hard spheres vanish quadratically as \( \phi \to 0 \) and this behavior is captured by \( \eta_n \). Also plotted is the Krieger (1972) model of \( \eta_s = (1 - \phi)^{-1.82} \). This standard form differs in its divergence and is larger than our modeled \( \eta_s \) for most \( \phi \) but is otherwise similar. All behavior predicted in this study would be qualitatively the same if the Krieger form were employed for \( \eta_s \). Qualitative differences occur only extremely near \( \phi = \phi_{\text{max}} \) and are due to the fact that the ratio \( q(\phi) = \eta_n / \eta_s \) would diverge rather than tend to a constant as in our modeling; \( q \) appears below in analysis of the wide-gap Couette flow, and is presented for \( K_n = 0.75 \) and \( K_s = 0.1 \) in Fig. 1(b). In the present modeling, \( q \) is monotonically increasing and tends to a finite value as \( \phi \to \phi_{\text{max}} \). While \( q \) must increase initially because \( \eta_n \) vanishes as \( \phi \to 0 \), it is an unproven assumption that the divergences of \( \eta_n \) and \( \eta_s \) are the same. The basis for this modeling is that the dominant normal and shear stresses in concentrated suspensions both result from the same particle interactions, namely from moments of hydrodynamic lubrication forces and contact forces.

In taking all normal stress components as compressive, we contradict conclusions of...
Laun (1994). Laun experimentally measured $N_1 \approx -2N_2 \approx -|\tau|$ ($\tau$ is the shear stress) in a study of electrostatically stabilized 280 nm styrene/ethyl acrylate particles suspended in glycol at a reported particle volume fraction of 0.587; the rheology was measured in a cone-and-plate rheometer and the suspensions were found to be extremely shear thickening. Laun interpreted the measured normal stresses as $\Sigma_{11} = -\frac{1}{2} |\tau|$, $\Sigma_{22} = \frac{1}{2} |\tau|$, and $\Sigma_{33} = 0$. This implies a tensile $\Sigma_{22}$, which is difficult to reconcile in terms of the relation of microstructure to rheological properties. In the absence of any other mechanism for tensile stresses besides hydrodynamics, the microstructure required for a tensile stress in a concentrated suspension has an excess of close pairs of particles with their center-to-center separation vector aligned with the bulk extensional axis. This is in striking contrast to structure determined from simulation [Phung et al. (1996); Katyal (1998)] and theoretical predictions [Brady and Morris (1997)] in which the accumulation of particle pairs is along the axis of compression and the axis of extension is essentially void of pairs at contact. These findings are supported by the experimental evaluation of pair structure in suspensions of noncolloidal spheres performed by Leighton and Rampall (1993). We shall discuss in Sec. III an interpretation of the results of Laun (1994) which requires consideration of suspension pressure, after presenting the predictions of our analysis for the cone-and-plate flow.

### III. CURVILINEAR-FLOW PREDICTIONS

The bulk suspension balance, for neutrally buoyant Stokes-flow suspensions, satisfies $\nabla \cdot \Sigma = 0$ at all times. During the period of migration beginning from onset of flow of a uniformly concentrated suspension, the phase stress divergences are both nonzero but are locally equal and opposite:

$$\nabla \cdot \Sigma_p = -\nabla \cdot \Sigma_f.$$  \hspace{1cm} (20)

The implication is that stress gradients, much like simple pressure gradients, are developed in the two phases of the flowing mixture. These stress gradients drive the components in opposite directions, resulting in a bulk segregation.

The unsteady problem is taken up for wide-gap Couette flow in Sec. III D. Before addressing that problem, we shall consider the steady state following migration. This state, in which the particle stress variation satisfies $\nabla \cdot \Sigma_p = 0$, is examined for the parallel-plate, cone-and-plate, and wide-gap Couette flows, with comparison to experimental observations providing estimates of parameters in the modeling wherever comparison is possible. The analysis assumes that the suspensions undergo a smooth variation in shear rate up to solid surfaces and satisfy noslip, and we have interpreted the experiments as also having this behavior. This neglects the possibility of "slip" of the particle phase at the walls.

#### A. Steady parallel-plate torsional flow

The steady or developed flow which occurs after particle migration is complete satisfies $\nabla \cdot \Sigma_p = 0$. In Secs. III A–C, we shall only need to consider the particle contributions to the stress and the subscript $p$ will be dropped from the particle stress.

For the torsional flow between two parallel circular plates generated by rotating one plate about its center at an angular velocity $\Omega$, the shear rate depends linearly upon distance from the center of the plates, $\dot{\gamma} = \partial u_\theta / \partial z = \Omega r / H$, with $H$ the distance between the plates. Assuming invariance of the shear rate in the direction normal to the plates ($z$) and in the angular direction ($\theta$) yields $(\nabla \cdot \Sigma)_r = 0$, which is explicitly written
For this flow, \( \theta \) and \( r \) are the flow and vorticity directions, respectively, and thus we may rewrite the previous expression as

\[
\frac{1}{r} \frac{\partial}{\partial r} (r \Sigma_{rr}) = \frac{\Sigma_{\theta \theta}}{r}.
\]  

(21)

Using the constitutive law, Eqs. (15) and (16), in this expression, with the assumption that variations of all quantities except \( u \) are in the \( r \) direction only, yields

\[
\lambda_3 \frac{d(\eta_n \dot{\gamma})}{dr} = \frac{\eta_n \dot{\gamma}(1-\lambda_3)}{r},
\]  

(23)

which may be solved, using \( \dot{\gamma} \sim r \), to obtain

\[
\eta_n(\phi) = A_1 r^{(1-2\lambda_3)\lambda_3},
\]  

(24)

with \( A_1 \) a constant determined by requiring \( \phi(r) \) to sum to the initial \( \phi_{\text{bulk}} \). The prediction is that the relationship between \( \phi \) and \( r \) is independent of the magnitude of the normal stress and depends only upon the functional form of \( \eta_n(\phi) \), as the leading constant \( K_n \) in \( \eta_n \) may be absorbed into \( A_1 \).

The experiments of Chapman (1990) and Chow et al. (1994) indicate that migration in the parallel-plate flow is weak or zero. This lack of migration can be predicted within the present modeling: a constant \( \eta_n \), implying constant \( \phi \), can be achieved in Eq. (24) if \( \lambda_3 = \frac{1}{2} \). If we do not have \( \lambda_3 = \frac{1}{2} \), \( \phi \) varies and the slope of \( d\phi/dr \) at \( r = 0 \) is seen from Eq. (24) to be infinite, suggesting a stress law with “nonlocality” is needed. While the precise \( \lambda_3 = \frac{1}{2} \) condition is excessively strict, the weakness of migration observed for this flow suggests that \( \lambda_3 = 0.5 \) is valid, and analytical studies at small particle fraction [Brady and Morris (1997)] support use of this value. In later sections, we shall simply take \( \lambda_3 = \frac{1}{2} \).

It is, however, possible to model a nonlocal stress. This has been performed by assuming a suspension temperature-dependent normal stress [Nott and Brady (1994); Morris and Brady (1998)], and alternatively by assuming that a transient network of particles forms which is capable of exerting a normal stress [Mills and Snabre (1995)]. Here, we take a modeling approach which allows the stress to depend in some fashion upon the spatial average of shear rate in a finite volume, \( \Delta V(x) \), centered on the point of interest \( x \)—the parallel-plate flow has nonzero average shear rate in any finite \( \Delta V \), despite the vanishing local average shear rate at \( r = 0 \). The approach of Nott and Brady (1994) has been developed and is known to require an additional dependent field variable (and consequently an additional dynamical equation) to describe the flow. The approach suggested here can, by contrast, be incorporated to the scheme developed in this work without additional field variables. For the parallel-plate flow, if we assume the size of \( \Delta V \) is constrained to the scale of the plate separation \( H \), a nonlocal stress contribution can be modeled as proportional to the gradient of the shear rate,

\[
\Sigma_{nl} = -K_{nl} \eta_n H \nabla \dot{\gamma} I.
\]

For simplicity, we have used \( \eta_n \) to describe the \( \phi \) dependence of this stress. \( \Sigma_{nl} \) is taken to be isotropic, in order that the nonlocal stress itself not cause deformation at a point.
where the average shear rate vanishes. Adding $\Sigma_{nl}$ to the constitutive law, Eqs. (15) and (16), for the stress, a relationship for $\eta_n$ as a function of position analogous to Eq. (24) is found:

$$\eta_n(\phi) = A_1 \left( r + K_{nl} \frac{H}{R} \right)^{(1 - 2\lambda_3)/\lambda_3}.$$  \hspace{1cm} (25)

In Fig. 2, the normalized particle fraction in a parallel-plate device of $H/R = 0.1$ for $\phi_{bulk} = 0.8\phi_{max}$, using $\lambda_3 = \frac{1}{2}$ in the local model and $\lambda_3 = 0.4$ and 0.6 with $K_{nl} = 1$ in the nonlocal model.

**FIG. 2.** The predicted normalized particle fraction in a parallel-plate device of $H/R = 0.1$ for $\phi_{bulk} = 0.8\phi_{max}$, using $\lambda_3 = \frac{1}{2}$ in the local model and $\lambda_3 = 0.4$ and 0.6 with $K_{nl} = 1$ in the nonlocal model.

The manner in which the nonlocal contribution to the stress was evaluated results in an aphysical finite $d\phi/dr$ at $r = 0$; our purpose is to illustrate the utility of the nonlocal stress concept and the simplicity with which it may be implemented without additional field variables, and we shall not pursue this issue here.

**B. Steady wide-gap Couette flow**

Particle migration in the wide-gap Couette flow confined between two concentric cylinders has been studied using nuclear magnetic resonance (NMR) imaging of the particle fraction for nearly monodisperse concentrated suspensions of spherical noncolloidal particles by Abbott et al. (1991) and Phillips et al. (1992); agreement between the results of the two studies for the steady particle fraction is excellent. We use the data of Phillips et al. for comparison to our predictions; the external radius of the inner cylinder in these experiments was $R_i = 0.64$ cm and the internal radius of the outer cylinder was $R = 2.38$ cm, yielding $R_i/R = 0.239$.

As in the parallel-plate flow, the radial momentum balance at steady state reduces to
\[
\frac{1}{r} \frac{\partial}{\partial r} \left( r \Sigma_{rr} \right) = \frac{\Sigma_{\theta \theta}}{r}. \tag{26}
\]

In this flow, \( \theta \) and \( r \) are the flow and gradient directions, respectively. Thus, in terms of the first normal stress difference, the previous balance is given by

\[
\frac{\partial \Sigma_{rr}}{\partial r} = \frac{\Sigma_{\theta \theta} - \Sigma_{rr}}{r} = \frac{N_1}{r}. \tag{27}
\]

Using the constitutive relations, Eqs. (15) and (16) and replacing \( N_1 \) by its form given in Eq. (17) yields

\[
\lambda_2 \frac{d(\eta_\varphi \dot{\gamma})}{dr} = \frac{\eta_\varphi \gamma(1 - \lambda_2)}{r}, \tag{28}
\]

which is solved in conjunction with the shear stress balance

\[
\frac{1}{r} \frac{\partial}{\partial r} \left( r^2 \Sigma_{r \theta} \right) = 0, \quad \Sigma_{r \theta} = \eta_\varphi(\phi) \dot{\gamma},
\]

and

\[
\dot{\gamma} = r \frac{\dot{u}_\theta}{r}. \tag{29}
\]

The shear stress balance yields

\[
\dot{\gamma} = \frac{C}{r^2 \eta_\varphi(\phi)}, \tag{30}
\]

which is substituted in Eq. (28) to yield

\[
\frac{\eta_\varphi(\phi)}{\eta_\varphi(\phi)} = q(\phi) = A_2 r^{(1 + \lambda_2) / \lambda_2}, \tag{31}
\]

with \( A_2 \) a constant determined by requiring \( \phi(r) \) to average to the imposed \( \phi_{\text{bulk}} \).

The predicted particle fraction variation for a bulk particle fraction of \( \phi_{\text{bulk}} = 0.5 \) and a maximum packing fraction of \( \phi_{\text{max}} = 0.68 \) for a wide-gap Couette flow with \( R/R_i \) matching that employed by Phillips et al. (1992) is shown in Fig. 3(a); Fig. 3(b) shows \( u_\theta/\Omega R_i \). [The choice of \( \phi_{\text{max}} = 0.68 \) is made in order to be consistent with the observations of Phillips et al. (1992) and allow for direct comparison of predictions with their results. The typical value for random close packing of monodisperse spheres is \( \phi_{\text{rcp}} = 0.63 \), but sufficient polydispersity or other factors allow shearing at higher fractions for the suspensions of Phillips et al. (1992)]. The results are presented for \( \lambda_2 = 0.6, 0.8 \) and 1. The values of Phillips et al. (1992) are judged to be matched most closely by the predictions obtained for \( \lambda_2 = 0.8 \) and \( K_s = 0.1 \), although \( \lambda_2 = 0.6 \) and 1 also provide reasonable agreement. From Eq. (31), the relationship which is indicated by \( \lambda_2 = 0.8 \) is \( q \propto r^{2.25} \), and thus when viewed as a function of \( r \) for the device of interest, \( q(R)/q(R_i) = (R/R_i)^{2.25} = 19.2 \), i.e., the ratio of normal to shear viscosity must undergo a large variation.

The predictions for \( \phi_{\text{bulk}} = 0.45, 0.5, \) and 0.55 with \( \phi_{\text{max}} = 0.68 \) are presented, using values of \( \lambda_2 = 0.8 \) and \( K_s = 0.1 \) determined to provide a good fit of the \( \phi_{\text{bulk}} = 0.5 \) data, in Figs. 4(a) and 4(b). Also plotted are the experimental \( \phi \) from Phillips.
et al. (1992) for these bulk particle fractions. All results are plotted in the form $\phi / \phi_{\text{max}}$. Agreement between predictions and the observed particle fractions is generally excellent. Because of the migration, the normalized velocity of the suspension varies much more rapidly near the inner cylinder—where $h_s$ is smallest—than would a uniform-viscosity Newtonian fluid.

It is of interest to consider other particle fractions, and different geometries of the wide-gap Couette apparatus. Predictions for the steady bulk fractions from $\phi_{\text{bulk}} / \phi_{\text{max}} = 0.1−0.9$ in increments of 0.2 are presented in Fig. 5. The results of Fig. 5 provide a guide to the migration to be expected for all fractions in this wide-gap Couette device, provided the rheological model is reasonably accurate for dilute fractions. A notable feature is that as $\phi_{\text{bulk}} / \phi_{\text{max}}$ increases, the curvature of the particle fraction curve, $d^2 \phi / dr^2$, increases. This is consistent with the experimental data of Phillips et al. (1992), as seen from Fig. 3, and can be understood in the context of the rheological basis for the migration. Figure 1(b) illustrates that for the larger fractions, the necessary variation of $q(R)/q(R_i) = 19.2$ requires that $\phi / \phi_{\text{max}}$ pass from the slowly varying region to
the rapidly varying region of the curve occurring at $\phi / \phi_{\text{max}} = 0.85$. A related point is that there is a limit to the maximum bulk fraction which can be described by this model, as maximum packing is achieved at the outer wall for some value $\phi_{\text{bulk}} / \phi_{\text{max}} < 1$. The predicted maximum bulk fraction with $\lambda_2 = 0.8$, $K_s = 0.1$, and $K_n = 0.75$ is $\phi_{\text{bulk}} = 0.91 \phi_{\text{max}}$.

Figures 6(a) and 6(b) provide predictions for the steady $\phi(r)$ and $u_\theta(r)$, respectively, of a suspension of $\phi_{\text{bulk}} / \phi_{\text{max}} = 0.8$ for $R_i/R = 0.3, 0.5, 0.7,$ and $0.85; \lambda_2 = 0.8$ is used in the modeling. One result which may be surprising is that even for $R_i/R = 0.85$, where $u_\theta$ apparently varies little from linearity with respect to $r$ over the gap, the particle fraction is predicted to vary by almost 10% across the gap in order that $q(R)/q(R_i) = 1.44$ as required by the solution.

The steady predicted and experimentally-measured [Phillips et al. (1992)] particle fraction for $\phi_{\text{bulk}} = 0.45, 0.5,$ and $0.55$ (with $\phi_{\text{max}} = 0.68$) for a wide-gap Couette flow with the ratio of inner to outer radius of $0.269$. The values $K_s = 0.1, K_n = 0.75, \lambda_2 = 0.8,$ and $\lambda_3 = 1/2$ were used in the model.

FIG. 4. The steady predicted and experimentally-measured [Phillips et al. (1992)] particle fraction for $\phi_{\text{bulk}} = 0.45, 0.5,$ and $0.55$ (with $\phi_{\text{max}} = 0.68$) for a wide-gap Couette flow with the ratio of inner to outer radius of $0.269$. The values $K_s = 0.1, K_n = 0.75, \lambda_2 = 0.8,$ and $\lambda_3 = 1/2$ were used in the model.

FIG. 5. Predicted steady volume fractions for $\phi_{\text{bulk}} / \phi_{\text{max}} = 0.1–0.9$ for a wide-gap Couette suspension flow with the ratio of inner to outer radius of $0.269$. The values $K_s = 0.1, K_n = 0.75, \lambda_2 = 0.8,$ and $\lambda_3 = 1/2$ were used in the model.
C. Steady cone-and-plate flow

In small-angle cone-and-plate flow, the shear rate is essentially constant under the cone. This flow has been studied for concentrated suspensions by Chow et al. (1995) using NMR imaging techniques, and it was observed that particles migrate radially outward. Specifically, in a truncated cone device, it was found that $\phi$ grew progressively larger with time under shear in the cone region at larger $r$, while being depleted in the essentially parallel-plate flow under the truncated cone at small $r$. The detailed dependence of $\phi$ upon $r$ was not determined.

In the cone-and-plate flow $\phi$ is the angle about the cone axis and is the flow direction, $\theta$ is the azimuthal (cone) angle and is the gradient direction, while $r$ is the vorticity direction. The maximum value of $r$ is denoted by $R$ and the radius of the truncated region by $R_i$.

The shear rate in a cone-and-plate device is

$$\dot{\gamma} = \frac{1}{r} \frac{\partial u_\phi}{\partial \theta}.$$
The radial momentum balance at steady state reduces to $(\nabla \cdot \Sigma)_r = 0$, which expands to

$$
\frac{\partial \Sigma_{rr}}{\partial r} = \frac{\Sigma_{\phi \phi} + \Sigma_{\theta \theta} - 2 \Sigma_{rr}}{r} = \frac{N_1 + 2N_2}{r}.
$$

(32)

Introducing the constitutive model, Eqs. (15) and (16), this can be rewritten

$$
\lambda_3 \frac{\partial (\eta_n \dot{\gamma})}{\partial r} = \frac{1 + \lambda_2 - 2 \lambda_3}{r} \eta_n \dot{\gamma},
$$

(33)

which is solved assuming $\dot{\gamma}$ constant to yield

$$
\eta_n(\phi) = A_3 r^{(1 + \lambda_2 - 2 \lambda_3) / \lambda_3},
$$

(34)

with $A_3$ a constant determined by requiring $\phi$ to sum to $\phi_{\text{bulk}}$. With $\lambda_3 = 1$, this will predict $\eta_n \sim r^{2 \lambda_2}$ and therefore outward migration for all positive $\lambda_2$. Consideration of the manner in which $\lambda_2$ and $\lambda_3$ enter in the general relation of $\phi$ to $r$ expressed by Eq. (34) shows that an isotropic normal stress with $\lambda_2 = \lambda_3 = 1$ would yield no migration. Furthermore, the presence of any shear-induced anisotropic normal stress is predicted to result in migration with the steady-state particle fraction independent of the magnitude of the normal stress, i.e., the magnitude of $K_n$ does not influence the results. Hence, the influence of normal stresses on the steady state is found to be singular.

In Fig. 7, the predicted steady particle fraction profiles in a full cone-and-plate for varying $\lambda_2$, with fixed $\lambda_3 = 1$, are shown for a bulk particle fraction of $\phi_{\text{bulk}} = 0.8 \phi_{\text{max}}$. In Fig. 8, predictions are presented for the local $\phi$ for $\phi_{\text{bulk}} = 0.8 \phi_{\text{max}}$ in a truncated cone of the same geometry as was used by Chow et al. (1995). (A truncated cone with radius of truncation region $R_i = 5.535$ mm, overall plate radius of $R = 21$ mm, and gap in the truncation region of $H = 0.55$ mm was used, with particles of radius 50 and 25 $\mu$m; comparison is made only with experimental results using 50 $\mu$m radius particles.) In Fig. 9, we show the predictions of our model for a truncated cone device along with the modeling of Leighton in the Chow et al. (1995) study. The present model is found to agree well with that work for a value of $\lambda_2 = 0.58$, under the assumption of $\phi_{\text{bulk}} = 0.68$. Leighton used the diffusive flux model [Phillips et al. (1992)] with additional curvature terms [Krishnan et al. (1996)]. The results shown were determined.
to be the best fit of the time dependence of the experimentally determined ratio of the average of \( \phi \) in the outer region to its average in the truncated cone region. We have not modeled the unsteady cone-and-plate migration. In Fig. 10, the predicted steady-state volume fractions for a truncated cone-and-plate device for a range of volume fractions are shown using \( \lambda_2 = 0.8 \), and \( \lambda_3 = 0.5 \). Figures 7–10 indicate that as \( \lambda_2 \) increases, the outward migration is predicted to become more pronounced, leaving the apex region of a full cone device nearly free of particles. The truncated region is predicted to have a constant \( \phi \), because it is a parallel-plate arrangement and \( \lambda_3 = \frac{1}{2} \). It is interesting that if \( 1 + \lambda_2 - 2\lambda_3 < 0 \), inward migration is predicted.

1. Interpretation of Laun results

We have noted in Sec. II B that Laun (1994) interpreted the values of \( N_1 = -2N_2 \) in cone-and-plate rheometry of a shear-thickening dispersion to mean \( \Sigma_{11} = -|\tau|/2 \), \( \Sigma_{22} = |\tau|/2 \), and \( \Sigma_{33} = 0 \). The results for the normal stresses of the Laun

![Diagram](image_url1)

**FIG. 8.** Predicted steady volume fractions in a truncated cone-and-plate device, with \( R_i/R = 0.264 \) for a range of values of \( \lambda_2 \). The bulk particle fraction is \( \phi_{\text{bulk}}/\phi_{\text{max}} = 0.8 \) and \( \lambda_3 = 0.5 \) in all cases.

![Diagram](image_url2)

**FIG. 9.** Predicted steady volume fractions in a truncated cone-and-plate device at \( \phi_{\text{bulk}} = 0.5 \), with \( \lambda_3 = 0.5 \), and \( \lambda_2 = 0.5, 0.58, \) and 0.8; \( \lambda_2 = 0.58 \) is judged in best agreement with predictions by Leighton for \( \phi_{\text{bulk}} = 0.5 \) [from Chow et al. (1995)]. It is assumed that \( \phi_{\text{max}} = 0.68 \) for both sets of data.
experiments are, like all such measurements, ambiguous in the sense that the first normal stress and hence the presence of a net isotropic normal stress could not be measured. The stresses could thus be interpreted more generally as $S_{11} = \frac{P}{3}U_{1} \frac{U_{1}}{2}$, $S_{22} = \frac{P}{3}U_{2} \frac{U_{2}}{2}$, and $S_{33} = \frac{P}{3}$, with all stresses compressive if $P > \frac{3U_{1}U_{2}}{2}$. Laun has taken the form with $P = 0$, a result difficult to reconcile with microstructural theory because it results in a tensile $S_{22}$. The ratios of the normal stresses obtained from this interpretation are quite different from the values determined to be appropriate in the present study. However, the extreme thickening observed suggests the mixture is much closer to maximum packing than those to which we make comparisons. The physical situation may be dominated by the constraints of being close to this singular state, forcing it to respond in a manner necessary to keep the concentration from varying; the extreme thickening observed suggests that even a small local increase in particle fraction could generate a very large increase in the stress in that vicinity, and this would violate the normal stress balance. Interestingly, this interpretation is suggested by the analysis presented above: from Eq. (32), noting that $\dot{\gamma}$ is constant and variation of $\Sigma_{rr}$ therefore reflects variation of $\phi$, we observe that the requirement for vanishing migration in the cone-and-plate flow is $N_{1} + 2N_{2} = 0$, the relationship found experimentally by Laun.

D. Unsteady wide-gap Couette flow

The particle fraction evolution is governed in general by Eq. (14). For the case of the wide-gap Couette flow, the average convective derivative, $\langle \mathbf{u} \rangle \cdot \nabla \phi$, vanishes to yield

$$\frac{\partial \phi}{\partial t} = -\frac{2a^2}{9\eta} \nabla \cdot [f(\phi)\nabla \Sigma_{p}],$$

which in the wide-gap Couette flow, where variations are assumed to be only in the $r$ direction, becomes

$$\frac{\partial \phi}{\partial t} = -\frac{2a^2}{9\eta} \frac{1}{r} \frac{\partial}{\partial r} \left[ f(\phi) \left( \frac{\partial \Sigma_{rr}}{\partial r} - N_{1} \right) \right].$$

FIG. 10. Predicted steady volume fractions in a truncated cone-and-plate device for several bulk volume fractions, with $\lambda_2 = 0.8$ and $\lambda_3 = 0.5$. 

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Clearly \( \partial \phi / \partial t \) depends upon the normal stresses through \( \Sigma_{rr} \) and \( N_1 \), and available experimental data of \( \phi(r,t) \) in unsteady wide-gap Couette flow thus provides information on the magnitude of shear-induced normal stresses. We compare the predictions of Eq. (36) with the evolving-flow experiments of Phillips et al. (1992). In the experiments, \( \phi(r,t) \) was measured and reported as a function of the number of revolutions of the inner cylinder, with the outer held fixed.

The algorithm used to solve Eq. (36) is an explicit scheme employing a Euler method for the time dependence, i.e., \( \Delta \phi(r_i) = \mathcal{R}(r_i) \Delta t \), where \( \mathcal{R}(r_i) \) is shorthand for evaluation of the RHS of Eq. (36) at a generic mesh point \( r_i \). The value of \( \mathcal{R} \) for interior points was determined by a central difference scheme. The values needed to evaluate the differences are those at the prior time. The shear rate is determined from Eq. (30) and the requirement that the velocity have specified values at the inner and outer walls using \( \phi(r_i, t_{i-1}) \), the particle fraction at the prior time step. For numerical purposes, the migration problem requires consideration only of Eq. (36).

The boundary conditions on \( \phi \) are that the particle flux normal to the boundary must vanish. The streamlines are assumed to be purely \( \theta \) directed, and the condition therefore requires \( j_{\perp,r} \cdot n_{\text{walls}} = j_{\perp,r} \sim (\nabla \cdot \Sigma_p) \bigg|_{\text{walls}} = 0 \). For a general point, the vanishing of \( j_{\perp,r} \) yields

\[
\frac{\partial \phi}{\partial r} = \frac{q}{q^r} \frac{1 + \lambda_2}{\lambda_2 r},
\]

which is derivable by differentiation from the steady-state condition, Eq. (31), for \( \phi(r) \) in this flow. The condition, Eq. (37), was enforced at the boundaries at all times in the numerical solution, by requiring \( \partial \phi / \partial r \) be evaluated numerically between the boundary node and the interior node directly adjacent to have the specified value. In discretized form, at the inner boundary (where \( r = R_i \)),

\[
\phi(R_i, t_i) = \phi(R_i, t_{i-1}) - \Delta r \frac{q}{q^r}(R_i, t_{i-1}) \frac{1 + \lambda_2}{\lambda_2 R_i},
\]

in which \( R_i \) denotes the radial position of the interior node adjacent to the inner boundary, and \( \Delta r = R_i - R_{i-1} > 0 \). A similar expression holds at the outer wall.

Having specified the initial particle fraction and imposed vanishing particle flux at inner and outer boundaries, the solution is fully specified and \textit{ad hoc} renormalization of the particle fraction should not be required; conservation of particle mass is satisfied identically by the algorithm. The deviation of the bulk average particle fraction in the computed \( \phi(r, t) \) was typically 0.1% or less for a 100 node radial mesh, and this deviation is attributable to discretization error.

The predicted time-dependent solution for a suspension of \( \phi_{\text{bulk}} / \phi_{\text{max}} = 0.8 \) and \( a/R = 0.0142 \) is shown in Fig. 11 after 100, 200, 800, and 12,000 revolutions of the inner cylinder, along with the experimentally determined particle fractions in a suspension with \( \phi_{\text{bulk}} = 0.55 \) [Phillips et al. (1992)] after 200 and 12,000 revolutions. Agreement with experiment is excellent at 200 revolutions for \( K_p = 0.75 \): this value was judged by eye to be an optimal fit to the data of Phillips et al. While the experimental data were found to undergo negligible change from 800 to 12,000 revolutions for this condition, the model predicts some variation after 800 revolutions. A point of interest is that the particle fraction is predicted to become nonmonotonic with respect to \( r \) in the early stages of the migration, developing a local minimum at \( r/R \approx 0.83 \); the experiments of Phillips et al. seem to have similar behavior, but the slightly nonuniform initial
loading may be misleading. Using $K_n = 0.75$, the evolution of $\phi(r,t)$ was predicted for $a/R = 0.0021$, with results presented in Fig. 12. This matches the conditions of Phillips et al. for particles of 50 $\mu$m mean radius in the apparatus previously described, and the agreement with the experimental data at 8000 revolutions is excellent, confirming that dependence of the migration rate upon particle size as $a^2$ is valid for these conditions, as was also shown by Phillips et al.

In the predictions presented in Figs. 11 and 12, $\alpha = 4$ was used in the hindrance function, $f(\phi) = (1 - \phi)^\alpha$ which appears in Eq. (36). Although the steady-state particle fraction is independent of $f$, the rate of migration depends upon $f$ as well as $\eta_n$. As the focus of the present study is upon the rheological basis for particle migrations, we have

FIG. 11. The volume fraction for the evolving flow in a wide-gap Couette flow, with $a/R = 0.0143$, after 100, 200, 800, and 12,000 revolutions of the inner cylinder. The predicted profiles are judged to be in the best agreement with the experimental data of Phillips et al. (1992) for $K_n = 0.75$, with $K_s = 0.1, \lambda_2 = 0.8$, and $\lambda_3 = 1$. No-slip of the particle phase is assumed in comparison of the model predictions with the experimental data.

FIG. 12. The predicted volume fraction for the evolving flow in a wide-gap Couette flow of $R_i/R = 0.269$ and $a/R = 0.0021$. The predicted profiles are obtained using parameter values $K_s = 0.1, K_n = 0.75$, and $\lambda_2 = 0.8$. Experimental data of Phillips et al. (1992) for the same value of $a/R$ at 8,000 revolutions are also shown.
not attempted to thoroughly decouple these two influences upon the rate of migration (the available data would, in fact, not support such an effort). To provide an indication of the influence of upon migration rate, unsteady results for three values of in a suspension of / = 0.8 and / / = 0.014 are presented in Fig. 13 for / = 0.1 and / = 0.75. The parameter values are / = 0.1, / = 0.75, and / = 0.68. The results shown are the particle fraction after 200 revolutions of the inner cylinder. The values for / = 2, 4, and 6 are plotted, and as increases, the rate of migration is dramatically reduced, although the shape of the curve is not substantially altered.

The condition \( \mathbf{j}_w \cdot \mathbf{n} = 0 \) used in solution of the wide-gap Couette flow migration problem must hold for impermeable boundaries in general. This condition is forced, in this work, to hold at arbitrarily short times after the initial condition using the same rheological model employed in the steady state, and as a result requires an instantaneous change in the near-surface \( \phi \). This is a consequence of the implicit assumption that the suspension microstructure necessary to yield normal stresses is developed instantaneously. In reality, a time of \( O(\gamma^{-1}) \times \text{fxn}(\phi) \) [\text{fxn}(\phi) here implies a function of \( \phi \) only, presently unknown] is required before the structural anisotropy is developed and normal stresses are exerted [Phan-Thien (1995); Brady and Morris (1997)]. Here, the time scales of interest for the bulk migration are vastly greater than the time for development of local structure and we ignore effects on the shorter time scale. Another factor worth noting is that the microstructure, and hence the rheology, will be influenced by the presence of a wall. This is also neglected, based on the fact that the particles are small relative to the flow scale for the wide-gap Couette flow and parallel-plate flow to which we have made comparisons. This may not be true in the case of the truncated portion of the cone in the cone-and-plate flow data [Chow et al. (1995)] to which primarily qualitative comparison has been made, and thus care should be taken in interpreting results for this geometry. Valid modeling requires better understanding of this issue, but it is expected that boundary-induced structural variations will be the primary feature leading to nonuniformity in \( \phi \) for suspension flows where migration in the bulk is not expected, such as a simple shear between parallel plates.

**FIG. 13.** The predicted volume fractions after 200 revolutions of the inner cylinder for / = 2, 4, and 6 in the hindrance function model, \( f(\phi) = (1 - \phi)^\alpha \), in a suspension of / = 0.0143, / = 0.8. The parameter values are / = 0.1, / = 0.75, and / = 0.68.
IV. CONCLUDING REMARKS

This work has demonstrated that a rheological approach in which shear-induced normal stresses provide the driving force for migration is able to explain the migration phenomena observed in curvilinear flows of concentrated suspensions. Because normal stress differences are dynamically relevant in curvilinear flows, the anisotropy of the stresses is influential in the migration process. Prediction of flows ultimately requires modeling of the rheology and other transport coefficients: $\eta_n(\phi)$, $\eta_s(\phi)$, and $f(\phi)$. However, the basic concept that migration is driven by normal stress rheology is demonstrated by analysis of mass and momentum conservation for the suspension. The analysis presented in Sec. II and at the beginning of each of Secs. III A–C shows that the observed migrations in wide-gap Couette flow, parallel-plate, and cone-and-plate flow can all be rationalized by including compressive normal stresses proportional to $\eta_y$.

To make full use of the relationship between rheology and particle migration requires unambiguous normal stress data for concentrated suspensions. The work of Gadala-Maria (1979) in this area has not been followed by an abundance of further data. One difficulty in obtaining such data is the smallness of the normal stresses until the suspensions become concentrated, because of the $\Sigma_{ij} \sim \phi^2$ dependence for $\phi \rightarrow 0$. Simulation has provided insight into the rheology, with determinations even of the suspension pressure [Yurkovetsky (1997)] which this and other work [Nott and Brady (1994); Morris and Brady (1998)] has shown to be important to the migration phenomenon, but cannot substitute for experiment.

There remain many questions in regard to the general utility of the approach presented here for the prediction of flows of concentrated suspensions. Polydispersity is one obvious difficulty: Krishnan et al. (1996) showed that tracer particles migrate inward or outward in a parallel plate flow if they are smaller or larger, respectively, than the other particles of an otherwise monodisperse suspension. Rheology in more general flows must also be considered, as in most practical cases the suspension will be subjected to flows other than simple shear. Another issue of substantial practical concern is the influence of viscoelasticity in the suspending fluid upon migration of particles, which has been demonstrated experimentally by Tehrani (1996). In a suspension with a viscoelastic suspending fluid, modeling of the normal stresses in the mixture will be complicated because normal stress contributions arise from both particles and fluid. The rheological modeling used here assumed that interparticle noncontact forces or surface roughness are able to balance the hydrodynamic force and produce an anisotropic, rate-independent microstructure for noncolloidal (Pe $\rightarrow \infty$) suspensions. It is of substantial interest to examine suspension normal stress rheology and migration phenomena at finite Pe, where Brownian motion influences the rheology.

A natural but complicating factor in the approach presented here is that some $\phi$ dependent function, which we have taken as the sedimentation hindrance function $f(\phi)$, must be used to relate the particle flux to the “driving force” for migration provided by divergence of the particle stress. Results from Sec. III D showed that this function has substantial impact upon the rate of migration. Use of $f$ as a resistance to the bulk relative migration of the two phases works well and appears well founded, but further study is needed to decouple the influence of $\nabla \cdot \Sigma_p$ from $f$ in the migration flux.

We close by emphasizing that the influence of shear-driven normal stresses is found to be singular, in the sense that their presence results in a qualitatively different steady particle fraction and bulk flow than would be found for the equal $\phi$ Newtonian suspension. Modeling of the normal stresses was kept relatively simple in this study to allow analytical progress in examining the influence of the anisotropy. Understanding of the $\phi$
dependence of $\lambda_3 = \Sigma_{22}^{p}/\Sigma_{11}^{p}$ and $\lambda_3 = \Sigma_{33}^{p}/\Sigma_{11}^{p}$ is needed. The simplification of taking these ratios constant with respect to $\phi$ used here is certainly not generally valid, although it is expected to provide qualitatively correct results.

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References


