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Nonlinear effects in the rheology of dilute suspensions

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An analysis is presented of the deformation of a solid-like, viscoelastic sphere suspended in the infinite Stokesian flow field of a Newtonian fluid undergoing an arbitrary time-dependent homogeneous deformation far from the particle. The results of the analysis are then used to deduce the macroscopic rheological behaviour of a dilute monodisperse suspension of slightly deformable spheres.

Even though inertial effects and second-order terms in the particle deformation are neglected, it is found that non-linear rheological effects can arise, because of the interaction between the deformed particle and the flow. As a consequence, the rheological relation obtained here differs from those presented earlier by Fröhlich & Sack (1946) and by Oldroyd (1955) through the appearance of certain terms which are non-linear in the deformation rate.

When the suspended particles are purely elastic in their behaviour the rheological equation presented here reduces for certain flows to a special case of Oldroyd's (1958) phenomenological model, with material constants which can be directly related to suspension properties.

1. Introduction

The problem of deducing theoretically the macroscopic rheological behaviour of microscopically heterogeneous fluids has received considerable attention, dating from the celebrated early work of Einstein (1906, 1911) on the viscosity of dilute suspensions of solid spheres in Newtonian liquids.

Owing to their particular relevance to an understanding of elastic effects in emulsions as well as solutions of deformable macromolecules, mathematical models for suspensions of deformable elastic particles have been the subject of several works. Following the work of Fröhlich & Sack (1946) on the irrotational flow of dilute suspensions of elastic spheres, Oldroyd (1953, 1955) treated suspensions of solid and liquid spheres exhibiting complex interfacial effects. Cerf (1951) has also considered suspensions of solid viscoelastic spheres, in connexion with a study of flow birefringence of polymer solutions, but he did not analyse the rheology of such suspensions in great detail. More recently, Giesekus (1962) has made both experimental and theoretical studies of deformable particles in certain types of shear fields, but the theoretical studies deal mainly with simplified hydrodynamic models.

The present study was undertaken for two reasons. First of all, it was desired to determine the effects of shear-induced particle deformation and rotation on suspension behaviour for situations where Brownian effects are absent. In the previous works of Fröhlich & Sack (1946) and of Oldroyd (1953), dealing with spherical particles or drops, the rate of particle deformation is accounted for by a matching of velocities at a fluid–particle interface, but both these authors assume that the deviation of the particle from a spherical shape is sufficiently small so that interfacial matching conditions can be imposed at the surface of the original sphere. While this approximation appears to be exact to terms of the first order in the *rates* of deformation of both the fluid and the particle, the present analysis will show that the ellipticity of the deformed sphere gives rise to additional stresses at the particle surface, which involve bilinear terms in the fluid deformation rate and the particle *strain*. As will be shown, these terms can give rise to non-linear effects in the suspension behaviour, even if terms of the second order in particle strain are considered negligible.

A second objective of the present work is to present a derivation of the macroscopic rheology of dilute suspensions which proceeds directly from a knowledge of individual particle behaviour at infinite dilution. The present technique appears to be more convenient in general than the 'cavity' technique employed by Fröhlich & Sack (1946) and later by Oldroyd (1953), since it requires no *a priori* assumption as to the form of the rheological equation being sought.

1.1. *Motion of a single viscoelastic sphere in a homogeneous velocity-gradient field*

We wish to derive here the equations describing the simultaneous rotation and deformation of a single viscoelastic particle placed in a time-dependent flow field of an incompressible Newtonian fluid having a homogeneous, i.e. spatially uniform, velocity gradient far from the particle.

We shall assume that the particle is composed of a homogeneous and isotropic, solid-like material and that, in its undeformed or stress-free state, the particle is spherical in shape. Furthermore, we shall suppose that the rheological constitutive equation of the solid is known so that, once the stress history is completely specified over the surface of the particle, its instantaneous deformation can in principle be determined.

The problem consists therefore of determining the motion and deformation of the particle when it is placed in an infinite flow field whose (time-dependent) velocity distribution, the 'undisturbed' flow, is prescribed far from the particle. This is a well-known type of problem which involves the simultaneous solution of the equations of motion for the fluid and for the particle, with a matching of the local stress and velocity (or displacement) at the particle surface. Since we shall postulate here the absence of interfacial effects, this matching can be imposed on all the components of the velocity and surface-stress vectors.

As regards the forces acting in our system, we shall assume that the density of the particles is the same as that of the fluid, or that buoyancy forces are otherwise negligible, and, as an approximation, that inertial forces are everywhere negligible beside elastic and viscous forces. In this case, the force balance becomes

$$\nabla . \mathbf{T} = 0 \tag{1.1}$$

in the regions occupied by the fluid and the particle; and, as a consequence, the equations of motion for the fluid reduce to the well-known Stokes equations:

$$\mu\nabla^2\mathbf{v} = \boldsymbol{\nabla}p, \quad \boldsymbol{\nabla}.\mathbf{v} = 0, \tag{1.2}$$

where $\mathbf{T} = \mathbf{T}(\mathbf{r},t)$ is the stress tensor, $\mathbf{v} = \mathbf{v}(\mathbf{r},t)$ and $p = p(\mathbf{r},t)$ are the vector velocity field and the pressure field in the fluid, and μ is the fluid viscosity, with $\mathbf{r}$ and t denoting, respectively, the position vector and time. In a later paragraph we shall offer some criteria for validity of our omission of inertial effects.

Once the appropriate rheological equation for the particle is specified a second equation of motion, the analogue of (1.1), can be written down for the region occupied by the particle. Letting primed quantities refer to this region and matching velocity and stress at the surface of the particle $\mathscr{S}'(t)$, say, we have then that

$$\left.\begin{aligned} \mathbf{v}(\mathbf{r},t) &= \mathbf{v}'(\mathbf{r},t) \\ \text{and} \quad \mathbf{T}(\mathbf{r},t).\mathbf{n} &= \mathbf{T}'(\mathbf{r},t).\mathbf{n} \end{aligned}\right\} \text{ for } \mathbf{r} \text{ on } \mathscr{S}'(t), \tag{1.3}$$

where $\mathbf{n}$ denotes the unit normal to $\mathscr{S}'$. As is done above, we shall employ Gibbs' dyadic notation in the following analysis, with vectors and tensors denoted by bold face lower- and upper-case letters, respectively.

Now, one further condition on fluid velocity, far from the particle, will suffice in principle for determination of the motion. Letting $\mathbf{r}$ denote the position vector referred, say, to the mass centre of the particle, we shall take this remaining condition to be

$$\mathbf{v} \to \mathbf{v}^{(0)} = \boldsymbol{\Gamma}^{(0)}.\mathbf{r}, \quad \text{for} \quad r \to \infty, \tag{1.4}$$

where $r = |\mathbf{r}|$ and $\boldsymbol{\Gamma}^{(0)}(t)$ is a velocity-gradient tensor, $\mathbf{v}^{(0)}$ denoting the 'undisturbed' flow velocity.

Having thus posed the problem, at least up to a specification of the rheological equation for the particle, we shall now introduce an hypothesis, based largely on the previous work of Fröhlich & Sack (1946) and Cerf (1951), which will greatly facilitate its solution. In particular, if the material of the particle is homogeneous and isotropic and if its instantaneous strain depends only on the past history of stress, we are led to suppose that, in the absence of surface tension or other interfacial effects, the motion of the original sphere will consist of purely *homogeneous* deformation, in which case the velocity-gradient field is homogeneous inside the particle. This supposition can be justified in a heuristic fashion by noting, first of all, that under a homogeneous deformation a spherical or ellipsoidal particle will be transformed at any instant into an ellipsoid; furthermore, one can deduce from classical work of Jeffery (1922) that, for the motion of *rigid* solid ellipsoids in homogeneous velocity-gradient fields, the fluid stress on the surface of an ellipsoid gives rise to a homogeneous stress field in its interior. Provided then, in the present case, that a homogeneous stress history gives rise to homogeneous strain in the particle, it remains only to show that Jeffery's result carries over to deformable spheres or ellipsoids. Indeed this is possible and, moreover, a complete solution to the present problem can be constructed by a slight modification of Jeffery's solution, as we now show. It should be noted that Cerf (1951) has presented a similar, but less complete, argument.

1.2. *Extension of Jeffery's result to deformable ellipsoids*

In order to construct a solution to the present problem from Jeffery's solution for the Stokes flow outside a rigid ellipsoid, we shall proceed as follows: we assume first that a particle of the type considered here undergoes a homogeneous deformation and, hence, that it always has an ellipsoidal shape. Next, we modify Jeffery's solution for the flow field outside a rigid ellipsoid in such a way as to obtain the exact solution for the Stokes flow outside a homogeneously deforming ellipsoid. Finally, we verify that there exists a homogeneous stress field $\mathbf{T}'(t)$, say, inside the deforming ellipsoid which matches with the fluid stresses on its surface. Hence, it follows that we shall have found at least one solution to the problem at hand, provided there exists a homogeneous particle deformation which is rheologically compatible with the homogeneous stress $\mathbf{T}'$. We shall be able to formulate this latter requirement more concisely after the following discussion.

We proceed, then, by assuming that the strain inside the particle is homogeneous; as a consequence, we can express the velocity $\mathbf{v}'$ in the first equation of (1.3) by

$$\mathbf{v}' = \mathbf{\Gamma}'.\mathbf{r}$$

where $\mathbf{\Gamma}' = \mathbf{\Gamma}'(t)$, independent of position, is the velocity-gradient tensor inside the particle. Next, decomposing $\mathbf{\Gamma}'$ into a symmetric strain-rate tensor $\mathbf{E}'$ and an antisymmetric 'vorticity' tensor $\mathbf{\Omega}'$, we have

$$\mathbf{\Gamma}' = \mathbf{E}' + \mathbf{\Omega}', \tag{1.5}$$

and we recall that the angular-velocity vector of the particle is simply $-\operatorname{Vec}\mathbf{\Omega}'$. The first equation in (1.3) can now be replaced by

$$\mathbf{v} = (\mathbf{E}' + \mathbf{\Omega}').\mathbf{r}, \quad \text{for } \mathbf{r} \text{ on } \mathscr{S}'(t), \tag{1.6}$$

where $\mathscr{S}'(t)$ is an ellipsoidal surface. It follows then that, if $\mathbf{E}'$ and $\mathbf{\Omega}'$ were specified, the fluid motion outside the particle could be determined from (1.2), (1.4) and (1.6).

In contrast to the foregoing problem statement, the problem treated by Jeffery requires the solution of (1.2) subject to the conditions

$$\left.\begin{aligned} \mathbf{v} &= \mathbf{\Omega}'.\mathbf{r}, \quad \text{for } \mathbf{r} \text{ on } \mathscr{S}'(t), \\ \mathbf{v} &\to \mathbf{\Gamma}^{(0)}.\mathbf{r}, \quad \text{for } r \to \infty, \end{aligned}\right\} \tag{1.7}$$

where again $\mathscr{S}'(t)$ is an ellipsoidal surface, and where $\mathbf{\Omega}' = \mathbf{\Omega}'(t)$ and $\mathbf{\Gamma}^{(0)} = \mathbf{\Gamma}^{(0)}(t)$. These equations govern the motion of a rigid ellipsoid in the absence of any externally applied force, and as shown by Jeffery the solution to this problem permits determination of $\mathbf{\Omega}'$ and, hence, of the particle rotation, once $\mathbf{\Gamma}^{(0)}$ as well as any extraneous torque on the particle are specified.

Considering here the case of zero torque only, we denote Jeffery's solution for the fluid velocity, the pressure field, and the particle vorticity, respectively, by

$$\mathbf{v} = \mathbf{u}[\mathbf{\Gamma}^{(0)}, \mathbf{G}'; \mathbf{r}, t], \quad p = q[\mathbf{\Gamma}^{(0)}, \mathbf{G}'; \mathbf{r}, t], \quad \mathbf{\Omega}' = \mathbf{W}[\mathbf{\Gamma}^{(0)}, \mathbf{G}'; t], \tag{1.8}$$

where $\mathbf{G}' = \mathbf{G}'(t)$ is a symmetric second-order tensor (to be defined in (1.13)) with principal axes which determine the instantaneous orientation of the ellipsoidal surface $\mathscr{S}'$.

Without writing any of the quantities in (1.8) down explicitly at this point, we note that $\mathbf{u}, q$ and $\mathbf{W}$ are all linear functions of their first argument $\mathbf{\Gamma}^{(0)}$, which of course is a consequence of the linearity of (1.2) and (1.7). Because of this fact and owing to the absence of time derivatives in the problem at hand, it follows that the functions

$$\left.\begin{aligned} \mathbf{v} &= \mathbf{u}[\mathbf{\Gamma}^{(0)} - \mathbf{E}', \mathbf{G}'; \mathbf{r}, t] + \mathbf{E}'.\mathbf{r}, \\ p &= q[\mathbf{\Gamma}^{(0)} - \mathbf{E}', \mathbf{G}'; \mathbf{r}, t], \\ \mathbf{\Omega}' &= \mathbf{W}[\mathbf{\Gamma}^{(0)} - \mathbf{E}', \mathbf{G}'; t] \end{aligned}\right\} \tag{1.9}$$

will satisfy (1.2), (1.4) and (1.6) as well as the condition of zero torque, provided of course that the surface $\mathscr{S}'(t)$ in (1.7) is taken to coincide instantaneously with that in (1.6), and provided further that

$$\mathbf{\nabla}.(\mathbf{E}'.\mathbf{r}) \equiv \mathrm{tr}(\mathbf{E}') \equiv \mathbf{\nabla}.\mathbf{v}' = 0, \tag{1.10}$$

where 'tr' denotes the trace of a tensor. This latter condition corresponds to incompressibility of the particles, and we shall assume henceforth that the particles are indeed composed of an incompressible material.

Following Cerf, we observe now that Jeffery's equations for the fluid stress acting on the surface of a rigid ellipsoid can be expressed as $\mathbf{S}.\mathbf{n}$, where $\mathbf{n}$ is the unit normal and where

$$\mathbf{S} = \mathbf{S}[\mathbf{\Gamma}^{(0)}, \mathbf{G}'; t] \tag{1.11}$$

is a tensor independent of position on the surface and linear in $\mathbf{\Gamma}^{(0)}$. Hence, it follows readily by (1.1), by the second equation of (1.3), and by (1.9) that the fluid stresses on the surface of a homogeneously deforming ellipsoid produce in its interior a stress field which is given by the homogeneous tensor

$$\mathbf{T}' = \mathbf{S}[\mathbf{\Gamma}^{(0)} - \mathbf{E}', \mathbf{G}'; t] + 2\mu\mathbf{E}' \tag{1.12}$$

plus perhaps an additive field $\mathbf{T}''(\mathbf{r}, t)$, say. This latter field must satisfy (1.1) inside $\mathscr{S}'$ and the condition

$$\mathbf{T}''.\mathbf{n} = \mathbf{0}$$

on $\mathscr{S}'$. However, since the only homogeneous stress satisfying these conditions is the trivial one, $\mathbf{T}'' \equiv \mathbf{0}$, it is seen that (1.12) provides a unique expression for the *homogeneous* stress field necessary to balance the fluid stresses on the surface of a homogeneously deforming ellipsoid. Hence, by taking $\mathbf{T}'' \equiv \mathbf{0}$, thereby rejecting any extraneous non-homogeneous stress, we have found one possible solution for (1.2) to (1.4), provided that the rheological relation between $\mathbf{T}'(t)$ and $\mathbf{E}'(t)$ for the particle is compatible with (1.12). Otherwise stated, the rheological relation for the particle together with (1.12), $\mathbf{S}$ being given explicitly by Jeffery's result, represent two relations between the unknowns $\mathbf{T}'$ and $\mathbf{E}'$. The existence of a solution to these equations will guarantee the existence of a solution to the present problem, of the type assumed.

We have not been able to establish here the general restrictions on particle rheology which are necessary for the existence of a unique solution. Instead, we shall restrict ourselves to an investigation of the limiting form of (1.12) for small particle deformations, which will be presented in the following section, and then in §2.2 we shall consider a specific rheological model, for which it appears that a unique solution exists.

We should again point out that previous work on related problems would tend to support our hypothesis of homogeneous particle deformation in the absence of surface effects. In particular, the analysis of Fröhlich & Sack (1946) for purely elastic solid spheres indicates a homogeneous deformation of the spheres, a condition presupposed by Cerf (1951) in a similar problem. On the other hand, in Oldroyd's (1953) later, more comprehensive analysis involving viscoelastic spheres with interfacial tension, it is found that the motion of the particles consists in general of the sum of both a homogeneous and a non-homogeneous mode. However, for the special case of vanishing surface tension, one finds on closer inspection that, within the framework of the 'cavity' technique employed by Oldroyd, the magnitude of the non-homogeneous mode can be shown to be proportional to second-order terms in the volume fraction ϕ, say, of the particulate phase. Since, as Oldroyd indicates later in his analysis, the accuracy of the technique which he employed is limited to terms of order one in ϕ, all terms of the second order could have been discarded at an early stage in his analysis. Indeed, one finds from Oldroyd's work (equations (25) to (28)) that, for vanishing surface tension, a particle suspended in an infinite body of fluid (corresponding to $A = 0$ in Oldroyd's equations) would deform homogeneously ($A' = 0$ in the same equations). Thus, we are led to conclude that for vanishing surface tension the non-homogeneous motion is probably an artifact of the cavity technique itself.

Of course, in the case of a finite surface tension, a non-homogeneous particle deformation would ensue, giving rise to a 'circulatory' motion and to the associated stresses in the particle necessary to balance with surface forces. In this regard, it should be noted that, in the absence of surface tension, Oldroyd's equations would cease to apply once the particle deviates appreciably from spherical shape, whereas the analysis presented here could still remain valid, subject possibly to certain restrictions on the rheology of the particulate phase. Since we cannot offer *a priori* any rigorously defined restrictions, it would seem reasonable, as anticipated at the outset, to limit the discussion to 'solid-like' materials, characterized by a unique 'undeformed' state of zero stress. For, without surface tension, it becomes somewhat meaningless in general to speak of liquid droplets.

1.3. *Motion of a slightly deformable sphere*

We shall now restrict our attention to spherical particles with sufficient rigidity to ensure that their deformations are always small. In particular, and exactly as is done in the classical (linear) theory of elasticity, we shall assume that second- and higher-order terms in the components of the strain tensor are all negligible. The necessary conditions for small strains in the present problem will be stated more precisely below.

We now let a_0 denote the radius of the undeformed sphere and $a_i(t)$, $i = 1, 2, 3$, the semiprincipal axes of the ellipsoid resulting from its deformation. Denoting further the finite strain tensor by $\mathbf{C}'$, and the Cauchy–Green deformation tensor by $\mathbf{G}'$, we shall define the latter by taking its components to be

$$G'_{ij} = \begin{Bmatrix} (a_0/a_i)^2 & \text{for } i = j, \\ 0 & \text{for } i \neq j, \end{Bmatrix} \tag{1.13}$$

on an orthogonal Cartesian co-ordinate system $x_i (i = 1, 2, 3)$, chosen to coincide with the principal axes of deformation, while we define the former by taking

$$\mathbf{G}' = \mathbf{I} - 2\mathbf{C}' \tag{1.14}$$

on an arbitrary system. The equation of the (ellipsoidal) surface of the particle is merely that of the ellipsoid of the tensor $\mathbf{G}'$, i.e.

$$\sum_{i=1}^{3} \left(\frac{x_i}{a_i}\right)^2 = \frac{\mathbf{r}.\mathbf{G}'.\mathbf{r}}{a_0^2} = 1,$$

the last equality holding, of course, in any co-ordinate frame.

By differentiating the preceding relation with respect to t and by noting that

$$d\mathbf{r}/dt = \mathbf{v}' = \mathbf{\Gamma}'.\mathbf{r} \quad \text{for } \mathbf{r} \text{ on } \mathscr{S}'(t),$$

one has that
$$(d\mathbf{G}'/dt) + (\mathbf{\Gamma}')^{\dagger}.\mathbf{G}' + \mathbf{G}'.\mathbf{\Gamma}' = \mathbf{0},$$

or, in terms of $\mathbf{C}'$ and $\mathbf{E}'$, that

$$\mathbf{E}' = (d\mathbf{C}'/dt) + (\mathbf{\Gamma}')^{\dagger}.\mathbf{C}' + \mathbf{C}'.(\mathbf{\Gamma}'). \tag{1.15}$$

Here $(\mathbf{\Gamma}')^{\dagger}$ denotes the transpose or dyadic conjugate of $\mathbf{\Gamma}'$ which, by (1.5) is

$$(\mathbf{\Gamma}')^{\dagger} = \mathbf{E}' - \mathbf{\Omega}', \tag{1.16}$$

since $(\mathbf{E}')^{\dagger} = \mathbf{E}'$ and $(\mathbf{\Omega}')^{\dagger} = -\mathbf{\Omega}'$. Equation (1.15) will be recognized as the definition of $\mathbf{E}'$ in terms of a 'convected' derivative of $\mathbf{C}'$.

In order to express some of Jeffrey's results in the present notation, we recall that the components of $\mathbf{C}'$ on the axes of the ellipsoid are

$$C'_{ij} = \begin{cases} \frac{1}{2}[1-(a_0/a_i)^2] & \text{for } i = j, \\ 0 & \text{for } i \neq j, \end{cases}$$

$i, j = 1, 2, 3$. Then for the present purposes, we can replace the semiprincipal axes of the ellipsoid in Jeffery's paper by the functions of time, $a_i(t)$. However, by first restricting ourselves to the case of small strains, we have that

$$a_1/a_0 = 1 + C'_{11} + \tfrac{3}{2}C'^2_{11} + O(C'^3_{11}) \tag{1.17}$$

with similar equations for $i = 2, 3$. If then these expressions for $a_i(t)$, in terms of the $C_{ii}(t)$, are substituted into Jeffery's expression‡ for the fluid stress on the surface of a rigid ellipsoid, one finds after some algebra that the components of the stress tensor $\mathbf{S}$ in (1.12), as expressed on the axes of the ellipsoid, are

$$\left.\begin{aligned} S_{11} &= \mu[5E^{(0)}_{11}(1+\tfrac{6}{7}C'_{11}) + \tfrac{4}{7}[E^{(0)}_{22}(C'_{11}-C'_{22}) \\ &\qquad + E^{(0)}_{33}(C'_{33}-C'_{11})] + O(C'^2) + p^{(0)}, \\ S_{12} &= 5\mu E^{(0)}_{12}(1-\tfrac{3}{7}C'_{33}) + O(C'^2), \end{aligned}\right\} \tag{1.18}$$

with similar expressions for S'_{22}, S'_{23}, etc., obtained by cyclic permutation of the indices 1, 2, 3. Here $p^{(0)} = p^{(0)}(t)$ is the undisturbed pressure field far from the ellipsoid, $E^{(0)}_{ij} = E^{(0)}_{ij}(t)$ are the components of the deformation-rate tensor for the

‡ After correction of certain typographical errors, noted already by Giesekus (1962, footnote 28, p. 60) and by Cerf (1951).

undisturbed flow (again expressed on the axes of the ellipsoid), and $O(C'^2)$ denotes quantities which involve terms of the second order in C'_{11}, C'_{22} and C'_{33}. To derive the preceding relations from those given by Jeffery, we have made use of the fact that the condition for incompressibility of the solid sphere,

$$a_1 a_2 a_3 = a_0^3,$$

reduces for small C' to

$$C'_{11} + C'_{22} + C'_{33} - 2(C'_{11}C'_{22} + C'_{11}C'_{33} + C'_{22}C'_{33}) + O(C'^3) = 0. \tag{1.19}$$

In this regard we should note that it is necessary to retain the terms of the second order in (1.19) as well as those in (1.17) if one is to arrive at the correct expression for terms of $O(C')$ in (1.18). (The derivation of expansions for the integrals, denoted by $\alpha_0, \beta_0, \gamma_0, \ldots$, in Jeffery's paper, in terms of the C'_{11}, involves division by terms $O(C')$.)

Now, (1.18) can be expressed in dyadic notation by noting that, on the present co-ordinate system, it is merely

$$\left.\begin{aligned} \mathbf{S} &= \mu[5\mathbf{E}^{(0)} + \tfrac{15}{7}[\mathbf{E}^{(0)}.\mathbf{C}' + \mathbf{C}'.\mathbf{E}^{(0)}] + \tfrac{4}{7}(\mathbf{E}^{(0)}\colon \mathbf{C}')\,\mathbf{I}] - p^{(0)}\mathbf{I} + O(C'^2), \\ \mathbf{E}^{(0)} &= \tfrac{1}{2}[\mathbf{\Gamma}^{(0)} + (\mathbf{\Gamma}^{(0)})^{\dagger}], \end{aligned}\right\} \tag{1.20}$$

and

$$\mathbf{E}^{(0)}\colon \mathbf{C}' = \operatorname{tr}(\mathbf{E}^{(0)}.\mathbf{C}').$$

Also, $\mathbf{I}$ denotes the unit tensor and $\mathbf{E}^{(0)}$ is, of course, the deformation rate tensor for the undisturbed flow in Jeffery's problem. Stated in the form (1.20) the above relation must be valid now on any co-ordinate system. When (1.20) is substituted into (1.12), with the modification indicated there, one obtains the desired expression for the stress field $\mathbf{T}'$ inside the deforming ellipsoid of the present problem. However, since we are dealing here with incompressible, isotropic materials, it is necessary to consider only the deviatoric or 'extra' stress tensor, defined in general by

$$\mathbf{P} = \mathbf{T} + p\mathbf{I}, \tag{1.21}$$

where as before $\mathbf{T}$ is the stress tensor and

$$p = -\tfrac{1}{3}\operatorname{tr}\mathbf{T} \equiv -\tfrac{1}{3}\mathbf{T}\colon\mathbf{I} \tag{1.22}$$

is the 'mean' pressure. In terms of the deviatoric particle stress $\mathbf{P}'$, (1.20) and (1.12) give

$$\mathbf{P}'(t) = 5\mu[\mathbf{A} + \tfrac{3}{7}(\mathbf{A}.\mathbf{C}' + \mathbf{C}'.\mathbf{A}) - \tfrac{2}{7}(\mathbf{A}\colon\mathbf{C}')\,\mathbf{I}] + 2\mu\mathbf{E}' + O(C'^2), \tag{1.23}$$

where

$$\mathbf{A} = \mathbf{A}(t) = \mathbf{E}^{(0)}(t) - \mathbf{E}'(t) \tag{1.24}$$

and

$$\mathbf{C}' = \mathbf{C}'(t),$$

$\mathbf{E}'$ and $\mathbf{C}'$ being related by (1.15).

Now, (1.15) contains $\mathbf{\Omega}'(t)$, the (unknown) rotation tensor for the particle, but we can also derive equations for this tensor for Jeffery's results. In fact, one can show easily that Jeffery's formula for rotation in the absence of torque can be expressed in the present notation as

$$\Delta\mathbf{\Omega} + \mathbf{C}'.(\Delta\mathbf{\Omega}) + (\Delta\mathbf{\Omega}).\mathbf{C}' = \mathbf{E}^{(0)}.\mathbf{C}' - \mathbf{C}'.\mathbf{E}^{(0)}, \tag{1.25}$$

where

$$\Delta\mathbf{\Omega} = \mathbf{\Omega}' - \mathbf{\Omega}^{(0)}, \tag{1.26}$$

$\boldsymbol{\Omega}^{(0)}$ denoting the rotation tensor of the undisturbed flow. (Giesekus (1962) has put this result in a different form, involving a third-order tensor.) Again, it follows from (1.9) in the preceding section that for deforming ellipsoids the tensor $\mathbf{E}^{(0)}$ in (1.25) should be replaced by the tensor $\mathbf{A}$ of (1.24). Hence, with the approximation of small strain, the resulting equation for $\Delta\boldsymbol{\Omega}$ yields, on solution by 'successive approximations',

$$\Delta\boldsymbol{\Omega} = \mathbf{A}.\mathbf{C}' - \mathbf{C}'.\mathbf{A} + O(C'^2), \tag{1.27}$$

where as before $O(C'^2)$ denotes terms involving the squares of the components of $\mathbf{C}'$. Thus, the rotation rate of the particle differs from the vorticity of the undisturbed flow by terms of order *one* in the particle strain.

It should also be pointed out here that, as is the case for a rigid ellipsoid, the resultant force as well as the torque on a deforming ellipsoid can readily be shown to vanish, which means in the present context that the ellipsoid moves with the mean velocity of the undisturbed flow, as presupposed implicitly in (1.4).

In summary, now, we note that (1.15), (1.23) and (1.27), together with the appropriate rheological equation for the particle relating $\mathbf{C}'(t)$ to $\mathbf{P}'(t)$ or, more generally, to the stress history ($\mathbf{P}'(t_1)$, $-\infty \leqslant t_1 \leqslant t$) would represent four equations involving four unknown tensor quantities $\mathbf{P}'$, $\mathbf{C}'$, $\mathbf{E}'$ and $\boldsymbol{\Omega}'$. After a discussion of the macroscopic rheology of particle suspensions, we shall consider below a specific and simple rheological model for the particulate phase, for which it appears that the above system is determinate.

2. Dilute suspensions of viscoelastic particles

Here we wish to present the method to be used in the present work for determining the macroscopic rheological behaviour of dilute suspensions, starting from a knowledge of individual particle behaviour at infinite dilution. Hashin (1964) and Happel & Brenner (1965) have recently given reviews, with extensive bibliographies, of past theoretical work on the rheology of suspensions. However, despite the wealth of papers dealing with dilute systems, it would appear that none of the techniques previously employed can be readily applied to the present problem. Thus, it is appropriate to discuss next the assumptions involved in the technique to be used here, before actually applying it.

2.1. *Dilute suspensions of arbitrary particles*

The statistical foundations for a rheological theory of heterogeneous systems have been rather thoroughly discussed elsewhere (e.g. by Frisch 1964), but for the present purposes we shall rely on some of the simplifying assumptions discussed by Hashin (1964). In particular, we consider now a representative volume of our suspension containing a large number of particles; it will be convenient then to refer to the dimensions of the volume under consideration and to those of the particles as the *macroscopic* and the *microscopic* scales, respectively. We suppose now that the sample of suspension under consideration occupies a region $\mathscr{R}(t)$ of constant volume V bounded by a closed surface $\mathscr{S}(t)$. Furthermore, we denote by $\mathscr{R}'(t)$ the subregion of $\mathscr{R}(t)$ containing the particles and by $\mathscr{S}'(t)$ its boundary, consisting of the fluid–particle interfaces.

We imagine now that by application of the appropriate stresses on $\mathscr{S}(t)$ the sample is subjected to a macroscopically homogeneous deformation, such that the velocity at the surface $\mathscr{S}(t)$ is given by

$$\mathbf{v}(\mathbf{r},t) = \mathbf{v}^{(0)}(\mathbf{r},t) = \mathbf{\Gamma}^{(0)}.\mathbf{r} \tag{2.1}$$

up to an additive velocity term independent of position, with $\mathbf{\Gamma}^{(0)} = \mathbf{\Gamma}^{(0)}(t)$ being a velocity-gradient tensor, and $\mathbf{r}$ the position vector relative to an observer moving with (the mass centre, say, of) the sample.

Then, by taking the integral over $\mathscr{R}(t)$ of the resultant velocity-gradient tensor $\mathbf{\Gamma}(\mathbf{r},t)$, which varies of course from point to point on the microscopic scale, we have that

$$\iiint_{\mathscr{R}(t)} \mathbf{\Gamma}(\mathbf{r},t)\,dV = \iiint_{\mathscr{R}(t)} \nabla\mathbf{v}(\mathbf{r},t)\,dV$$

$$= \iint_{\mathscr{S}(t)} \mathbf{v}(\mathbf{r},t)\,\mathbf{n}\,dS = V\mathbf{\Gamma}^{(0)}(t), \tag{2.2}$$

where $\mathbf{n}$ is the unit outer normal to $\mathscr{S}(t)$. The first equality here follows from the definition of $\mathbf{\Gamma}$ and the second from an elementary result of vector calculus, provided that $\mathbf{v}$ is continuous across the particle boundaries $\mathscr{S}'(t)$; finally, the third equality is a consequence of (2.1), together with the (dyadic) relation

$$\iint_{\mathscr{S}(t)} \mathbf{r}\mathbf{n}\,dS = V\mathbf{I},$$

(or its transpose), where V is the volume of $\mathscr{R}(t)$. Denoting volume averages over the entire sample by brackets $\langle\ \rangle$, we shall have then, by (2.2), that

$$\langle\mathbf{\Gamma}\rangle \stackrel{\text{def}}{=} \frac{1}{V}\iiint_{\mathscr{R}(t)} \mathbf{\Gamma}(\mathbf{r},t)\,dV \equiv \mathbf{\Gamma}^{(0)}(t)$$

and that

$$\langle\mathbf{E}\rangle \equiv \mathbf{E}^{(0)} \stackrel{\text{def}}{=} \mathbf{\Gamma}^{(0)} + (\mathbf{\Gamma}^{(0)})^{\dagger}. \tag{2.3}$$

That is to say, the volume averages of the velocity gradient and deformation rate are the same as the 'macroscopic' values imposed by (2.1). This relation has already been cited by Hashin (1964). As indicated by his discussion, the question arises now as to the appropriate definition of the macroscopic stress in terms of $\mathbf{T}(r,t)$, the microscopic stress field in the sample.

One possible definition of stress is obtained as follows: we note that if there are no interfacial effects at the particle boundaries $\mathscr{S}'(t)$, such that $\mathbf{T}.\mathbf{n}$ is continuous across $\mathscr{S}'(t)$, then the 'divergence' theorem gives

$$\iint_{\mathscr{S}(t)} \mathbf{r}\mathbf{T}.\mathbf{n}\,dS = \iiint_{\mathscr{R}(t)} \nabla.(\mathbf{r}\mathbf{T})\,dV$$

(where $\nabla.(\mathbf{r}\mathbf{T})$ has components $\partial(x_i T_{jk})/\partial x_k$ on an orthogonal Cartesian frame). With the further assumption of negligible inertia, as embodied in (1.1), we have that

$$\nabla.(\mathbf{r}\mathbf{T}) = \mathbf{T},$$

and, therefore, the preceding relation can be expressed as

$$\langle \mathbf{T} \rangle = \frac{1}{V} \iint_{\mathscr{S}(t)} \mathbf{rT}.\mathbf{n} dS, \tag{2.4}$$

where $\langle \mathbf{T} \rangle$ denotes the volume-average stress tensor. Hence, we can show readily by (2.1), (2.3) and (2.4) that

$$\langle \mathbf{\Gamma} \rangle . \langle \mathbf{T} \rangle = \langle \mathbf{\Gamma} . \mathbf{T} \rangle$$

and, as a consequence, that

$$\langle \mathbf{E} \rangle : \langle \mathbf{T} \rangle = \langle \mathbf{E} : \mathbf{T} \rangle. \tag{2.5}$$

The latter relation implies that the stress work derived from the individual volume averages of the stress and deformation-rate tensors is the same as the volume average of the stress work. Otherwise stated, the volume-average stress represents an 'energy preserving' definition of stress, under the imposed macroscopic deformation of (2.1). In this respect, (2.5) is essentially equivalent to a result cited by Hashin (1964). This same criterion has served as the basis for definitions of macroscopic rheological parameters in many past theoretical studies, and we shall likewise adopt it here.

Under the preceding assumption, we are now in a position to formulate the rheological equation for the suspension. In particular, with the above definition of the macroscopic stress tensor, it follows that the macroscopic value of the deviatoric stress is also given by the volume average $\langle \mathbf{P} \rangle$ of the deviatoric stress $\mathbf{P}(\mathbf{r}, t)$ in the suspension.

Therefore, recalling that, in the subregion of $\mathscr{R}(t)$ occupied by the (Newtonian) fluid, the deviatoric stress is given by

$$\mathbf{P} = 2\mu \mathbf{E},$$

one sees that

$$\langle \mathbf{P} \rangle - 2\mu \langle \mathbf{E} \rangle \equiv \langle \mathbf{P} - 2\mu \mathbf{E} \rangle = \phi \langle \mathbf{P}' - 2\mu \mathbf{E}' \rangle$$
$$\equiv \phi [\langle \mathbf{P}' \rangle - 2\mu \langle \mathbf{E}' \rangle], \tag{2.6}$$

where the primes denote now an average over the region occupied by particulate phase $\mathscr{R}'(t)$, and where ϕ denotes the ratio of the volume of $\mathscr{R}'(t)$ to that of $\mathscr{R}(t)$; i.e. ϕ is the volume fraction of the particulate phase.‡ Now, (2.6) will provide a rheological relation for a suspension, once the stress and deformation-rate tensors, $\mathbf{P}'(\mathbf{r}, t)$ and $\mathbf{E}'(\mathbf{r}, t)$ for a given particle, are specified in terms of the imposed deformation rate $\langle \mathbf{E} \rangle = \mathbf{E}^{(0)}$ of (2.3).

Up to this point, we have made no assumption as to the concentration of the particulate phase. But now, in order to apply (2.6) in the present work, we shall assume that the volume fraction ϕ of the particles is sufficiently small that interactions of the individual particles are negligible. Thus following Happel & Brenner (1965), we let the sample volume V of $\mathscr{R}(t)$ tend to infinity and we assume that the boundary condition of (2.1) can be replaced by that of (1.4). In other words, considering an arbitrarily chosen particle, the velocity distribution

‡ Equation (26), which is a generalization for deformable particles of a formula cited by Giesekus (1962), has also been proposed by Landau & Lifschitz (1959, pp. 76–9), who, however, offer no detailed justification for its use.

in its vicinity is assumed to be governed by (1.2) to (1.4). Although the general validity of this assumption has been questioned by Happel & Brenner (1965), it can be justified within the framework of the present method. Thus it is reasonable to suppose for $\phi \to 0$ that the difference between the actual values of $\mathbf{E}'$ and $\mathbf{P}'$ for a particle in the suspension and the values for a single particle, in a large volume of fluid subjected to the deformation (2.1), should be at most $O(\phi)$. This would imply then that the use of the latter values of $\langle \mathbf{E}' \rangle$ and $\langle \mathbf{P}' \rangle$ in (2.6) would involve an error there $O(\phi^2)$ relative to the leading terms $\langle \mathbf{P} \rangle$ and $\langle \mathbf{E} \rangle$. As we are concerned here only with terms $O(\phi)$ we shall make use of the above approximation.

Finally, since we have restricted this analysis to particles which are intrinsically spherical and have further assumed the absence of torques on the particle arising from external fields or Brownian effects, it is reasonable to expect that, subsequent to any initial state of rest or of isotropic stress in the sample of suspension, the motion and orientation of the individual particles will be identical, provided of course that the particles all have the same rheological behaviour. Assuming this to be the case and making use of (2.3), we can then replace (2.6) by

$$\langle \mathbf{P} \rangle = 2\mu \mathbf{E}^{(0)} + \phi(\mathbf{P}' - 2\mu \mathbf{E}'), \tag{2.7}$$

where $\mathbf{P}'(t)$ and $\mathbf{E}'(t)$, the same now for *all* particles, are to be determined from the relation given in the preceding section, together with a rheological equation for the particles.

2.2. *Suspensions of viscoelastic spheres*

We consider now a suspension of viscoelastic spheres of an incompressible, isotropic solid. Since we have already restricted our discussion to small strains, we shall assume here that the rheological behaviour of the particles is adequately described by a constitutive equation of the form

$$\mathbf{C}'(t) = \mathscr{F}'[\mathbf{P}'(t)], \tag{2.8}$$

$\mathscr{F}'$ denoting a 'quasi-linear' viscoelastic operator

$$\mathscr{F}' = \frac{1}{2k}\left(\frac{1+\alpha_1(\mathscr{D}'/\mathscr{D}t)+\alpha_2(\mathscr{D}'/\mathscr{D}t)^2+\ldots}{1+\beta_1(\mathscr{D}'/\mathscr{D}t)+\beta_2(\mathscr{D}'/\mathscr{D}t)^2+\ldots}\right), \tag{2.9}$$

where k, α_1, α_2, ... and β_1, β_2, ... are constants and where, for any second-order tensors $\mathbf{B}(t)$ associated with a particle, the operation $\mathscr{D}'/\mathscr{D}t$ is defined by

$$(\mathscr{D}'/\mathscr{D}t)\,\mathbf{B}(t) = (d/dt)\,\mathbf{B}(t) + \mathbf{B}\,.\,\mathbf{\Omega}' - \mathbf{\Omega}'\,.\,\mathbf{B}, \tag{2.10}$$

$\mathbf{\Omega}'$ being the vorticity tensor for the particle. This operation, which is essentially the Jaumann derivative (Prager 1961), ensures that (2.9) has the proper material frame-indifference, with account being taken of material rotation.

To facilitate the following discussion we consider first and foremost the simplest form of (2.9), the purely elastic particle, in which case (cf. Fröhlich & Sack 1946)

$$\alpha_1 = \alpha_2 = \ldots = \beta_1 = \beta_2 = \ldots = 0 \quad \text{and} \quad \mathbf{P}' = \mathbf{C}'/2k, \tag{2.11}$$

the constant k (with dimensions of stress) now designating the Hookean elastic modulus of the material.

For the purposes of the analysis to follow, we define the 'magnitude' of a tensor $\mathbf{B}$ by the scalar invariant

$$|\mathbf{B}| = |\mathbf{B}:\mathbf{B}|^{\frac{1}{2}} \equiv (\operatorname{tr}\mathbf{B}^2)^{\frac{1}{2}}, \tag{2.12}$$

and, in connexion with order-of-magnitude estimates, we shall employ both the symbols $O(\mathbf{B})$ and $O(\epsilon)$ to denote tensors with individual components or magnitudes which are $O(|\mathbf{B}|)$ or $O(\epsilon)$. Also, we shall henceforth include in the O-notation certain quantities having physical dimensions, simply to indicate the pertinent dimensions involved in various relations.

Now, in the case of the elastic particle defined by (2.11), (1.23) yields the following expression for the particle strain

$$\tau^{-1}\mathbf{C}' = \tfrac{5}{3}\{\mathbf{A}+\tfrac{3}{7}(\mathbf{A}.\mathbf{C}'+\mathbf{C}'.\mathbf{A})-\tfrac{2}{7}(\mathbf{A}:\mathbf{C}')\,\mathbf{I}\}+\tfrac{2}{3}\mathbf{E}'+O(\mathbf{A}.\mathbf{C}'^2), \tag{2.13}$$

where $\mathbf{A} = \mathbf{E}^{(0)}-\mathbf{E}'$, as in (1.24), and

$$\tau \overset{\text{def}}{=} 3\mu/2k \tag{2.14}$$

is a characteristic time parameter for the suspension. The condition of small particle strain postulated in § 1.3 can now be formulated precisely, by requiring that the fluid stress on the particle be much smaller than the elastic modulus. Thus we have, by (2.13), that

$$\mathbf{C}' = O(\tau\mathbf{A})$$

for $\tau \to 0$, and, as we shall later confirm, the preceding relation implies that

$$\mathbf{A} = O(\gamma),$$

and, hence, that

$$\mathbf{C}' = O(\epsilon), \quad \text{for } \epsilon \overset{\text{def}}{=} \gamma\tau \to 0, \tag{2.15}$$

where

$$\gamma = |\mathbf{E}^{(0)}|$$

is the macroscopic 'shear' rate. The following analysis is based then on the postulate that the dimensionless group $\epsilon = \epsilon(t)$ is always small relative to unity.

By means of (1.15) and (1.27) the particle deformation rate $\mathbf{E}'$ can be related to the strain $\mathbf{C}'$, in a manner consistent with (2.15), by

$$\mathbf{E}' = (\mathscr{D}\mathbf{C}'/\mathscr{D}t)+\mathbf{E}'.\mathbf{C}'+\mathbf{C}'.\mathbf{E}'+O(\gamma\epsilon^2) \tag{2.16}$$

where, for a second-order tensor $\mathbf{B}(t)$,

$$\mathscr{D}\mathbf{B}/\mathscr{D}t = (d\mathbf{B}/dt)+\mathbf{B}.\mathbf{\Omega}-\mathbf{\Omega}.\mathbf{B} \tag{2.17}$$

with $\mathbf{\Omega} \equiv \mathbf{\Omega}^{(0)}$ being the macroscopic vorticity tensor.

We have now, in the form of (2.13) and (2.16), a number of relations sufficient to determine the particle strain $\mathbf{C}'$ and deformation rate $\mathbf{E}'$, to terms $O(\epsilon)$, once the appropriate initial conditions, as well as the form of the macroscopic velocity gradient $\mathbf{\Gamma}^{(0)}(t)$, are specified. With the further relations of (1.23) and (2.7), this will permit the specification of the macroscopic rheological equation for the suspension, up to terms which are either $O(\epsilon^2)$ or $O(\phi^2)$.

In the following paragraphs, we shall omit the brackets $\langle\ \rangle$ and the superscripts zero which were introduced in § 1 to distinguish macroscopic variables. Also, we note at this point that the operation defined by (2.17) is ultimately to be interpreted as the macroscopic *Jaumann* derivative (Prager 1961), and, hence, the ordinary time derivative in (2.16) must eventually be replaced by the well-known material or substantial derivative.

Equations (2.13) and (2.16) are now to be solved simultaneously for $\mathbf{E}'$ and $\mathbf{C}'$. However, because of (2.15), we can immediately express $\mathbf{E}'$ in terms of $\mathbf{C}'$ by successive substitutions on the right-hand side of (2.16). Taking account of the incompressibility condition in (1.10), we find thus that

$$\mathbf{E}' = \left\{\frac{\mathscr{D}\mathbf{C}'}{\mathscr{D}t} + 2\mathscr{S}\!d\left[\mathbf{C}'.\frac{\mathscr{D}\mathbf{C}'}{\mathscr{D}t}\right]\right\}\{1+O(\epsilon^2)\}+O(\gamma\epsilon^2), \tag{2.18}$$

where, for the sake of brevity, we have introduced the notation $\mathscr{S}\!d[\mathbf{B}]$ to denote the *symmetric part of the deviator* of a tensor $\mathbf{B}$; i.e.

$$\mathscr{S}\!d[\mathbf{B}] \overset{\text{def}}{=} \tfrac{1}{2}\{\mathbf{B}+\mathbf{B}^{\dagger}-\tfrac{2}{3}(\operatorname{tr}\mathbf{B})\,\mathbf{I}\}. \tag{2.19}$$

In applying (2.19), it should be recalled that all the stress and deformation tensors heretofore introduced, as well as their derivatives $\mathscr{D}/\mathscr{D}t$, are symmetric.

When (2.18) is substituted into (2.13) to eliminate $\mathbf{E}'$, we obtain the following differential equation for $\mathbf{C}'$,

$$\frac{\mathscr{D}\mathbf{C}'}{\mathscr{D}t}+\frac{1}{\tau}\mathbf{C}' = \tfrac{5}{3}\mathbf{E}+\mathscr{S}\!d\left[\frac{10}{7}\left(\mathbf{E}-\frac{12}{5}\frac{\mathscr{D}\mathbf{C}'}{\mathscr{D}t}\right).\mathbf{C}'\right]+O(\gamma\epsilon^2), \tag{2.20}$$

in which we recall that $\mathbf{E}$ denotes the macroscopic deformation rate. In a similar way one finds, by (1.23) and (2.18), that the quantity of ultimate interest, which appears on the right-hand side of (2.7), is given by

$$\mathbf{P}'-2\mu\mathbf{E}' = 5\mu\left\{\left(\mathbf{E}-\frac{\mathscr{D}\mathbf{C}'}{\mathscr{D}t}\right)+\mathscr{S}\!d\left[\frac{6}{7}\left(\mathbf{E}-\frac{10}{3}\frac{\mathscr{D}\mathbf{C}'}{\mathscr{D}t}\right).\mathbf{C}'\right]+O(\gamma\epsilon^2)\right\}. \tag{2.21}$$

Hence, the problem consists essentially of solving (2.20) in such a way as to yield expressions for $\mathbf{C}'$ and $\mathscr{D}\mathbf{C}'/\mathscr{D}t$ with an error $O(\epsilon^2)$, which will be suitable for substitution into (2.21); this can be accomplished formally as follows.

First of all, we note that in (2.20) the terms occurring inside the argument of $\mathscr{S}\!d[\ ]$ are $O(\epsilon)$ relative to the remaining, explicit terms. Therefore, we can once again employ a method of successive approximations based on small ϵ, to treat (2.21). Thus, neglecting the second term on the right-hand side of (2.20), we have, as a first approximation, that

$$\mathbf{C}' = \tfrac{5}{3}\mathscr{C}\{\tau\mathbf{E}\}+O(\epsilon^2), \tag{2.22}$$

and, on substitution of this expression into the right-hand side of (2.20), as a second approximation, that

$$\mathbf{C}' = \tfrac{5}{3}\mathscr{C}\{\tau\mathbf{E}+\tfrac{10}{7}\tau\mathscr{S}\!d[\mathscr{B}\{\mathbf{E}\}.\mathscr{C}\{\tau\mathbf{E}\}]+O(\epsilon^2)\}, \tag{2.23}$$

where $\mathscr{C}$ and $\mathscr{B}$ denote the linear operations

$$\mathscr{C}\{\mathbf{B}\} = [1+\tau(\mathscr{D}/\mathscr{D}t)]^{-1}\mathbf{B} \quad \text{and} \quad \mathscr{B}\{\mathbf{B}\} = [1-3\tau(\mathscr{D}/\mathscr{D}t)]\,\mathscr{C}\{\mathbf{B}\}. \tag{2.24}$$

By means of a calculus for the Jaumann derivative, discussed recently by Goddard & Miller (1966), it is possible to express the operations of (2.24) in closed form, i.e. to integrate the differential equations implied there, subject of course to the appropriate initial conditions. In the present work we shall leave these and subsequent, related operations in the 'implicit' form (2.24), noting simply that they can also be rendered explicit by means of the expansion

$$\mathscr{C}\{\mathbf{B}\} = \left(1-\tau\frac{\mathscr{D}}{\mathscr{D}t}+\tau^2\frac{\mathscr{D}^2}{\mathscr{D}t^2}-\tau^3\frac{\mathscr{D}^3}{\mathscr{D}t^3}+\ldots\right)\mathbf{B}, \tag{2.25}$$

provided that

$$\left|\tau^n\frac{\mathscr{D}^n}{\mathscr{D}t^n}\mathbf{B}\right| \leqslant \lambda^n|\mathbf{B}|, \tag{2.26}$$

where $0 < \lambda < 1$ is a constant. Since the macroscopic vorticity tensor $\mathbf{\Omega}$ can be made to vanish by appropriate choice of the reference frame (which incidentally is the basis for the aforementioned calculus of $\mathscr{D}/\mathscr{D}t$), the inequality in (2.26) is effectively a restriction on the time rates of change of $\mathbf{B}$.

In view of the expression (2.22) for $\mathbf{C}'$ we note that, even when $\mathbf{E}$ is a rapidly or discontinuously varying function of time (as in the case of a suddenly imposed macroscopic deformation), our original assertion that the particle strain $\mathbf{C}'$ is $O(\epsilon)$ should remain valid, subject only to the previous restriction on the magnitude of $\mathbf{E}$, $\epsilon \ll 1$; this can be inferred simply from (2.22) by consideration of an irrotational flow, $\mathbf{\Omega} \equiv 0$, in which case the operations in (2.24) can easily be rendered explicit. In like manner one concludes that $\mathscr{D}\mathbf{C}'/\mathscr{D}t$ and, hence, $\mathbf{E}'$ are either $O(\gamma\epsilon)$, whenever (2.26) holds, or at most $O(\gamma)$, for rapidly varying $\mathbf{E}$, which in either case confirms the estimate (2.15).

To obtain an expression for the derivative $\mathscr{D}\mathbf{C}'/\mathscr{D}t$, we now substitute the expression (2.23) for $\mathbf{C}'$ into all the terms of (2.20), except the term $\mathscr{D}\mathbf{C}'/\mathscr{D}t$ on the left-hand side. We obtain thus an expression for that derivative which is accurate to terms $O(\epsilon)$ and which, therefore, is suitable for subsequent substitution into (2.21), together with the expression (2.22) for $\mathbf{C}'$. The resulting equation is, of course, to be employed then in (2.7). Omitting the algebraic details, we consequently obtain the rheological equation for the suspension, up to terms that are presumably $O(\phi^2)$,

$$\mathbf{P}-2\mu\mathbf{E} = 5\mu\phi(\mathscr{A}\{\mathbf{E}\}+\tfrac{50}{21}\mathscr{C}\{\mathscr{S}d[\mathscr{B}\{\mathbf{E}\}.\mathscr{C}\{\tau\mathbf{E}\}]\} \\ -\tfrac{20}{21}\mathscr{S}d[\mathscr{A}\{\mathbf{E}\}.\mathscr{C}\{\tau\mathbf{E}\}]+O(\gamma\epsilon^2)), \tag{2.27}$$

where

$$\mathscr{A}\{\mathbf{E}\} = [1-\tfrac{2}{3}\tau(\mathscr{D}/\mathscr{D}t)]\mathscr{C}\{\mathbf{E}\}, \tag{2.28}$$

and the operations $\mathscr{B}$, $\mathscr{C}$ and $\mathscr{S}d$ are given by (2.24) and (2.19).

As pointed out previously, the various operations in (2.27) could be rendered explicit by means of a calculus for the Jaumann derivative (Goddard & Miller 1966). However, this would be somewhat superfluous whenever $\mathbf{E}$ satisfies (2.26) as, for example, is the case with steady simple-shearing flows. For then, by applying the inverse of the operation $\mathscr{C}$ to both sides of (2.27), making use of the expansion (2.25), and retaining only those terms of $O(\epsilon)$ relative to the leading terms, one can reduce (2.27) to the more explicit form

$$\mathbf{P}+\tau\frac{\mathscr{D}\mathbf{P}}{\mathscr{D}t} = 2\mu\left[(1+\tfrac{5}{2}\phi)\mathbf{E}+(1-\tfrac{5}{3}\phi)\tau\frac{\mathscr{D}\mathbf{E}}{\mathscr{D}t}+\tfrac{25}{7}\phi\tau(\mathbf{E}^2-\tfrac{1}{3}\operatorname{tr}(\mathbf{E}^2)\mathbf{I})+O(\epsilon^2\gamma)\right]. \tag{2.29}$$

Equation (2.29) could of course be cast into other forms, by means of expansions similar to (2.29), but these would all be equivalent to terms $O(\epsilon)$.

We note that if the non-linear terms in $\mathbf{E}$ and $\mathbf{\Omega}$ are omitted from (2.27) and (2.29) the resulting equations differ only by terms that are $O(\phi^2)$ from the relation given previously by Fröhlich & Sack (1946). The non-linear terms derived here result evidently from the effect of stress-induced ellipticity of the particles on the flow field of the suspending fluid.

Now, the preceding analysis for elastic spheres is applicable, with certain restrictions, to suspensions of the viscoelastic spheres defined by (2.9). In particular, because of (1.27) it appears that one can replace the derivative $\mathscr{D}'/\mathscr{D}t$ in (2.9) by the macroscopic Jaumann derivative of (2.17), with an absolute error of $O(\epsilon^2)$, provided that the constants in (2.9) satisfy the relations

$$\alpha_n = O(\tau^n), \quad \beta_n = O(\tau^n), \quad \text{for } \tau \to 0, \tag{2.30}$$

for $n = 1, 2, 3, \ldots$, where as before τ is the characteristic time defined by (2.14). In this case, (2.20) to (2.28) remain equally valid for a suspension of viscoelastic spheres, if the time constant τ is everywhere replaced by the *operator*

$$\mathscr{T} = \tau\left(\frac{1+\alpha_1(\mathscr{D}/\mathscr{D}t)+\alpha_2(\mathscr{D}^2/\mathscr{D}t^2)+\ldots}{1+\beta_1(\mathscr{D}/\mathscr{D}t)+\beta_2(\mathscr{D}^2/\mathscr{D}t^2)+\ldots}\right), \tag{2.31}$$

derived from (2.9). Although it appears that weaker conditions than (2.30) might be justified in certain cases, we shall not pursue this question further here. We note, however, that if non-linear terms in $\mathbf{E}$ and $\mathbf{\Omega}$ are again omitted from (2.27) one obtains, to terms $O(\phi)$, Oldroyd's (1953) generalization of the Fröhlich–Sack equation. In passing, it should also be noted that in many instances operators such as (2.9) and (2.31) could more appropriately be expressed in closed form by means of a calculus for the Jaumann derivative (Goddard & Miller 1966).

As a limitation on (2.27), we should discuss briefly here the assumption of negligible inertia embodied in (1.1). Since the suspending fluid is presumed to be Newtonian, the approximation of negligible inertia can be based on the usual 'Reynolds-number' criterion, which takes the form here:

$$\gamma a_0^2 \rho/\mu \ll 1, \tag{2.32}$$

where, as above, a_0 is the undeformed particle radius, γ the magnitude of the deformation rate for the fluid, μ its viscosity, and ρ its density (cf. Happel & Brenner 1965). On the other hand for the solid particles considered here, having approximately the same density as that of the fluid, inertial effects in the particle should be negligible provided that

$$\rho\gamma^2 a_0^2/k \equiv \tfrac{2}{3}\epsilon(\gamma a_0^2 \rho/\mu) \ll 1, \tag{2.33}$$

where k is the elastic modulus of the particle. One sees, however, that (2.33) follows *a fortiori* from (2.32) and the condition $\epsilon \ll 1$ postulated above.

Since (2.32) involves parameters which do not enter into the preceding analysis, the condition stated there can in principle be realized independently of our previous assumptions. For example, (2.32) would always hold for very small particles, although it should be noted that in practice there is a lower limit on particle size for which the orientation effects of Brownian motion can be neglected. At any rate, one can conceive of suspensions where the non-linear terms appearing in (2.27) are important, even when inertial effects play a negligible role in the particle deformation at the microscopic level.

In closing here, it is interesting to note that (2.29) is a special case of the well-known phenomenological equation proposed by Oldroyd (1958); by means of the analysis presented in his paper one deduces that, in a steady simple-shearing flow, a fluid described by (2.29) would exhibit 'shear thinning' and unequal normal stresses in all three directions, each directly proportional to τ.

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