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RHEOLOGY OF A DILUTE SUSPENSION OF DIPOLAR SPHERICAL PARTICLES IN AN EXTERNAL FIELD

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I. INTRODUCTION

Einstein's formula (1, 2) for the viscosity of a dilute suspension of rigid spherical particles uniformly dispersed in a Newtonian fluid is valid only when the suspended particles rotate with the local vorticity of the fluid motion. Free rotation of this nature is possible only when no external couples act upon the individual particles. However, circumstances are easily envisioned in which this condition will not be met. For example, if the center of mass of each particle is displaced from its geometric center owing to inhomogeneities in the internal mass distribution, couples will arise from the action of gravity whenever the embedded gravity dipole does not lie parallel to the direction of the gravitational field. Similarly, permanent magnetic or electric dipoles embedded within the suspended particles will give to couples in the presence of external magnetic or electric fields.

The net external couple acting upon each particle is governed by the orientation of its dipole vector relative to the direction of the external field. When rotary Brownian motion is negligible, this orientation is governed by a balance between the hydrodynamic shear tending to rotate the sphere, thereby carrying the dipole with it, and by the external field tending to align the dipole in the direction of the field.

The existence of couples results in a hindered rotation, whereby the particles no longer rotate freely with the fluid. Indeed, free rotation will prevail only for the special case where the local vorticity vector is colinear with the direction of the external field. The phenomenon of hindered rotation results in a greater rate of mechanical energy dissipation (3) and, hence, a larger apparent viscosity than would exist in the absence

of such couples — all other things being equal. Since the distribution of dipole orientations will generally change in response to changes in the rate of shear, such a suspension will manifest non-Newtonian behavior in the sense that the apparent viscosity will be shear-rate dependent. Moreover, for a specified experimental configuration and shear rate, the magnitude of the apparent viscosity will also depend upon the orientation of the apparatus relative to the external field — giving rise to a type of configurational anisotropy.

Working independently, Hall and Busenberg (4) and Brenner (5) each developed a partial theoretical analysis of the above phenomena for the case of dilute suspensions of rigid spheres, and utilized their results to provide a qualitative interpretation of the respective ferromagnetic suspension viscosity experiments of McTague (6) and Rosensweig _et al_ (7). The two theoretical analyses complement one another, and their union furnishes an essentially complete description of the phenomenon for the case where the suspended particles are sufficiently large to ignore the effects of rotary Brownian motion. The main purpose of this paper is to effect this union and to point out some consequences of the general theory by means of several illustrative examples.

The Hall-Busenberg analysis treats the general case where the angle between the directions of the vorticity vector and the external field may have any value whatsoever, whereas Brenner treats only the two special cases where the vorticity vector is colinear with, or perpendicular to, the external field. On the other hand, Hall and Busenberg's energy dissipation analysis furnishes only the apparent viscosity of the suspension, (and then only for a Couette flow), whereas Brenner's treatment furnishes a complete dynamical theory of the stresses extant in such dipolar suspensions. The contrast is that which exists between a scalar (the apparent viscosity) and a second-rank tensor (the stress). In particular, Brenner's theory points up the fact that the stress tensor in a dipolar suspension will not be symmetric owing to the existence of external body couples. The increase in apparent viscosity, over and above the Einstein value, is ultimately attributable to this asymmetry.

The effects of electric and magnetic fields on the macroscopic properties of dilute, dipolar suspensions of ellipsoids of revolution have been extensively studied, especially with regard to their effects on streaming birefringence and suspension viscosity. These are reviewed in a series of papers by Chaffey and Mason (8, 9, 10), to which should be added the work of Saito and Kato (11) and Argyropoulos (12). None of this prior work is relevant to the present investigation for one or more of the following reasons: (i) the analysis is concerned solely with the case where the dipoles are induced by the field, rather than being permanent dipoles as in the present investigation; (ii) the analysis assumes that rotary Brownian motion, rather than the hydrodynamic or external field, constitutes the dominant influence, whence the calculation proceeds by assuming a small perturbation about the randomly-oriented state; (iii) calculation of the rheological properties of the suspension is based upon the original energy-dissipation approach of Jeffrey (13) and thereby furnishes, at most, the apparent viscosity of the suspension rather than the complete dynamical stress-strain relation; (iv) the analysis is confined only to the cases where the vorticity vector is either parallel or perpendicular to the external field.

II. GENERAL DYNAMICAL THEORY

Consider a dilute suspension of identical rigid spheres each of which possesses an identical permanent dipole locked into it. The homogeneous carrier fluid is assumed to be incompressible and Newtonian, its viscosity being μ_o and density ρ. For definiteness we regard the dipole as being a gravity dipole, and imagine the suspension to be present in a gravity field characterized by the vector $\mathbf{g}$, equal in magnitude to the strength $g = |\mathbf{g}|$ of the gravity field and pointing in the direction of the field. Such a particle is conveniently referred to as a "loaded" sphere. Comparable results for permanent magnetic or electric dipoles in magnetic or electric fields may be adduced at once by merely redefining the meaning of the symbols which appear in the present analysis. The mean density of each particle is assumed to be identical to that of the carrier fluid. The suspended particles therefore experience no net external force and, hence, display no tendency to settle. Finally, inertial effects will be systematically neglected throughout the analysis compared with viscous forces.

For the circumstances described above, Brenner (5) has shown that the fundamental linear momentum, angular momentum, and continuity equations appropriate to such a suspension, regarded as a macroscopic continuum, are

$$\nabla \cdot \mathbf{P}' + \rho \mathbf{g} = \mathbf{0} \qquad [1]$$

$$\mathbf{T}_\times + \mathbf{G} = \mathbf{0} \qquad [2]$$

$$\nabla \cdot \mathbf{v} = 0 \qquad [3]$$

Here,

$$\mathbf{P}' = -\mathbf{I}\, p' + \mathbf{T} \qquad [4]$$

is the macroscopic pressure tensor, $\mathbf{I}$ the dyadic idemfactor, p' the pressure, $\mathbf{T}$ the deviatoric stress tensor, $\mathbf{G}$ the external body couple density per unit volume exerted by the surroundings on the suspension, and $\mathbf{v}$ the mass-average velocity of the suspension. In addition, $\mathbf{T}_{\times} = -\boldsymbol{\epsilon}:\mathbf{T}$ is the vector invariant of the deviatoric stress[1], in which $\boldsymbol{\epsilon}$ is the unit alternating isotropic triadic. The double-dot notation follows the nesting convention of Chapman and Cowling (14).

These relations are identical to the usual continuum-mechanical equations for a continuum when macroscopic inertial forces are negligible and couple stresses are absent (or negligible). It should be emphasized, however, that these equations have not merely been postulated to be valid on the basis of a priori arguments as to the applicability of established continuum-mechanical and continuity principles to heterogeneous continua. Rather, they have been derived from 'first' principles, beginning with the corresponding "microscopic" interstitial fluid-mechanical equations, coupled with the equations of rigid-body mechanics applied to the individual suspended particles.

The external force density per unit volume of suspension, $\rho\mathbf{g}$, may be absorbed into the stress by defining the hydrodynamic pressure, $p = p' - \rho\mathbf{g} \cdot \mathbf{R}$, and the hydrodynamic pressure tensor $\mathbf{P} = \mathbf{P}' + \mathbf{I}\,\rho\mathbf{g} \cdot \mathbf{R}$, where $\mathbf{R}$ is the position vector. In place of Eqs. [1] and [4] we then have

$$\nabla \cdot \mathbf{P} = \mathbf{0} \qquad [5]$$

and

$$\mathbf{P} = -\mathbf{I}\, p + \mathbf{T} \qquad [6]$$

Brenner (5) has shown that the deviatoric stress in the suspension is given by

$$\mathbf{T} = \mathbf{T}^{s} + \mathbf{T}^{a} \qquad [7]$$

where the symmetric and antisymmetric portions are given by the constitutive relations

$$\mathbf{T}^{s} = \mu \left[\nabla\mathbf{v} + (\nabla\mathbf{v})^{\dagger} \right] \qquad [8]$$

and

$$\mathbf{T}^{a} = \frac{1}{2}\boldsymbol{\epsilon} \cdot \mathbf{T}_{\times} \qquad [9]$$

in which

$$\mathbf{T}_{\times} = \zeta\, (\boldsymbol{\omega} - \overline{\boldsymbol{\Omega}}) \qquad [10]$$

In the above,

$$\mu = \mu_o \left(1 + \frac{5}{2}\phi\right) \qquad [11]$$

is the coefficient of shear viscosity, in which ϕ is the volume fraction of suspended particles, and

$$\zeta = 6\mu_o\, \phi \qquad [12]$$

is the coefficient of spin viscosity; $\boldsymbol{\omega} = \frac{1}{2}\nabla \times \mathbf{v}$ is the vorticity of the suspension, and $\overline{\boldsymbol{\Omega}}$ is the mean angular velocity of the suspended particles.

The above system of dynamical and kinematical relations is valid only to the first order in ϕ. Consider two identical experiments, one performed with the homogeneous carrier fluid alone, and the other performed with the suspension. The same macroscopic boundary conditions are to be maintained in both experiments, e.g., no slip at the solid apparatus boundaries. One may then write, correctly to the first order in ϕ, that

$$p = p_o + \phi\, p_1 \qquad [13]$$

$$\mathbf{v} = \mathbf{v}_o + \phi\, \mathbf{v}_1 \qquad [14]$$

where $(\mathbf{v}_o, p_o)$ is the undisturbed field arising from the motion of the carrier fluid, and $(\mathbf{v}_1, p_1)$ is the perturbation field arising from the presence of the suspended particles in the flow. It follows that $\boldsymbol{\omega} = \boldsymbol{\omega}_o + O(\phi)$, where

$$\boldsymbol{\omega}_o = \frac{1}{2} \nabla \times \mathbf{v}_o \qquad [15]$$

is the vorticity vector associated with the undisturbed flow. Since the spin-viscosity coefficient is already of order ϕ, it follows that Eq. [10] may be replaced by

$$\mathbf{T}_\times = \zeta (\boldsymbol{\omega}_o - \bar{\boldsymbol{\Omega}}) \qquad [16]$$

correctly to terms of the first order in ϕ.

The constitutive relation [8] for the symmetric portion of the deviatoric stress corresponds to that given originally by Einstein (1, 2). The constitutive relation for the antisymmetric stress is given independently by Afanas'ev and Nikolaevskii (15, 16) and Brenner (5). This relation is intimately related to the angular momentum equation [2] for the following reason. In dilute systems, where hydrodynamic interactions among the suspended particles are negligible, Faxén's law (17) furnishes the following expression for the quasistatic hydrodynamic couple exerted by the fluid on the ith particle:

$$\mathbf{L}_i = 8\pi\mu_o a^3 (\boldsymbol{\omega}_o - \boldsymbol{\Omega}_i) \qquad [17]$$

where a is the sphere radius and $\boldsymbol{\Omega}_i$ the angular velocity of the sphere. Upon neglecting inertial effects stemming from the moment of inertia of the particle, the net couple on the particle must vanish. This requires that $\mathbf{L}_i + \mathbf{L}_i^{(e)} = \mathbf{0}$, where $\mathbf{L}_i^{(e)}$ is the body couple exerted on the particle

from outside of the system by the action of the external field on the embedded dipole. If there are N particles contained in a volume V of the suspension, then the total external couple exerted on these particles (and, hence, on the entire contents of V) is $\sum_i \mathbf{L}_i^{(e)} = -N8\pi\mu_o a^3 (\boldsymbol{\omega}_o - \bar{\boldsymbol{\Omega}})$, where $\bar{\boldsymbol{\Omega}} = \sum_i \boldsymbol{\Omega}_i / N$. Since $N/V = \phi / \frac{4}{3}\pi a^3$, the external body couple per unit volume of suspension is thus $\mathbf{G} = -6\mu_o \phi (\boldsymbol{\omega}_o - \bar{\boldsymbol{\Omega}})$. On substitution into Eq. [2] we recover Eq. [16]. Balancing the hydrodynamic and external couples on each particle is therefore implicitly equivalent to satisfying the angular momentum equation [2]. Consequently, the latter need not be explicitly considered as an independent relation in the subsequent analysis. Rather, it will automatically be satisfied by balancing the couples on each particle.

III. RELATIONS BETWEEN THE MICROSCOPIC AND MACROSCOPIC VIEWS OF DIPOLAR SUSPENSIONS

The mean angular velocity $\bar{\Omega}$ may be calculated in dilute systems by focussing attention on the behavior of isolated particles. This will ultimately yield $\bar{\Omega}$ correct to terms of $O(1)$ in ϕ, and hence will furnish $T_\times$ to $O(\phi)$ via Eq. [16]. Let d_i be the vector drawn from the geometric center of sphere i to its center of mass, and put $d_i = e_i\, d$. Here, $d = |d_i|$ is the same for all particles and e_i is a unit vector locked into the ith sphere. The instantaneous orientation of the particle may be specified by giving the orientation of e_i relative to a set of Cartesian axes fixed in space. This vector may conveniently be regarded as a radial unit vector in a system of spherical polar coordinates.

The gravitational body couple exerted on the mass of the ith sphere is

$$L_i^{(e)} = \rho \frac{4\pi}{3} a^3 d_i \times g \qquad [18]$$

The latter therefore depends on the orientation e_i of the particle relative to the direction of the gravity field. In conjunction with Eq. [17], the condition that the particle experience no net couple at each instant requires that

$$\Omega = \omega_o + \lambda\, \omega_o\, e \times \hat{g} \qquad [19]$$

where the subscript i has been dropped for brevity. Here, $\omega_o = |\omega_o|$; $\hat{g} = g/g$ is a unit vector parallel to gravity, and

$$\lambda = \frac{\rho g d}{6\mu_o\, \omega_o} \qquad [20]$$

is a dimensionless constant specifying the relative strengths of the dipolar and hydrodynamic couples. Since the vector e is locked into the particle, its time rate of change as measured by a space-fixed observer is

$$\frac{de}{dt} = \Omega \times e \qquad [21]$$

The instantaneous orientation of each particle is thus governed by the following nonlinear differential equation:

$$\frac{de}{dt} = \omega_o \left[\hat{\omega}_o \times e + \lambda\, (\hat{g} - e\, e \cdot \hat{g}) \right] \qquad [22]$$

in which $\hat{\omega}_o = \omega_o/\omega_o$ is a unit vector parallel to the local unperturbed vorticity vector. The nonlinearity arises from the quadratic term in e.

Equation [22] has been solved by Brenner (5) for the special cases of $\hat{\omega}_o$ colinear and perpendicular, respectively, to $\hat{g}$, and by Hall and Busenberg (4)[2] in the general case where the angle $\gamma = \cos^{-1}(\hat{\omega}_o \cdot \hat{g})$ $(0 \le \gamma \le \pi)$ between the directions of the vorticity and gravity vectors may be arbitrary. The two independent results are concordant for the cases $\gamma = 0$, $\pi/2$, and π considered by both. The solution $e \equiv e\{t\}$ of the differential equation depends, in general, upon the initial orientation of the particle at $t = 0$, and upon the parameters γ and λ.

Excluding the special case where $\gamma = \pi/2$ and $0 \le \lambda < 1$, which is discussed separately in the Appendix, Hall and Busenberg have shown, by an ingenious application of the Poincare-Bendixson stability theorem (18), that in all other circumstances the particle ultimately achieves a unique, stable, terminal orientation, corresponding to the situation in which $de/dt = 0$; that is, irrespective of its initial orientation, as $t \to \infty$ the vector e tends to a definite orientation relative to axes fixed in space. From

Eq. [22] the orientation $\mathbf{e}_s \equiv \mathbf{e}_s\left\{\hat{\boldsymbol{\omega}}_o, \hat{\mathbf{g}}, \lambda\right\}$ corresponding to this terminal state may be obtained by solving the equation

$$\hat{\boldsymbol{\omega}}_o \times \mathbf{e}_s = -\lambda\,(\hat{\mathbf{g}} - \mathbf{e}_s\mathbf{e}_s \cdot \hat{\mathbf{g}})$$

as follows: Upon squaring both sides of the above, one obtains $\sin\theta_s = \lambda \sin\xi_s$ where $\theta_s = \cos^{-1}(\mathbf{e}_s \cdot \hat{\boldsymbol{\omega}}_o)$ and $\xi_s = \cos^{-1}(\mathbf{e}_s \cdot \hat{\mathbf{g}})$. Furthermore, dot multiplying both sides of the above by $\hat{\boldsymbol{\omega}}_o$ yields $\cos\theta_s \cos\xi_s = \cos\gamma$. These two equations may be solved simultaneously for the angles θ_s and ξ_s. In this manner one obtains[3] (cf. Fig. 1)

$$\mathbf{e}_s = \hat{\mathbf{i}}_1 \sin\theta_s \cos\phi_s + \hat{\mathbf{i}}_2 \sin\theta_s \sin\phi_s + \hat{\mathbf{i}}_3 \cos\theta_s$$

where

$$\hat{\mathbf{i}}_1 = \frac{(\hat{\boldsymbol{\omega}}_o \times \hat{\mathbf{g}}) \times \hat{\boldsymbol{\omega}}_o}{\sin\gamma} \;;\; \hat{\mathbf{i}}_2 = \frac{\hat{\boldsymbol{\omega}}_o \times \hat{\mathbf{g}}}{\sin\gamma} \;;\; \hat{\mathbf{i}}_3 = \hat{\boldsymbol{\omega}}_o$$

are unit vectors in a right-handed system of Cartesian coordinates. Equivalently, since $\hat{\mathbf{g}} = \hat{\mathbf{i}}_1 \sin\gamma + \hat{\mathbf{i}}_3 \cos\gamma$, we obtain

$$\mathbf{e}_s = \hat{\boldsymbol{\omega}}_o \cos\theta_s \sin^2\phi_s + \hat{\mathbf{g}}\,\frac{\sin\theta_s \cos\phi_s}{\sin\gamma} + \hat{\boldsymbol{\omega}}_o \times \hat{\mathbf{g}}\,\frac{\sin\theta_s \sin\phi_s}{\sin\gamma} \qquad [23]$$

In these expressions,

$$\sin\theta_s = \left\{\frac{1}{2}(1+\lambda^2) - \left[\frac{1}{4}(1+\lambda^2)^2 - \lambda^2 \sin^2\gamma\right]^{1/2}\right\}^{1/2} \qquad [24]$$

and

$$\sin\phi_s = \frac{\sin\theta_s}{\lambda \sin\gamma} \qquad [25]$$

in which $0 \le \theta_s \le \pi$ and $0 < \phi_s < \pi/2$. These angles are shown in Fig. 1. Of the two possible values of θ_s satisfying Eq. [24] only that value of θ_s lying in the same quadrant as γ is to be selected.

That the orientation given by Eq. [23] is stable to <u>small</u> disturbances may be confirmed by a linearized stability analysis involving a small perturbation $\mathbf{e}'$ (perpendicular to $\mathbf{e}_s$) from the orientation $\mathbf{e}_s$. Employing Eq. [22], the two eigenvalues of the resulting stability matrix are found to be $-\mathbf{e}_s \cdot \hat{\mathbf{g}} \pm i\,\lambda\,\mathbf{e}_s \cdot \hat{\boldsymbol{\omega}}_o$ where $i = \sqrt{-1}$. Stability requires that the real portion of this be negative, i.e. that $\mathbf{e}_s \cdot \hat{\mathbf{g}} > 0$. Since $\mathbf{e}_s \cdot \hat{\mathbf{g}} = \cos\gamma/\cos\theta_s$, it is this condition which requires that θ_s lie in the same quadrant as does γ. Moreover, Eq. [25] and Footnote 4 show that $\sin\phi_s$ and $\cos\phi_s$ must both be positive (i.e., $\hat{\mathbf{i}}_1 \cdot \mathbf{e}_s > 0$ and $\hat{\mathbf{i}}_2 \cdot \mathbf{e}_s > 0$) and, consequently, that ϕ_s must lie in the first quadrant, as already stated.

More generally, Hall and Busenberg's analysis shows that the orientation described by Eq. [23] is stable against disturbances of arbitrary amplitude and orientation.

The rotation of the sphere in its terminal orientation is simple to describe. Since $d\mathbf{e}/dt = \mathbf{0}$, it follows from Eq. [21] that, in its terminal state, $\boldsymbol{\Omega}$ is colinear with $\mathbf{e}_s$, so that the sphere rotates about the $\mathbf{e}_s$ axis. Upon setting $\boldsymbol{\Omega} = \mathbf{e}_s\,\Omega$ it follows upon dot multiplying Eq. [19] by $\mathbf{e}_s$ that $\Omega = \boldsymbol{\omega}_o \cdot \mathbf{e}_s$. Hence, from Eq. [23][4] (see also Fig. 1),

$$\boldsymbol{\Omega} = \mathbf{e}_s\,\omega_o \cos\theta_s \qquad [26]$$

For the rotational "slip velocity" we have $\boldsymbol{\omega}_o - \boldsymbol{\Omega} = (\mathbf{I} - \mathbf{e}_s\mathbf{e}_s) \cdot \boldsymbol{\omega}_o$, corresponding to that component of $\boldsymbol{\omega}_o$ lying perpendicular to $\mathbf{e}_s$.

In the present circumstances, all particles contained in a region where ω_o is sensibly constant will possess the same e_s value and rotate with the same angular velocity Ω. Hence, $\bar{\Omega} = \Omega$, where Ω is given by Eq. [26]. It follows from Eqs. [16], [12] and [26] that

$$T_\times = 6\phi\,\mu_o\,\omega_o\,(\hat{\omega}_o - e_s \cos\theta_s) \qquad [27]$$

so that the deviatoric stress is given to the first order in the volume fraction ϕ by the expression

$$T = \mu\,[\nabla v + (\nabla v)^\dagger] + 3\phi\,\mu_o\,\omega_o\,\epsilon \cdot (\hat{\omega}_o - e_s \cos\theta_s) \qquad [28]$$

Dot multiplication of Eq. [27] by Ω shows that $T_\times$ is perpendicular to $\bar{\Omega}$, and hence that G is perpendicular to $\bar{\Omega}$. Physically, this means that no work is done by the external field acting upon the suspension. This conclusion is plausible; since a particle rotates about an axis which passes through both its centers of mass and buoyancy, it does not undergo any change in its orientational potential energy, $-(4\pi/3)\,a^3\,\rho\,d_i \cdot g$, as it rotates. That is, the latter remains constant during the rotation.

The preceding expressions simplify considerably in the "strong field" limit, $\lambda \to \infty^5$. In this case $e_s \to \hat{g}$, $\theta_s \to \gamma$, and $\phi_s \to 0$. These make

$$\Omega = \hat{g}\,\omega_o \cos\gamma \qquad [29]$$

or, equivalently, $\Omega = \hat{g}\,\hat{g} \cdot \omega_o$. The physical statement of this relation is that each sphere rotates about an axis colinear with the direction of the gravity field, the magnitude of its angular velocity being equal to the projection of the vorticity vector onto the direction of gravity.

Thus if we write $\omega_o = \omega_\parallel^o + \omega_\perp^o$, where $\omega_\parallel^o = \hat{g}\,\hat{g} \cdot \omega_o$ and $\omega_\perp^o = (I - \hat{g}\,\hat{g}) \cdot \omega_o \equiv \hat{g} \times (\omega_o \times \hat{g})$ are the respective vector components of ω_o parallel and perpendicular to gravity, then $\Omega = \omega_\parallel^o$. Moreover, we note in the strong field limit that $T_\times = 6\phi\,\mu_o\,\omega_\perp^o$, so that this axial vector is perpendicular to the gravity field.

From Eqs. [3] and [5]-[9] the dynamical and kinematical equations governing the macroscopic motion of the suspension are

$$-\nabla p + \mu\,\nabla^2 v = \frac{1}{2}\nabla \times T_\times \qquad [30]$$

$$\nabla \cdot v = 0 \qquad [31]$$

In accordance with Eqs. [27] and [23], the vector $T_\times$ may be regarded as a known function whenever the undisturbed motion (v_o, p_o) is known. Now the dynamical equation for the slow motion of a homogeneous Newtonian fluid in an external field of force is

$$-\nabla p + \mu\,\nabla^2 v + F = 0$$

where F is the external force per unit volume exerted on the fluid. It follows on comparison that Eq. [30] may be physically interpreted in terms of the existence of a prescribed nonconservative external force density possessing a vector potential, viz, $F = -\frac{1}{2}\nabla \times T_\times$. Indeed, this interpretation may be utilized to generate solutions of Eqs. [30]-[31], since the theory of Stokes equations in the presence of external force fields (19) shows that a particular integral of Eqs. [30]-[31] is

$$\underset{\sim}{v}(\underset{\sim}{r}) = - \frac{1}{8\pi\mu} \nabla \times \int_{V'} \frac{\underset{\sim}{T}_\times (\underset{\sim}{r}')}{|\underset{\sim}{r} - \underset{\sim}{r}'|} d^3 \underset{\sim}{r}' \quad [32a]$$

$$p(\underset{\sim}{r}) = \text{constant} \quad [32b]$$

to which may be added any solution of the homogeneous Stokes equations so as to satisfy the specified boundary conditions.

IV. COUETTE FLOW

Consider a simple shearing flow taking place in the x - y plane between two flat plates, x = 0 and x = h, the lower plate being held at rest and the upper plate moving parallel to itself with velocity U. In the absence of suspended particles the flow field is

$$\underset{\sim}{v}_o = \hat{\underset{\sim}{y}}\, Gx, \qquad p_o = \text{constant}$$

where $\hat{\underset{\sim}{x}}, \hat{\underset{\sim}{y}}, \hat{\underset{\sim}{z}}$ are unit vectors parallel to the coordinate axes and G = U/h is the shear rate. This gives $\underset{\sim}{\omega}_o = \hat{\underset{\sim}{z}}\, G/2$ whence $\hat{\underset{\sim}{\omega}}_o = \hat{\underset{\sim}{z}}$ and $\omega_o = G/2$. Since $\underset{\sim}{\omega}_o$ is constant throughout the fluid, then so is $\underset{\sim}{e}_s$ and, hence, $\underset{\sim}{T}_\times$ (provided that ϕ is uniform throughout the suspension). This makes $\nabla \times \underset{\sim}{T}_\times = \underset{\sim}{0}$, whence it follows that for the same boundary conditions the solution of Eqs. [30]-[31] is

$$\underset{\sim}{v} = \hat{\underset{\sim}{y}}\, Gx, \qquad p = \text{constant}$$

Upon substitution into Eq. [28] we find that the components of the deviatoric stress are

$$T_{xx} = T_{yy} = T_{zz} = 0 \quad [33a]$$

$$T_{xy} = \mu_o G \left(1 + \frac{5}{2}\phi + \frac{3}{2}\phi \sin^2 \theta_s\right) \quad [33b]$$

$$T_{yx} = \mu_o G \left(1 + \frac{5}{2}\phi - \frac{3}{2}\phi \sin^2 \theta_s\right) \quad [33c]$$

$$T_{yz} = - T_{zy} = - \frac{3}{2}\phi\, \mu_o G \csc\gamma \sin\Theta \sin\theta_s \cos\theta_s \cos(\phi_s + \Phi) \quad [33d]$$

$$T_{zx} = - T_{xz} = - \frac{3}{2} \phi \mu_o G \csc \gamma \sin \Theta \sin \theta_s \cos \theta_s \sin (\phi_s + \Phi) \qquad [33e]$$

where Θ and Φ are the spherical polar angles corresponding to the resolution of the gravity vector along the x, y, z axes, and are defined by the relation

$$\hat{g} = \hat{x} \sin \Theta \cos \Phi + \hat{y} \sin \Theta \sin \Phi + \hat{z} \cos \Theta \qquad [34]$$

where $0 \leq \Theta \leq \pi$ and $0 \leq \Phi < 2\pi$.

The apparent viscosity η of the suspension is defined in terms of the force dF_y in the y direction exerted on the surface element dS_x lying in the plane x = constant. It is defined by the relation $dF_y = \eta G dS_x$. Since, from the definition of stress, $dF_y = dS_x P_{xy}$, and since $P_{xy} = T_{xy}$, Eq. [33b] then yields

$$\eta = \mu_o (1 + \frac{5}{2} \phi + \frac{3}{2} \phi \sin^2 \theta_s) \qquad [35]$$

This agrees exactly with the expression obtained by Hall and Busenberg via scalar arguments based on the additional rate of mechanical energy dissipation[6] incurred by the addition of particles to the flowing fluid. In their analysis they make no reference to the antisymmetric state of stress existing in the suspension, nor does their analysis suggest the existence of the complex system of stresses represented by Eqs. [33c]-[33e]. For example, there exists a force on the plane x = constant in the z direction given by $dF_z = dS_x T_{xz}$. This force is perpendicular to the x - y plane despite the two-dimensional character of the flow.

The stress system given by Eqs. [33] simplifies considerably in the "strong field" limit, $\lambda \to \infty$. Here we find that

$$T_{xx} = T_{yy} = T_{zz} = 0$$

$$T_{xy} = \mu_o G (1 + \frac{5}{2} \phi + \frac{3}{2} \phi \sin^2 \gamma)$$

$$T_{yx} = \mu_o G (1 + \frac{5}{2} \phi - \frac{3}{2} \phi \sin^2 \gamma)$$

$$T_{yz} = - T_{zy} = - \frac{3}{2} \phi \mu_o G g_x \cos \gamma$$

$$T_{zx} = - T_{xz} = - \frac{3}{2} \phi \mu_o G g_y \cos \gamma$$

where (g_x, g_y, g_z) are the components of g along the x, y and z directions. The apparent viscosity in this case is

$$\eta = \mu_o (1 + \frac{5}{2} \phi + \frac{3}{2} \phi \sin^2 \gamma) \qquad [36]$$

as was first pointed out by Hall and Busenberg. The anisotropy of the apparent viscosity with respect to the relative directions of the external field and the vorticity vector is clearly evident.

V. MORE GENERAL FLOW FIELDS
($\gamma = \pi/2$, $\lambda \geq 1$)

The apparent viscosity is not generally an intrinsic property of the suspension alone, but depends rather on the physical details of the apparatus in which the rheological experiment is conducted. However, as we now show, in experimental configurations where the unperturbed flow is such that the vorticity vector $\underset{\sim}{\omega}_o$ is everywhere perpendicular to the gravity field $\underset{\sim}{g}$, and for situations where $\lambda \geq 1$ throughout the flow field, the apparent viscosity will be independent of the nature of the experiment performed. As important examples of flow fields where $\underset{\sim}{\omega}_o$ is perpendicular to $\underset{\sim}{g}$ at each point we have: (i) all two-dimensional flows taking place in vertical planes (e.g., two-dimensional Couette and Poiseuille flows between vertically separated flat plates); (ii) all axisymmetric three-dimensional flows for which the symmetry axis is vertical (e.g., Poiseuille flow in a vertical circular tube; a falling-ball viscometer).

In these circumstances, $\gamma = \pi/2$, so that, from Eqs. [24] and [25], $\theta_s = \pi/2$ and $\phi_s = \sin^{-1}(1/\lambda)$ $(0 < \phi_s < \pi/2)$. Equation [23] gives for this case,

$$\underset{\sim}{e}_s = \hat{\underset{\sim}{g}} \sqrt{1 - \lambda^{-2}} + \lambda^{-1} \hat{\underset{\sim}{\omega}}_o \times \hat{\underset{\sim}{g}} \qquad [37]$$

which is everywhere perpendicular to $\underset{\sim}{\omega}_o$. Furthermore, Eq. [26] shows that $\underset{\sim}{\Omega} = \underset{\sim}{0}$, and hence that $\overline{\underset{\sim}{\Omega}} = \underset{\sim}{0}$. The particles are therefore completely immobilized, unable to rotate at all. It thus follows from Eq. [16] that

$$\underset{\sim}{T}_\times = 6\phi \mu_o \underset{\sim}{\omega}_o \qquad [38]$$

In the absence of the particles the equations of motion are

$$-\nabla p_o + \mu_o \nabla^2 \underset{\sim}{v}_o = \underset{\sim}{0} \qquad [39a]$$

$$\nabla \cdot \underset{\sim}{v}_o = 0 \qquad [39b]$$

Consequently,

$$\begin{aligned} \nabla \times \underset{\sim}{T}_\times &= 3\phi \mu_o \nabla \times (\nabla \times \underset{\sim}{v}_o) \\ &= -3\phi \mu_o \nabla^2 \underset{\sim}{v}_o \\ &= -3\phi \nabla p_o \end{aligned} \qquad [40]$$

Equations [30]-[31] governing the flow of the suspension therefore become

$$-\nabla P + \mu_o \nabla^2 \underset{\sim}{v} = \underset{\sim}{0} \qquad [41a]$$

$$\nabla \cdot \underset{\sim}{v} = 0 \qquad [41b]$$

where

$$P = \frac{\mu_o}{\mu} \left(p - \frac{3}{2} \phi p_o \right) \qquad [42]$$

For the same boundary conditions, Eqs. [41] possess the same solution as do Eqs. [39], whence $\underset{\sim}{v} = \underset{\sim}{v}_o$ and $P = p_o$. On substituting the latter into Eq. [42] and taking account of Eq. [11], it follows that $p = (1 + 4\phi)\, p_o$.

To terms of the first order in ϕ the above analysis shows that the macroscopic velocity field describing the suspension flow is the same as for the undisturbed case, but that the pressure is uniformly increased by a factor of $1 + 4\phi$ at each point of the fluid. The pressure and velocity fields for the suspension are, therefore, indistinguishable from those for a Newtonian fluid whose viscosity is $\mu_o (1 + 4\phi)$. On this basis, an experimentalist would interpret his results by assigning to the suspension a viscosity

$$\eta = \mu_o (1 + 4\phi) \qquad [43]$$

This is, of course, precisely the result we recover from Eq. [35] by setting $\theta_s = \pi/2$.

An experimenter who measured only the local velocity and pressure fields would conclude that the suspension was strictly Newtonian. On the other hand, a more open-minded and thoughtful investigator might go further and measure the stresses too. Much to his surprise he would discover that the fluid was not Newtonian, but rather obeyed the constitutive relation

$$\underset{\approx}{T} = \eta \left[\nabla \underset{\sim}{v} + (\nabla \underset{\sim}{v})^{\dagger} \right] - 3\mu_o \phi \, (\nabla \underset{\sim}{v})^{\dagger} \qquad [44]$$

where η is given by Eq. [43]. This result would surely prove perplexing to him since gravity plays the role of a hidden variable, and fails to manifest itself macroscopically in any of the conventional ways. Indeed, the strength of the gravity field does not appear in the velocity, pressure, or deviatoric stress fields, so that by varying it over a narrow range no alteration in the phenomenon would be observed.

To gain some idea of the value of λ in practical circumstances consider a suspension dispersed in water ($\rho = 1$ g cm^{-3}, $\mu_o = 1$ cP) at a shear rate of $G = 1$ sec^{-1} ($\omega_o = 0.5$ sec^{-1}), and a particle radius of $a = 10$ microns in an ordinary gravity field ($g = 980$ cm sec^{-2}). For a 3 percent displacement of the center of mass ($d/a = 3 \times 10^{-2}$) this yields $\lambda = 1.0$. In this calculation $\omega_o a^2 \rho/\mu_o = 0.5 \times 10^{-4} \ll 1$, whence the particle Reynolds number is indeed small. Furthermore, according to the Stokes-Einstein theory the rotary diffusion coefficient is $D_r = kT/8\pi \mu_o a^3$, where $k = 1.38 \times 10^{-16}$ g cm^2 sec^{-2} ($^oK)^{-1}$ is Boltzmann's constant and T is the Kelvin temperature. At room temperature this gives $D_r/G = 1.6 \times 10^{-4} \ll 1$, confirming that rotary Brownian motion effects are small.

For two-dimensional Poiseuille flow at mean velocity U between flat plates separated by a distance h the vorticity at the wall is $\omega_o = 3U/h$. The vorticity at any other point in the gap is less than this value, and vanishes at the midplane. In this situation the condition that $\lambda \geq 1$ at every point of the fluid will be met if $\rho\, dgh/\mu_o U \geq 18$.

For the falling-ball viscometer case the vorticity has a maximum magnitude of $\omega_o = 3U/2b$, which is achieved on the sphere surface (U = sphere velocity; b = sphere radius). Hence, the condition $\lambda \geq 1$ requires that $\rho\, dgb/\mu_o U \geq 9$.