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The stress system in a suspension of force-free particles

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The purpose of the paper is to consider in general terms the properties of the bulk stress in a suspension of non-spherical particles, on which a couple (but no force) may be imposed by external means, immersed in a Newtonian fluid. The stress is sought in terms of the instantaneous particle orientations, and the problem of determining these orientations from the history of the motion is not considered. The bulk stress and bulk velocity gradient in the suspension are defined as averages over an ensemble of realizations, these averages being equal to integrals over a suitably chosen volume of ambient fluid and particles together when the suspension is statistically homogeneous. Without restriction on the type of particle or the concentration or the Reynolds number of the motion, the contribution to the bulk stress due to the presence of the particles is expressed in terms of integrals involving the stress and velocity over the surfaces of particles together with volume integrals not involving the stress. The antisymmetric part of this bulk stress is equal to half the total couple imposed on the particles per unit volume of the suspension. When the Reynolds number of the relative motion near one particle is small, a suspension of couple-free particles of constant shape is quasi-Newtonian; i.e. the dependence of the bulk stress on bulk velocity gradient is linear. Two significant features of a suspension of non-spherical particles are (1) that this linear relation is not of the Newtonian form and (2) that the effect of exerting a couple on the particles is not confined to the generation of an antisymmetrical part of the bulk stress tensor. The role of surface tension at the particle boundaries is described.

In the case of a dilute suspension the contributions to the bulk stress from the various particles are independent, and the contributions arising from the bulk rate of strain and from the imposed couple are independent for each particle. Each particle acts effectively as a force doublet (i.e. equal and opposite adjoining 'Stokeslets') whose tensor strength determines the disturbance flow far from the particle and whose symmetrical and antisymmetrical parts are designated as a stresslet and a couplet. The couplet strength is determined wholly by the externally imposed couple on the particle; but the stresslet strength depends both on the bulk rate of strain and, for a non-spherical particle, on the rate of rotation of the particle relative to the fluid resulting from the imposed couple. The general properties of the stress system in a dilute suspension are illustrated by the specific and complete results which may be obtained for rigid ellipsoidal particles by use of the work by Jeffery (1922).

1. Introduction

Suspensions of small particles in fluid are common in nature and in several engineering fields, and it is desirable to know how they respond to imposed forces or motions at their boundaries. Provided the length scale of the motion imposed on the suspension is large compared with the average spacing between the particles, the suspension may evidently be regarded for some purposes as a homogeneous fluid. The problem is to determine the rheological properties of this equivalent homogeneous fluid from a knowledge of the properties of the particles and the ambient fluid in which they are suspended; in other words, to determine the relation between the macroscopic or bulk properties of the suspension and its microscopic structure on the particle scale.

The most important rheological property is usually the stress corresponding to a given bulk motion, and this alone will be investigated here.

The particles and the ambient fluid will be supposed to be incompressible, of uniform temperature, and permanent in constitution; no thermodynamical considerations will arise. It will also be assumed that the ambient fluid is Newtonian in its stress behaviour and that the particles are distributed with statistically uniform concentration throughout the fluid.

When the particles are spherical, and no external force or couple acts on them, the suspension has a wholly isotropic structure, and so behaves as a Newtonian fluid for sufficiently small rates of strain. In these circumstances the effect of the presence of the particles is simply equivalent to an increase in the shear viscosity of the suspension. The magnitude of this increase, expressed as a fraction of the viscosity μ of the ambient fluid, is a linear function of the concentration of the particles by volume (c, say) when $c \ll 1$, and the constant of proportionality depends on the constitution of the particles, being $2{\cdot}5$ for rigid spheres and $(\mu+2{\cdot}5\mu')/(\mu+\mu')$ for fluid spheres of viscosity μ'. In recent years research on suspension rheology has been directed towards extensions of these well-known results, in particular for the cases of (*a*) more concentrated suspensions of spherical particles, (*b*) deformable particles with both viscous and elastic properties, and (*c*) non-spherical rigid particles. None of these extensions is yet complete, and there are also other problems which have not yet been given much consideration, such as the effect of inertia forces in the relative motion near a particle, and the effect of an externally imposed couple on the particles. Theoretical analysis of the stress properties of a suspension of particles is still in its early stages.

Much of this paper is concerned with the formulation of the problem of determining the stress in a suspension of particles, in a way which provides a framework for known results and for further developments. This is not as straightforward as might be supposed, partly because the meaning of bulk stress in a suspension is not obvious; the view taken here is that it should be defined as a probability ensemble average. In view of the notorious difficulty in reconciling contributions to suspension mechanics written from different points of view, I have thought it worth while to set out the argument in fairly complete and general form. It is assumed that the body force per unit volume exerted by external means is uniform over the ambient fluid and particles alike, with the implication that there is no systematic translational motion of a particle through the ambient fluid. However, allowance will be made for the existence of a relative rotational motion of a particle and the ambient fluid due to the action of an externally imposed couple on the particle (as might be caused by an external magnetic or electric field for particles of suitable material—a situation with interesting possibilities for control of the mechanical properties of a suspension). The properties of the stress tensor will be considered for particles of general shape and constitution, and in the last section these properties are illustrated and interpreted with the aid of specific formulae for the case of rigid particles of ellipsoidal shape in a dilute suspension. In subsequent related papers, results which have been obtained by the author and colleagues at Cambridge for various particular suspensions will be described.

Aside from the obvious direct purpose of this paper and those to follow, namely, to derive results concerning the stress which are applicable, at least approximately, to certain naturally occurring suspensions, there is an underlying indirect objective. A major difficulty in the study of rheology is that one's intuition about the form of the constitutive stress relation appropriate to given circumstances is so poorly developed. It is often hard to know even in broad terms how a given material will behave, chiefly because we have at our disposal so few definite and well-understood constitutive relations for non-Newtonian fluids to provide guidance. This difficulty affects mathematical theory as well as the interpretation of observation, since, for lack of concrete results which can be used as a testing ground, the hypotheses on which analysis must perforce be based tend to be artificial and unmotivated. Now the microscopic structure of a suspension can be precisely specified, and it may be possible—and not only in principle—to *deduce* some of the macroscopic properties of the suspension and to see in explicit terms their relation to the microstructure. This seems to me to give the mechanics of suspensions an especially important place in current studies of rheology, in that we have the unusual opportunity of obtaining definite and explicable constitutive relations which are known to apply to specifiable materials and which may be used as a reliable guide for intuition. The development of understanding made possible by these concrete constitutive relations is the indirect objective referred to above.

It is useful to notice at the outset that two different and logically separate mechanical effects occur in a moving suspension of particles. In a material element of the suspension large enough to contain many particles, there will be at any instant a certain statistical distribution of particles with respect to shape, orientation, size and relative position, and this information we can regard as included in a specification of the instantaneous 'state' of that element of the suspension. The bulk stress in the element corresponding to a given local bulk motion and a given externally imposed couple on each particle depends only on this instantaneous state, in a way which will be examined and which involves a consideration of the motion of the ambient fluid near each particle. This dependence of the bulk stress on the instantaneous particle configuration is the first of the two effects referred to, and is one which poses mainly rheological questions. The second effect is the rotation and relative displacement of the particles under the influence of the local motion of the ambient fluid. The configuration of the particles, and so also the state of an element of the suspension, is changing in a way which depends on the nature of the instantaneous motion of the element; and in general a calculation of the state of an element at an arbitrary time after some given initial instant requries a knowledge of the history of the motion of the element. This is essentially a problem of microscopic dynamics.

Separate consideration of these two effects has some advantages. One is that the relation between the bulk stress and the bulk velocity gradient, for a given state of the suspension, is linear under certain conditions, whereas if the rate of change of the state of the suspension due to particle rotation is considered simultaneously this fundamental linearity is concealed. It also happens that there are some circumstances in which the orientation of each particle is constant, in which case the derived relation between the bulk stress and the instantaneous distribution of particle orientations is of immediate value; examples are: (1) a steady pure straining motion of a suspension of non-spherical couple-free particles (such as an ellipsoid, which takes up an asymptotic orientation in which the longest diameter is parallel to the greatest principal rate of strain and the shortest is parallel to the least principal rate of strain—see (6.7), near the end of this paper); and (2) a suspension of ferromagnetic particles on which is exerted a couple, by means of an external magnetic field, of sufficiently large magnitude to hold the particle orientation fixed despite the tendency for the particle to rotate with the surrounding fluid.

Only the dependence of the bulk stress on the instantaneous configuration of the particles is considered in this paper; 'history' effects are set aside for subsequent investigation in the context of particular flow systems.

2. The general form of the constitutive relation for a couple-free fluid of non-isotropic structure and small rates of strain

As a preliminary, we recall here the general form of the relation between the stress tensor and the local properties of the motion for any fluid material which does not have isotropic structure and on which no external body couple is imposed.

This general expression for the stress, for a given state of the fluid and for sufficiently small rates of strain, follows from the same phenomenological thermodynamic-type argument that is employed for an ordinary Newtonian viscous fluid. When the fluid as a whole is set into relative motion, an element of the fluid is disturbed from its state of mechanical equilibrium, and the nature and magnitude of the departure from equilibrium is measured by the local velocity gradient of the motion. Random exchanges of momentum between different parts of the element, acting on a molecular scale and perhaps also on a larger scale, inevitably tend to restore the element to equilibrium. The deviatoric part of the stress generated by the motion is a measure of the tendency to restoration of mechanical equilibrium, and, for sufficiently small magnitudes of the velocity gradient, can be expected to be a linear function of that gradient. This is the familiar hypothesis about the restoring flux accompanying small departures from equilibrium which underlies all molecular transport theory. It is important to note that this hypothesis is no less applicable when particles of macro-molecular dimensions are suspended in the fluid, provided the scale on which the motion occurs is sufficiently large, although before using it in this context we shall need to say what is meant by 'the stress' in a fluid which is inhomogeneous on a macro-molecular scale.

Thus the contribution to the stress tensor due to the motion of the fluid is of the general form

$$-P\delta_{ij}+A_{ijkl}\frac{\partial U_l}{\partial x_k}. \tag{2.1}$$

Here P is the scalar parameter in the isotropic part of the stress tensor and is interpreted as the pressure, U_i denotes the (i-component of the) local velocity, and A_{ijkl} is a tensor parameter determined by the local state of the fluid. By definition of the second term in (2.1) as the deviatoric part of the stress tensor, we have

$$A_{iikl} = 0. \tag{2.2}$$

Also the usual consideration of the balance of angular momentum for an element of volume of the fluid shows that, when no body couple acts on the medium, the stress tensor, and so also the tensor A_{ijkl}, must be symmetrical in the indices i and j.

The expression (2.1) for the stress tensor is expected to apply only for 'small' magnitudes of $\partial U_l/\partial x_k$. In the case of a suspension, departures from the linear relation will occur when the magnitude of the velocity gradient is so large that either the ambient fluid ceases to be Newtonian or the disturbance motion in the ambient fluid due to the presence of the particles ceases to be dominated by viscous forces; the latter restriction will normally be the more important, and will be given in more precise form later.

In the case of a fluid of isotropic structure, the expression (2.1) can be simplified considerably. The tensor parameter A_{ijkl} is here of isotropic form, and so must be representable as the sum of three terms which are products of two delta tensors (Jeffreys & Jeffreys 1966, ch. 3). Bearing (2.2) in mind, and also the need for symmetry of A_{ijkl} in i and j for a couple-free medium, we then have

$$A_{ijkl} = \mu(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}-\tfrac{2}{3}\delta_{ij}\delta_{kl}),$$

where μ is a scalar quantity determined by the local state of the fluid; and the expression for the stress due to the motion takes the usual Newtonian form (for an incompressible fluid)

$$-P\delta_{ij}+\mu\left(\frac{\partial U_j}{\partial x_i}+\frac{\partial U_i}{\partial x_j}\right).$$

These latter simplifications are not applicable in the case of a fluid of non-isotropic structure. However, the essential linearity of the expression (2.1) for the stress tensor in the velocity gradient *is* applicable, and it seems appropriate to label such a medium as *quasi-Newtonian*.

According to the point of view adopted here, the expression (2.1) for the stress is concerned only with local instantaneous quantities, and the parameter A_{ijkl} representing the state of the fluid is formally independent of the motion. However, in a consideration of the way in which the state of the material element of the fluid, and so the value of A_{ijkl}, changes with time, the relation between the current value and a given initial value of A_{ijkl} would be found in general to involve the local velocity and its spatial derivatives at different times. The point may be made clear by reference to the description of the stress in a Newtonian fluid of non-uniform temperature; so far as the instantaneous state of the fluid is concerned, the deviatoric stress is a linear function of the local rate of strain, but the constant of proportionality—the shear viscosity—depends on the temperature of the material element concerned and this changes in a way which brings in a further dependence on the history of the motion of the element. In the case of a suspension of non-spherical particles, the local state of the fluid changes mainly as a consequence of rotation of the particles. Only when the local and instantaneous state of a fluid is regarded as given is the constitutive relation linear in general.

The expression (2.1) shows explicitly that the number of scalar parameters required in general to describe the frictional properties of a suspension, regarded as a fluid of non-isotropic structure, is equal to the number of independent components of A_{ijkl}, and this, in view of (2.2) and the symmetry in i and j, and if we anticipate symmetry in k and l, corresponding to no effect of a bulk rigid rotation, is 30. Hand (1962) has investigated the frictional properties of a non-isotropic fluid on the assumption that the 'substructure' is determined by a symmetric second-order tensor; this we see is not sufficient in general, although it clearly is adequate in the particular case of a dilute suspension of similarly oriented and similarly shaped rigid ellipsoids, which was the only particular case considered by Hand.

3. The averaging process for a suspension

In crude terms, we wish to know what stress is generated in the suspension when a prescribed bulk motion is imposed on it. However, the velocity, pressure and stress all vary with position in the suspension, depending on proximity to a particle, and the terms 'bulk stress in the suspension' and 'bulk velocity gradient' have meaning only in some integral or average sense, and the method of averaging must now be specified.

There appear to be advantages in the use of the terminology and concepts of statistical mechanics and random function theory, as in the analogous fields of turbulence and kinetic theory of gases. The parallel with kinetic theory is obviously close, the primary mechanical difference being that interactions between the particles are a consequence of interparticle forces in one case and of continuum forces in the medium surrounding the particles in the case of a suspension.

A suspension is a system which is determinate (so far as an observer is concerned) only in a statistical sense, inasmuch as the exact location of the particles is different for different realizations of the suspension with the same macroscopic conditions (that is, with the same shape and motion of the boundary of the suspension). A large number of such realizations with the same macroscopic boundary conditions make up an ensemble, and an average over the values of some quantity occurring in these realizations is an ensemble average (and will be denoted by an over-bar). Thus, if $\mathbf{u}$ is the actual vector velocity at position $\mathbf{x}$ in the suspension—perhaps in the ambient fluid, perhaps in a particle—for one realization, the ensemble average $\bar{\mathbf{u}}$ may be defined and observed in principle. We shall adopt this procedure of averaging over an ensemble as the basic definition of the averaging process which yields bulk quantities (as suggested by Hashin (1964)), although we shall see that other averaging procedures which give identical results under certain conditions may be more convenient for calculation.

It is also possible to take an ensemble average of the values of the actual stress σ_{ij} at a point $\mathbf{x}$ (which again may be located either in the ambient fluid or inside a particle). However, here it must be remembered that the stress across a surface element is a resultant of the forces exerted across the element and the momentum flux across the element, and that when velocity fluctuations are concealed by an averaging process the momentum flux due to these fluctuations must be included in the definition of the bulk or macroscopic stress (in ways already familiar in kinetic theory and turbulence). Thus the 'bulk stress in the supension' is taken to be

$$\Sigma_{ij} = \overline{\sigma}_{ij} - \overline{m}_{ij}, \tag{3.1}$$ †

where m_{ij} is the momentum flux tensor. In the absence of any average relative motion of the two constituents of the suspension, m_{ij} has the familiar form $\rho u_i' u_j'$, where $\mathbf{u}' = \mathbf{u} - \bar{\mathbf{u}}$ and ρ is the common density of the ambient fluid and the particles.

Ensemble averages can neither be calculated directly nor observed conveniently, and it is necessary to consider the relation between ensemble averages and both calculable averages and those bulk quantities that are observed in at least the most common type of experiment. To establish this relation we shall take for granted the usual ergodicity property of equality of the ensemble average of some quantity and an integral average of the same quantity over any position co-ordinate with respect to which the quantity is statistically stationary. It will also be necessary to assume that the suspension under discussion is locally statistically homogeneous, in the sense that it is possible to find a length L which is large compared with the average particle spacing and over which the statistical properties of the suspension—defined as ensemble averages—do not vary appreciably. One of these statistical properties is the bulk deviatoric stress, which depends on the average velocity gradient, so that the velocity gradient should be included among the quantities whose average value must be slowly varying. In these circumstances a number of point functions such as u_i', $\partial u_i/\partial x_j$, and σ_{ij} are approximately stationary random functions of the position

† It is prudent to add the qualification that this definition of bulk stress is adopted with a view to use in the bulk momentum equation for the suspension. Whether the same definition is sensible and useful for all other purposes is, I think, still an open question.

vector $\mathbf{x}$ over distances of order L. An integral average of one of these quantities with respect to one or more components of $\mathbf{x}$ over a range of order L is then equal to the ensemble average of that quantity.

This leads us to equality of an ensemble average and a volume average, which is probably the most convenient kind of average for analytical purposes. The restrictions on the volume V over which the average is taken are that V should contain many particles and that the variation of the local statistical properties of the suspension over V should be negligible. Likewise there is equality of the ensemble average of some quantity and an average over a surface which makes an unbiased sample of the suspension and whose linear dimensions are large compared with the average particle spacing. In the particular case of the stress σ_{ij}, an average over a plane surface (which cuts through both ambient fluid and particles) normal to the direction represented by the suffix j has obvious appeal as a way of defining average stress, since it corresponds, although not to the point of realism, to the method of measuring stress in a homogeneous fluid.

It has become common practice in the literature to define macroscopic or bulk quantities as volume averages over the whole or part of the volume occupied by the suspension; this procedure has been adopted for instance by Landau & Lifschitz (1959, §22), Giesekus (1962), Peterson & Fixman (1963), Goddard & Miller (1967) and Roscoe (1967) among writers on suspension rheology, and by Hill (1963) writing on the elastic properties of dispersions of one solid material in another. The alternative starting-point adopted here—the ensemble average—gives identical results in circumstances in which a volume average can usefully be taken (namely, when a volume V satisfying the above restrictions can be found), and has the slight logical advantage, which will not be exploited here, that it is applicable also in circumstances in which the distance between the particles is too large, in relation to the distance over which the suspension properties vary, for a volume average to have significance (just as it is applicable in the comparable kinetic-theory context when the mean free path is not small compared with the boundary dimensions).

Turning now to the relation between ensemble averages and measured quantities, we must recognize that a relation which will apply to all kinds of experiment cannot be given. We shall consider here only the important and representative case in which the suspension is confined between two parallel rigid planes in steady relative shearing motion, with the stress being observed as the force per unit area on a section of one boundary with linear dimensions large compared with particle spacing. Near each of the rigid boundaries there is a layer, presumably with thickness comparable to a particle diameter, in which the statistical properties of the suspension (particle number density in particular) are affected by the presence of the boundary simply because the rigid boundary cannot intersect a particle. However, outside these two layers the suspension may be supposed to be statistically homogeneous, and so in this interior region any space average over a suitably extensive range is equal to the local ensemble average. Moreover, in view of the mechanical equilibrium (on average), the force-plus-momentum-flux per unit area across any large plane surface parallel to the boundaries (and which may cut through particles) is independent of position of the plane surface and in particular is equal to the force per unit area on one of the rigid plane boundaries; somewhat surprisingly, it is an exact inference from equilibrium that the average stress over a plane boundary which does not cut any particle is equal to the average stress-plus-momentum-flux over a parallel plane surface which does. Hence the observed average stress over a suitably extensive area of one of the rigid boundaries is equal to the volume-averaged stress-plus-momentum-flux in the interior homogeneous region and to the ensemble-averaged stress-plus-momentum-flux in that region.

It is also necessary to establish the relation between the theoretical average velocity gradient in the suspension and observable velocities. This can usually be done with the aid of the identity

$$\int \frac{\partial u_i}{\partial x_j}\, dV = \int u_i n_j\, dA. \tag{3.2}$$

In the above case of flow between two rigid parallel planes in steady relative shearing motion, we may choose the volume V to be the intercepted portion of a cylinder with generators in the y-direction normal to the two rigid planes, whence

$$\int \frac{\partial u}{\partial y} dV = \int_{A_2} u\, dA - \int_{A_1} u\, dA,$$

where u is the velocity component in the direction of relative motion of the rigid planes and A_1 and A_2 are the intercepts of the cylinder with the two rigid planes. On A_1 and A_2 we have $u = \alpha y$ say, giving the exact relation

$$\frac{1}{V} \int \frac{\partial u}{\partial y}\, dV = \alpha. \tag{3.3}$$

The volume over which this integral is taken includes the layers adjoining the two rigid boundaries in which conditions are untypical and which must be excluded from the range of integration in any space average if it is to be equal to the ensemble average. However, the two layers are thin, and, provided the distance between the rigid planes is large compared with the particle dimensions, they make a negligible contribution to the integral in (3.3). Thus the velocity gradient averaged over a volume confined to the homogeneous interior region is equal to α, that is, to the value which would be inferred from the relative velocity of the boundaries with the assumption of uniform bulk velocity gradient throughout the suspension.

4. The relation between the bulk stress and the detailed motion in the suspension

We now use the expression for the bulk stress in the suspension as a volume integral in order to establish its relation to the distributions of velocity and stress in the fluid near individual particles. The averaging volume V contains many particles, and the statistical properties of the suspension are stationary over V (implying that, in the case of a suspension which is inhomogeneous in the large, V must be suitably restricted in magnitude). The volume and surface of a typical particle in V will be denoted by V_0 and A_0. Then the bulk stress in the suspension is

$$\Sigma_{ij} = \frac{1}{V}\int (\sigma_{ij} - \rho u_i' u_j')\, dV,$$

and, since the ambient fluid is Newtonian, with viscosity μ, we have

$$\Sigma_{ij} = \frac{1}{V}\int_{V-\Sigma V_0}\left\{-p\delta_{ij} + \mu\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)\right\} dV + \frac{1}{V}\Sigma\int_{V_0}\sigma_{ij}\, dV - \frac{1}{V}\int \rho u_i' u_j'\, dV, \tag{4.1}$$

where the summation is over all the particles in V.

The volume-averaged velocity gradient, which plays an important part in the analysis and will be denoted by $\partial U_i/\partial x_j$ henceforth instead of $\partial \overline{u}_i/\partial x_j$, is

$$\frac{\partial U_i}{\partial x_j} = \frac{1}{V}\int \frac{\partial u_i}{\partial x_j}\, dV$$
$$= \frac{1}{V}\int_{V-\Sigma V_0}\frac{\partial u_i}{\partial x_j}\, dV + \frac{1}{V}\Sigma\int_{A_0} u_i n_j\, dA, \tag{4.2}$$

where n_i is an outward unit normal to A_0.

Also, we may write

$$\int_{V_0}\sigma_{ij}\, dV = \int_{A_0}\sigma_{ik} x_j n_k\, dA - \int_{V_0}\frac{\partial \sigma_{ik}}{\partial x_k} x_j\, dV. \tag{4.3}$$

It may happen that the interface of the ambient fluid and the particle is such that a surface tension must be supposed to act there. In such a case it is necessary to know whether the surface A_0 over which the integral in (4.3) is taken lies on the outside or the inside of the interfacial layer. Since we have already assumed that the volume $V - \Sigma V_0$ is wholly occupied by ambient fluid in which the stress has the Newtonian form, the volume V_0 must be regarded as including the interfacial layer. Thus the surface A_0 lies on the *outside* of the interfacial layer, and the stress in the surface integral in (4.3) is the boundary value of the Newtonian stress in the ambient fluid. It follows also that the range of integration for the volume integral on the right-hand side of (4.3) includes the interfacial layer within which some components of the stress tensor have very large values, but the stress divergence is non-singular everywhere and the interfacial layer makes no contribution to this volume integral.†

As already stated, we are assuming that the external body force per unit volume is uniform throughout the suspension. A uniform body force per unit volume can of course be equilibrated by a linearly varying isotropic stress which may be ignored. Hence, with allowance for inertia forces, we may put

$$\partial \sigma_{ij}/\partial x_j = \rho f_i' \tag{4.4}$$

at each point of the suspension, where f_i' is the local acceleration relative to the average value of the acceleration in V and the density ρ is assumed to be uniform throughout the suspension.

† Further explanation of the role of surface tension is given in the appendix.

With the aid of (4.2), (4.3) and (4.4), we may write the expression (4.1) for the bulk stress as

$$\Sigma_{ij} = -\frac{\delta_{ij}}{V}\int_{V-\Sigma V_0} p\,dV + \mu\left(\frac{\partial U_i}{\partial x_j}+\frac{\partial U_j}{\partial x_i}\right) + \Sigma_{ij}^{(p)},$$

where

$$\Sigma_{ij}^{(p)} = \frac{1}{V}\Sigma\int_{A_0}\{\sigma_{ik}x_j n_k - \mu(u_i n_j + u_j n_i)\}\,dA - \frac{1}{V}\Sigma\int_{V_0}\rho f'_i x_j\,dV - \frac{1}{V}\int \rho u'_i u'_j\,dV. \tag{4.5}$$

The first term in this expression for Σ_{ij} is a purely isotropic contribution of no particular interest; the second term is the deviatoric stress that would exist in the absence of the particles and with the same average velocity gradient; and the third represents the contribution to the bulk stress due to the presence of the particles. We shall call $\Sigma_{ij}^{(p)}$ the 'particle stress'. Only the deviatoric part of the particle stress is significant.

It may now be seen that the exertion of a couple on the particles by external means generates an antisymmetrical contribution to the bulk stress. The angular momentum equation for one particle gives

$$L_i = \epsilon_{ijk}\int_{A_0}\sigma_{jl}x_k n_l\,dA - \epsilon_{ijk}\int_{V_0}\rho f'_j x_k\,dV = \epsilon_{ijk}\int_{V_0}\sigma_{jk}\,dV, \tag{4.6}$$

where L_i is the pure couple exerted on the particle by external means, and it follows from (4.5) that

$$\epsilon_{ijk}\Sigma_{jk} = \epsilon_{ijk}\Sigma_{jk}^{(p)} = \frac{1}{V}\Sigma L_i.$$

The pure couple exerted on the particles thus accounts for the whole of the antisymmetrical part of the bulk stress. It would be tempting to suppose that the symmetrical part of the bulk stress is unaffected by the couple, but we see explicitly later that this is not so in general. We note also for later use that a relation like (4.6) holds for any small element of material, within which the stress tensor is approximately uniform, whence

$$\epsilon_{ijk}\sigma_{jk} = \mathscr{L}_i,$$

where $\mathscr{L}_i$ is the externally imposed couple per unit volume of material. This is a local relation, valid at each point of the suspension; at points in the ambient fluid $\mathscr{L}_i = 0$, whereas for the points within one particle we have

$$\int_{V_0}\mathscr{L}_i\,dV = L_i.$$

An alternative form of (4.5) which shows the symmetrical and antisymmetrical parts of the bulk stress tensor separately is thus

$$\Sigma_{ij}^{(p)} = \frac{1}{V}\Sigma\int_{A_0}\{\tfrac{1}{2}(\sigma_{ik}x_j+\sigma_{jk}x_i)\,n_k - \mu(u_i n_j + u_j n_i)\}\,dA + \frac{1}{2V}\Sigma\,\epsilon_{ijk}L_k$$
$$-\frac{1}{V}\Sigma\int_{V_0}\tfrac{1}{2}\rho(f'_i x_j + f'_j x_i)\,dV - \frac{1}{V}\int\rho u'_i u'_j\,dV. \tag{4.7}$$

Also of interest is the rate of dissipation of energy in the suspension. Dissipation occurs only through the action of internal friction at a rate $\frac{1}{2}\partial u_i/\partial x_j(\sigma_{ij}+\sigma_{ji})$ per unit volume at each point,† and the average rate over the volume V of the suspension is

$$\begin{aligned}\Phi &= \frac{1}{V}\int \frac{1}{2}\frac{\partial u_i}{\partial x_j}(\sigma_{ij}+\sigma_{ji})\,dV \\ &= \frac{1}{2}\frac{\partial U_i}{\partial x_j}(\overline{\sigma}_{ij}+\overline{\sigma}_{ji})+\frac{1}{V}\int\left\{\frac{\partial(u_i'\sigma_{ij})}{\partial x_j}-u_i'\frac{\partial\sigma_{ij}}{\partial x_j}-\frac{1}{2}\frac{\partial u_i'}{\partial x_j}(\sigma_{ij}-\sigma_{ji})\right\}dV \\ &= \frac{1}{2}\frac{\partial U_i}{\partial x_j}(\Sigma_{ij}+\Sigma_{ji})+\frac{\partial\overline{u_i'\sigma_{ij}}}{\partial x_j}+\frac{1}{V}\int\left\{\rho u_i'u_j'\frac{\partial U_i}{\partial x_j}-\rho u_i'f_i'+\tfrac{1}{2}\omega_i'\mathscr{L}_i\right\}dV, \end{aligned} \tag{4.8}$$

where $\boldsymbol{\omega} = \nabla\times\mathbf{u}$ is the local vorticity and $\boldsymbol{\omega}'$ is its fluctuating part. The second term on the right-hand side of (4.8) is non-zero only when the suspension is statistically non-uniform. Since the orders of magnitude of u_i' and of the variable part of σ_{ij} are ed and μe in the neighbourhood of a particle of dimension d and smaller elsewhere, where e is a representative magnitude of the bulk rate of strain, the ratio of the second term to μe^2 is never larger than the ratio of d to the distance over which the state of the suspension changes appreciably; thus the second term is negligibly small under the assumed conditions. (Use of the divergence theorem shows that this second term is zero if the velocity at the boundary of the volume V is exactly a linear function of position so that $\mathbf{u}' = 0$ there, and this condition has been adopted by most authors. Such a restriction seems artificial—although of course it fits the circumstances of some experiments—and the above argument shows that it is unnecessary.) In the third term on the right-hand side of (4.8) we use the fact that f_i' is the local fluctuation in acceleration to make the substitution

$$f_i' = \frac{\partial u_i'}{\partial t}+u_j'\frac{\partial U_i}{\partial x_j}+U_j\frac{\partial u_i'}{\partial x_j}+\frac{\partial}{\partial x_j}(u_i'u_j'-\overline{u_i'u_j'}).$$

Then, with neglect of another spatial derivative of an averaged quantity with similar justification, (4.8) becomes

$$\Phi+\left(\frac{\partial}{\partial t}+U_j\frac{\partial}{\partial x_j}\right)\tfrac{1}{2}\rho\overline{u_i'^2} = \frac{1}{2}\frac{\partial U_i}{\partial x_j}(\Sigma_{ij}+\Sigma_{ji})+\frac{1}{V}\Sigma\int_{V_0}\tfrac{1}{2}\omega_i'\mathscr{L}_i\,dV. \tag{4.9}$$

The interpretation of this relation when the imposed couple is zero is that the apparent rate of dissipation per unit volume, as estimated from the bulk quantities $\partial U_i/\partial x_j$ and $\frac{1}{2}(\Sigma_{ij}+\Sigma_{ji})$, gives the sum of the true dissipation of mechanical energy into heat and the rate of increase of 'microscopic' kinetic energy per unit volume of a material element moving with the mean flow; this sum is the rate at which bulk mechanical energy is being lost, and so is an effective rate of dissipation. However, when a couple is imposed, this apparent rate of dissipation as estimated from the bulk quantities must be supplemented by the second term on the right-hand side of (4.9) (which becomes $\Sigma\frac{1}{2}\omega_i'L_i/V$ when the particles are rigid) representing the rate of working by the externally imposed couple

† When surface tension acts at the particle boundaries, points on these interfaces require separate consideration. The consequent modifications to (4.8) and (4.9) are described in the appendix.

associated with the fluctuation in angular velocity. Thus here the rate of dissipation cannot in general be determined from the bulk stress and velocity gradient alone.

The expression (4.5), and the alternative form (4.7), hold for any shape of size or type of force-free particles, for any concentration of the particles, and for any magnitude of the average velocity gradient (provided only that the stress in the ambient fluid is Newtonian). These expressions contain allowance for two effects not normally included in investigations of the average stress in a suspension. One is the effect of rotational motion of the particles relative to the ambient fluid under the action of a couple imposed by external means, although the form of (4.5) is not affected explicitly by this effect. The other is the effect of inertia forces associated with the fluctuations about the average motion inside and outside the particles, which gives rise to the last two terms in (4.5) or (4.7). Both effects influence the values of σ_{ij} and u_i on the surface A_0. The form taken by (4.5) in the absence of these two effects appears to have been given first by Landau & Lifschitz (1959, §22).

The non-linear inertial effects associated with the fluctuations about the average motion are likely to be difficult to calculate in a specific case, and it is therefore important to know when they may be neglected. Completely general conditions are complicated, but we get some insight from a consideration of the Reynolds number of the disturbance or fluctuating motion arising from different causes separately. For simplicity suppose that a particle has the same representative linear dimension d in all directions. The velocity fluctuation caused by the presence of the particle in the imposed bulk straining motion (with e as a representative magnitude of the average rate of strain) is of order ed, and the corresponding Reynolds number for the disturbance motion in the ambient fluid is $ed^2\rho/\mu$. Another source of disturbance motion is the couple imposed by external means on the particle, which causes it to rotate with angular speed Ω relative to the ambient fluid and so produces a fluid motion whose Reynolds number is $\Omega d^2\rho/\mu$. Thus for these disturbance motions in the fluid to be free from the effect of inertia forces we require

$$\frac{ed^2\rho}{\mu} \ll 1, \quad \frac{\Omega d^2\rho}{\mu} \ll 1. \tag{4.10}$$

These conditions will frequently be satisfied in practice. (For a low-viscosity fluid like water, the condition that the Reynolds number of the translational motion of a particle due to gravity should be small will often be a more severe restriction on particle size.)

In the remainder of this paper we shall suppose that the effect of inertia forces associated with the fluctuations about the average motion is negligible. In these circumstances the last two terms in (4.5) and (4.7) may be ignored and we have for the contribution to the bulk stress due to the presence of the particles

$$\Sigma_{ij}^{(p)} = \frac{1}{V}\Sigma\int_{A_0}\{\sigma_{ik}x_j n_k - \mu(u_i n_j + u_j n_i)\}\,dA \tag{4.11a}$$

$$= \frac{1}{V}\Sigma\int_{A_0}\{\tfrac{1}{2}(\sigma_{ik}x_j + \sigma_{jk}x_i)\,n_k - \mu(u_i n_j + u_j n_i)\}\,dA + \frac{1}{2V}\Sigma\,\epsilon_{ijk}L_k. \tag{4.11b}$$

It will be noticed that, since no excess body force acts on a particle and momentum changes are being neglected, the point about which the first moment of the stress at the particle surface is calculated is arbitrary; it is thus permissible to choose with convenience a different origin for each particle.

The determination of σ_{ij} and u_i at the surface of each particle has now been reduced to a linear slow-viscous-motion problem. It is evident that the fluctuations in both σ_{ij} and u_i will be linear functions of the average velocity gradient in V, for either a dilute or concentrated suspension of particles of permanent form, and that they will vanish with this gradient, when the particles have neither translational nor rotational motion due to external influences. Thus in these latter circumstances the average deviatoric stress due to the particles is proportional to $\partial U_i/\partial x_j$ and the suspension is quasi-Newtonian, as expected from the general arguments in § 2.

Finally we notice alternative forms for the deviatoric part of (4.11) which will be needed later. At the boundary of a rigid particle, the no-slip condition gives $\mathbf{u}$ in terms of the particle velocity, and the integral of $u_i n_j + u_j n_i$ over the surface A_0 is zero for either translational or rotational motion (or a combination thereof) of a rigid particle. Nevertheless, we leave $u_i n_j + u_j n_i$ in the integrands in (4.11) not only to allow the possibility of application of (4.11) to non-rigid particles, but also because in the above form the integrals may be taken (when inertia forces are negligible) over any closed surface A lying entirely in the ambient fluid and enclosing the particle A_0 but no other particle. Also, if we define the stress fluctuation σ'_{ij} by

$$\sigma_{ij} = -p_0\delta_{ij} + \mu\left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i}\right) + \sigma'_{ij}, \tag{4.12}$$

where p_0 is constant in V, and $\mathbf{u} = \mathbf{U} + \mathbf{u}'$ as before, it may be seen that σ_{ij} and u_i in (4.11) may be replaced by σ'_{ij} and u'_i. And the replacement of the surface A_0 by A is still possible when this change has been made.

It is difficult to go beyond this point in the investigation of the stress in a concentrated suspension and very few definite results have been obtained. In particular, we do not yet know which statistical parameters of the state of the suspension determine the bulk stress. However, progress can be made in the case of a dilute suspension.

5. The particle stress in a dilute suspension

As the average distance between the particles in a suspension increases, the distributions of velocity and stress at the surface of one particle are less affected by the presence of all the others. And, in the limit $c \to 0$ (where c is the concentration of particles by volume), the flow near each particle is that which would be set up if it alone were immersed in an infinite body of ambient fluid with a uniform velocity gradient at large distances from the particle. Furthermore, the limiting value of this velocity gradient, as $c \to 0$, must be the average gradient $\partial U_i/\partial x_j$. The manner in which these limits are approached is not obvious (and needs to be considered in an investigation of the next approximation to the particle stress), but there can be no doubt about the limits themselves. The relation (4.11) shows that under these conditions the different particles in the volume V of the suspension make linearly additive contributions to the particle stress, and that the particle stress so obtained from (4.11) is correct to the order of the first power of the number density of the particles, that is, to the order of c. This is the standard basis of dilute suspension theory.

We now consider in general terms the flow near a single particle, on which an externally imposed couple (but no force) may act, with the assumption that the Reynolds number of the relative motion near the particle is sufficiently small for inertia forces to be negligible. Far from the particle the velocity gradient in the fluid is uniform and equal to the average velocity gradient in the whole suspension, which we shall write as

$$\partial U_i/\partial x_j = e_{ij} - \epsilon_{ijk}\,\Omega_k; \tag{5.1}$$

that is, the flow far from the particle is represented as the superposition of a pure straining motion characterized by the rate-of-strain tensor e_{ij} and a rigid-body rotation with angular velocity Ω_i. The velocity, pressure and stress in the fluid will be written as

$$\left.\begin{aligned} u_i = x_j\frac{\partial U_i}{\partial x_j} + u_i', \quad p = p_0 + p', \\ \sigma_{ij} = -p_0\,\delta_{ij} + 2\mu e_{ij} + \sigma_{ij}', \end{aligned}\right\} \tag{5.2}$$

where p_0 is a constant and u_i', p', σ_{ij}' are the disturbance quantities resulting from the presence of the particle. In the present circumstances, u_i', p' and σ_{ij}' are determinate, not random, quantities. The disturbance quantities satisfy the slow-viscous-motion equations

$$\left.\begin{aligned} \nabla p' = \mu\nabla^2\mathbf{u}' = -\nabla\times\boldsymbol{\omega}', \\ \nabla\,.\,\mathbf{u}' = 0, \end{aligned}\right\} \tag{5.3}$$

where $\boldsymbol{\omega}'$ is the disturbance vorticity $\nabla\times\mathbf{u}'$. The outer boundary condition is

$$u_i' \to 0, \quad p' \to 0, \quad \text{as} \quad r \to \infty,$$

where $r = |\mathbf{x}|$.

The inner boundary conditions to be satisfied by u_i' and p' depend on the constitution of the particle. If the material in the particle is undergoing deformation, the disturbance quantities inside the particle satisfy differential equations which may be different from (5.3), and the internal and external distributions of velocity and stress must satisfy the usual matching conditions at the particle surface; and the shape of the particle surface must itself be determined as a part of the matching problem. Investigation of the flow in and near a deformable particle presents a variety of special problems (contributions to which have been made by Taylor (1932, 1934), Roscoe (1967), Goddard & Miller (1967), and Cox (1969)), and inclusion of this case in the present general discussion would require rather complicated statements. I shall therefore consider here only the case of a rigid particle; this allows examination of the effects of particle shape and of an externally applied couple, and, if the more complete expression for particle stress given in § 4 is retained, of inertia forces in the relative motion near a particle.

Satisfaction of the no-slip condition at the surface A_0 of a rigid particle requires

$$u_i = v_i + \epsilon_{ijk}(\Omega_j + \Gamma_j)\, x_k \tag{5.4}$$

on A_0, where $\mathbf{v}$ is the velocity of the material point of the particle coinciding instantaneously with the origin and $\mathbf{\Gamma}$ is the angular velocity of the particle relative to that of the undisturbed ambient fluid. In this linear problem, the two causes of disturbance motion make independent contributions to $\mathbf{v}$ and $\mathbf{\Gamma}$, for given instantaneous position and orientation of the particle, and it is convenient to represent them separately. There are first the contributions $\mathbf{v}^{(s)}$, $\mathbf{\Gamma}^{(s)}$ due wholly to the straining motion in the undisturbed ambient fluid, which are determined by the two conditions that the fluid stress exerted at the surface of the particle yields zero force and zero couple on the particle; $\mathbf{v}^{(s)}$ and $\mathbf{\Gamma}^{(s)}$ depend only on the rate-of-strain tensor e_{ij}, aside from the shape and orientation of the particle, and are evidently linear functions of e_{ij}. Secondly, there will be contributions $\mathbf{v}^{(c)}$, $\mathbf{\Gamma}^{(c)}$ due to the couple $\mathbf{L}$ imposed on the particle by external means and determined by the conditions that the fluid stress at the surface of the particle, when it is moving in fluid at rest at infinity and instantaneously with the given position and orientation, yields zero force and a couple on the particle equal to $-\mathbf{L}$; $\mathbf{v}^{(c)}$ and $\mathbf{\Gamma}^{(c)}$ are linear functions of $\mathbf{L}$. On adding these contributions we obtain the inner boundary condition on the disturbance velocity in the form

$$u'_i = v_i^{(s)} + v_i^{(c)} + \epsilon_{ijk}(\Gamma_j^{(s)} + \Gamma_j^{(c)})\, x_k - e_{ij} x_j \tag{5.5}$$

on the surface A_0.

The conditions specified are now sufficient for the determination of the disturbance quantities u'_i, p', σ'_{ij}, and the contribution to the particle stress in the suspension due to the presence of this particular particle may then be calculated in principle from (4.11). Detailed results are available for an ellipsoidal rigid particle, and will be referred to in the next section. In this section we show some general features of the problem which apply irrespective of the shape of the particle.

These general features are associated with the asymptotic forms of the disturbance quantities far from the particle. Both the pressure and the vorticity in inertia-less flow of viscous fluid satisfy Laplace's equation, and can be written as series of spherical harmonics on and outside a sphere enclosing the body, the coefficients in these two series being related through (5.3). The forms of these series for a case in which, as here, p' and ω'_i both tend to zero at infinity are

$$\frac{p'}{\mu} = -D_j \frac{\partial r^{-1}}{\partial x_j} - D_{jk} \frac{\partial^2 r^{-1}}{\partial x_j \partial x_k} + \ldots, \tag{5.6}$$

$$\omega'_i = -\epsilon_{ijk} D_j \frac{\partial r^{-1}}{\partial x_k} - \epsilon_{ijk} D_{jl} \frac{\partial^2 r^{-1}}{\partial x_k \partial x_l} + \ldots, \tag{5.7}$$

where D_j, D_{jk}, ... are coefficients which are determined by the inner boundary condition. (The spherical harmonic r^{-1} does not appear because it is associated with a disturbance velocity which does not vanish as $r \to \infty$.) The first terms represent the type of singularity at the origin commonly called a Stokeslet, and it is known that the coefficient $\mathbf{D}$ is $1/4\pi\mu$ times the resultant force exerted on the fluid at the inner boundary; thus $\mathbf{D} = 0$ here. The second terms in the series represent the effect of a force doublet at the origin, about which more will be said in a moment.

The terms in the corresponding series for the disturbance velocity and stress are not spherical harmonics, but are simply related to them, and may be written down (Lamb 1932). Only the leading terms in the series will be needed for the present purposes, and these alone will be given explicitly. For the disturbance velocity we have

$$u_i' = D_{jk}\left(-\frac{3x_i x_j x_k}{2r^5} + \frac{x_j\delta_{ik} - x_k\delta_{ij}}{r^3}\right) + \ldots; \tag{5.8}$$

and the condition of zero volume flux across a surface enclosing the particle requires

$$D_{ii} = 0. \tag{5.9}$$

The series expression for the disturbance stress follows as

$$\begin{aligned}\sigma_{ij}' &= -p'\delta_{ij} + \mu\left(\frac{\partial u_i'}{\partial x_j} + \frac{\partial u_j'}{\partial x_i}\right)\\ &= \mu D_{kl}\left\{-\frac{3x_k(x_i\delta_{jl} + x_j\delta_{il})}{r^5} + \frac{15x_i x_j x_k x_l}{r^7}\right\} + \ldots.\end{aligned} \tag{5.10}$$

Now since the integral

$$\int\{\sigma_{ik}' x_j n_k - \tfrac{1}{3}\delta_{ij}\sigma_{lk}' x_l n_k - \mu(u_i' n_j + u_j' n_i)\}\, dA \tag{5.11}$$

has the same value when taken over any closed surface surrounding the particle, we are free to choose a sphere centred at the origin. It is evident that only the terms of even degree in r in the series for u_i', and the corresponding terms of odd degree in the series for σ_{ij}', make a non-zero contribution to the integral. Moreover, it follows from the general form of these terms and various integral theorems for spherical harmonics (Happel & Brenner 1965, § 3.2) that only the first such term of even degree in the series (5.8) contributes; alternatively, we can evaluate the integral for the particular choice of a sphere of large radius. Either way, the integral (5.11) is found to be equal to $4\pi\mu D_{ij}$. It follows then from the variant of (4.11 *a*) with u_i and σ_{ij} replaced by u_i' and σ_{ij}' that the particle stress in the suspension is given by

$$\Sigma_{ij}^{(p)} = \frac{4\pi\mu}{V}\Sigma D_{ij}, \tag{5.12}$$

the summation being over all the particles in V as before.

We also see from (4.6) that, with neglect of the inertial effect represented by the second integral,

$$L_i = 4\pi\mu\epsilon_{ijk}D_{jk},$$

that is

$$\tfrac{1}{2}(D_{ij} - D_{ji}) = \epsilon_{ijk}L_k/4\pi\mu. \tag{5.13}$$

The alternative form of (5.12) which shows the symmetrical and antisymmetrical parts of $\Sigma_{ij}^{(p)}$ separately, as in (4.11 *b*), is then

$$\Sigma_{ij}^{(p)} = \frac{4\pi\mu}{V}\Sigma\,\tfrac{1}{2}(D_{ij} + D_{ji}) + \frac{1}{2V}\Sigma\,\epsilon_{ijk}L_k. \tag{5.14}$$

The singularity at the origin that is associated with the asymptotic form of the disturbance motion due to a particle, and that represents the particle completely so far as its effect on the bulk stress in a dilute suspension is concerned, thus has two parts, one of which may be termed a *couplet* characterized by the axial vector L_i and the other a *stresslet* characterized by the symmetrical tensor $S_{ij} = 4\pi\mu\frac{1}{2}(D_{ij}+D_{ji})$, in which the factor $4\pi\mu$ has been included in order to achieve a numerical correspondence between S_{ij} and the effect of a force doublet at the origin. On rewriting the above relations in terms of the two quantities L_i and S_{ij}, we have the properties shown in table 1.

	Stresslet	Couplet
Disturbance pressure, p'	$-\frac{S_{ij}}{4\pi}\frac{\partial^2 r^{-1}}{\partial x_i \partial x_j}$	0
Disturbance vorticity, ω'_i	$-\epsilon_{ijk}\frac{S_{jl}}{4\pi\mu}\frac{\partial^2 r^{-1}}{\partial x_k \partial x_l}$	$\frac{L_j}{8\pi\mu}\frac{\partial^2 r^{-1}}{\partial x_i \partial x_j}$
Disturbance velocity, u'_i	$-\frac{3}{2}\frac{S_{jk}}{4\pi\mu}\frac{x_i x_j x_k}{r^5}$	$\epsilon_{ijk}\frac{L_j}{8\pi\mu}\frac{x_k}{r^3}$
Disturbance stress, σ'_{ij}	$\frac{S_{kl}}{4\pi}\frac{3x_k}{r^4}\left(-\frac{x_i\delta_{jl}+x_j\delta_{il}}{r}+\frac{5x_i x_j x_l}{r^3}\right)$	$\frac{3L_l}{8\pi}\frac{x_k}{r^5}(\epsilon_{jkl}+\epsilon_{ikl}x_j)$
Moment of disturbance surface force over a sphere of large radius, $\int\sigma'_{ik}x_j n_k dA$	$\frac{2}{3}S_{ij}$	$\frac{1}{2}\epsilon_{ijk}L_k$
Contribution to bulk stress in dilute suspension of total volume V	$\frac{S_{ij}}{V}$	$\frac{\frac{1}{2}\epsilon_{ijk}L_k}{V}$

TABLE 1. Properties of the distant flow field associated with a single force-free particle

The streamlines due to a stresslet at the origin are all radial lines, and the radial component of the velocity varies over the surface of a sphere in proportion to

$$-S_{11}\cos^2\theta - S_{22}\sin^2\theta\cos^2\phi - S_{33}\sin^2\theta\sin^2\phi,$$

where (r, θ, ϕ) are spherical polar co-ordinates with $\theta = 0$ in the direction of one principal axis of the tensor S_{ij} and $\theta = \frac{1}{2}\pi$, $\phi = 0$ in the direction of another. The streamlines due to a couplet, on the other hand, are circles about the line through the origin parallel to L_i, and spherical shells of fluid rotate rigidly with angular velocity $L_i/8\pi\mu r^3$ (as in inertia-less flow due to a rotating rigid sphere). These elementary properties are illustrated in figure 1, where it is also indicated that a couplet and each principal element of the stresslet tensor S_{ij} are equivalent to equal and opposite forces (or Stokeslets) applied to the fluid at two neighbouring points near the origin, the line joining the two points being normal to the forces in one case and parallel to them in the other.

It would be natural to suppose that the stresslet tensor S_{ij} is determined wholly by the need for the no-slip condition to be satisfied at the surface of a rigid particle immersed in fluid undergoing a pure straining motion and that it is independent of L_i. However, consideration of an ellipsoidal particle in the next section shows that this is false, except when the particle is spherical. It appears that, if a non-spherical particle is made to rotate by the action of an externally imposed couple, in the absence of any bulk deformation of the suspension, the resulting particle stress has a non-zero antisymmetrical part, as given by the second term on the right-hand side of (5.14), *and* a non-zero symmetrical part. This can be understood in terms of the stress acting at the surface of the rotating particle. When the disturbance surface force is everywhere normal to the position vector, as it is for (and only for) a rotating sphere and as is symbolized in figure 1 (*b*), the net effect of the surface force is a pure couple; but a non-zero component of the surface force parallel to the position vector will yield a resultant tension of the kind symbolized in figure 1 (*a*).

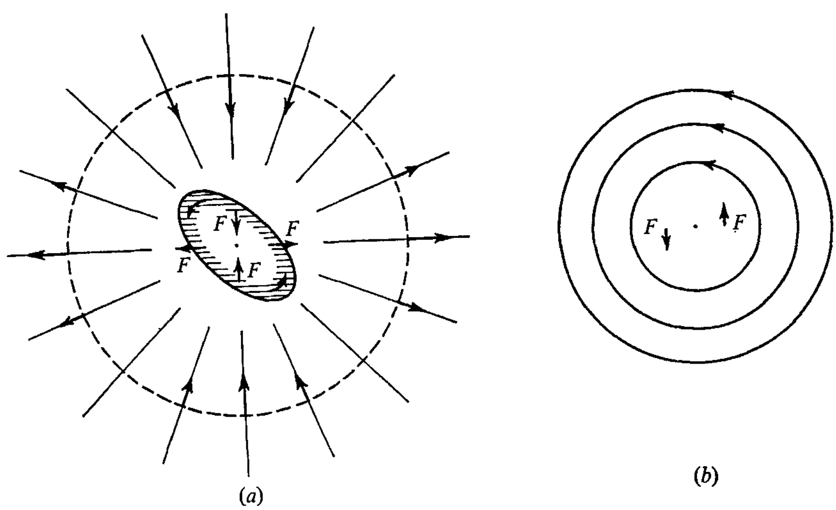

FIGURE 1. Streamlines in the plane normal to the x_3-axis for (*a*) a stresslet such that the principal axes of the defining tensor S_{ij} coincide with the co-ordinate axes and

$$S_{11} = -S_{22} = -2aF, \quad S_{33} = 0;$$

and (*b*) a couplet such that $L_1 = L_2 = 0$, $L_3 = 2aF$. The opposing forces of magnitude F are separated by a distance $2a$ in every case. The ellipse in (*a*) shows the position of an ellipsoid rotating about the x_3-axis under the action of an imposed couple which would generate a symmetrical particle stress of this kind (§6).

It is thus necessary to allow for two contributions to S_{ij}, one arising from the presence of the particle in a pure straining motion of the ambient fluid and necessarily linear in e_{ij}, and the other arising from the rotation of the particle under the action of the imposed couple and necessarily linear in L_i. We may write

$$\tfrac{1}{2}(D_{ij}+D_{ji}) = \frac{1}{4\pi\mu}S_{ij} = C_{ijkl}e_{kl}+\frac{1}{8\pi\mu}C_{ijk}L_k, \tag{5.15}$$

where C_{ijkl} and C_{ijk} are symmetrical in i and j and depend only on the shape, size and instantaneous orientation of the particle; also, in view of (5.9),

$$C_{iikl} = 0, \quad C_{iik} = 0,$$

and we may also suppose that C_{ijkl} is symmetrical in k and l since only its contraction with e_{kl} is relevant. Hence the particle stress in a dilute suspension of total volume V is

$$\Sigma_{ij}^{(p)} = \frac{1}{V}\Sigma\{4\pi\mu C_{ijkl}e_{kl} + \tfrac{1}{2}(C_{ijk}+\epsilon_{ijk})L_k\}, \tag{5.16}$$

the summation being over all the particles in V. For the particular case of a dilute suspension of particles in a Newtonian ambient fluid we have thus been able to show, without hypothesis, that the suspension is quasi-Newtonian; and comparison with the general form (2.1) for the symmetrical part of the stress due to the bulk deformation shows that

$$A_{ijkl} = \frac{4\pi\mu}{V}\Sigma C_{ijkl} \tag{5.17}$$

for a dilute suspension.

In all the above analysis, we have regarded the pure couple $\mathbf{L}$ exerted on the particle by external means as a given quantity. The effect of the couple is to give the particle an additional motion relative to the ambient fluid which is characterized by the quantities $\mathbf{v}^{(c)}$ and $\mathbf{\Gamma}^{(c)}$ (see (5.5)), both of which are linear functions of $\mathbf{L}$, and the viscous stresses set up at the particle surface by this motion then transmit the couple to the fluid. If the couple is exerted by means of an externally applied magnetic field acting on a particle with a magnetic moment, as in the recent experiments of McTague (1969), the couple depends on the particle orientation and may take a value which is itself affected by the bulk motion. For instance, if the magnetic field is sufficiently strong to prevent the magnetic axis of the particle from being turned by the bulk motion through more than a small angle, the value of the couple can be determined from hydrodynamic considerations and will clearly be a linear function of $\partial U_i/\partial x_j$. In these circumstances the rheological relation (5.16) takes a form which may be used conveniently for the determination of a whole flow field.

It is desirable to get some idea of the way in which the tensor coefficients C_{ijkl} and C_{ijk}, and the coefficients in the linear relations between $\mathbf{L}$ and $\mathbf{v}^{(c)}$ and $\mathbf{\Gamma}^{(c)}$, depend on particle shape and orientation. Ellipsoidal particles make a very convenient vehicle for a consideration of shape effects and we proceed to this case.

6. A dilute suspension of rigid ellipsoidal particles

The foregoing relations for a dilute suspension simplify a little when the surface of each particle is symmetrical about each of three orthogonal planes. Symmetry arguments show that, when such a rigid particle rotates about any axis through its centre in fluid at rest at infinity, the force exerted on it by the fluid is zero;† when it is in translational motion through fluid at rest at infinity, the couple exerted on it by the fluid is zero;† and, when it is immersed in fluid which is in pure straining motion far from the particle, the force on the particle

† These results for an orthotropic particle are given in the book by Happel & Brenner (1965, §5.6).

is zero if the centre moves with the velocity of the undisturbed pure straining motion at that same point. As a consequence, the centre of a force-free particle of this shape immersed in fluid which has the uniform velocity gradient (5.1) at infinity moves with a velocity equal to that of the undisturbed fluid at the same point, and we may put

$$v_i^{(s)} = 0, \quad v_i^{(c)} = 0$$

in (5.5). Moreover, the coefficient in the linear relation between the externally imposed couple $\mathbf{L}$ and the resulting angular velocity $\mathbf{\Gamma}^{(c)}$ of the particle in fluid at rest at infinity, namely

$$L_i = k_{ij}\Gamma_j^{(c)}, \tag{6.1}$$

is evidently a second-order tensor with non-diagonal elements which are zero when referred to axes coinciding with the intersections of the three planes of symmetry of the particle. There are presumably similar implications for the tensor coefficients C_{ijkl} and C_{ijk}, although these are less evident.

An ellipsoid has the above kind of symmetry, and it happens that complete detailed results for a dilute suspension of rigid ellipsoids can be obtained by using the well-known work by Jeffery (1922). Many writers on dilute suspension theory have made use of Jeffery's results to calculate the additional rate of dissipation due to the presence of couple-free particles, and have interpreted this in terms of an increased apparent viscosity of the suspension. As (5.17) indicates, such an interpretation is correct only for certain symmetry properties of the instantaneous distribution of particle orientations, and this in turn depends on the nature of the bulk motion and perhaps on the distribution at some initial time; failure to appreciate this has led some writers to describe the bulk properties of the suspension in terms of an effective viscosity in circumstances in which the stress system may not be of Newtonian form. The safe plan is to calculate the particle stress due to a particle of specified shape and orientation, which reduces to a determination of the tensor C_{ijkl} and C_{ijk}, and then to add the contributions from the different particles taking into account the distribution of their orientations which results from rotation in the given bulk motion (and from Brownian motion, in the case of very small particles).

Jeffery shows (see his equations (22), (23), (24) with (50)) that, when a rigid ellipsoid on which a couple, but no force, is imposed by external means is embedded in a pure straining motion, the disturbance velocity far from the ellipsoid is of the form (5.8) and that the components of the tensor D_{ij} for axes coinciding instantaneously with the principal axes of the ellipsoid are

$$\left.\begin{aligned} &\overline{D}_{11} = \tfrac{8}{3}A, \quad \overline{D}_{22} = \tfrac{8}{3}B, \quad \overline{D}_{33} = \tfrac{8}{3}C, \quad \overline{D}_{12} = \tfrac{8}{3}H, \quad \overline{D}_{21} = \tfrac{8}{3}H', \\ &\overline{D}_{23} = \tfrac{8}{3}F, \quad \overline{D}_{32} = \tfrac{8}{3}F', \quad \overline{D}_{31} = \tfrac{8}{3}G, \quad \overline{D}_{13} = \tfrac{8}{3}G', \end{aligned}\right\} \tag{6.2}$$

where expressions for the coefficients A, B, ... in terms of the angular velocity of the ellipsoid and the ambient rate of strain are available. This formula for the particle stress (which is what it is, effectively, in view of (5.12)) has been noted by Giesekus (1962) but has been otherwise neglected. Here we go a little further and obtain explicit expressions for C_{ijkl} and C_{ijk} and the angular resistance coefficient k_{ij}.

With unit vectors parallel to the principal axes of the ellipsoid denoted by $\mathbf{p}, \mathbf{q}, \mathbf{r}$, the relation between the components of the tensor D_{ij} for the general and special sets of axes is

$$D_{ij} = \bar{D}_{kl}\gamma_{ik}\gamma_{jl},$$

where $\quad \gamma_{i1} = \gamma_{1i} = p_i, \quad \gamma_{i2} = \gamma_{2i} = q_i, \quad \gamma_{i3} = \gamma_{3i} = r_i,$

and so

$$D_{ij} = \tfrac{8}{3}(Ap_ip_j + Bq_iq_j + Cr_ir_j + Hp_iq_j + H'p_jq_i + Fq_ir_j + F'q_jr_i + Gr_ip_j + G'r_jp_i). \qquad (6.3)$$

Jeffery's expressions for $A, B, \ldots$ (his equations (25), (26)) involve the components of the rate-of-strain tensor referred to his special choice of axes, and we must make the substitutions

$$\bar{e}_{11} = e_{kl}\gamma_{k1}\gamma_{l1} = e_{kl}p_kp_l,$$

$$\bar{e}_{12} = e_{kl}\gamma_{k1}\gamma_{l2} = e_{kl}p_kq_l, \quad \text{etc.}$$

The expression for D_{ij} for an ellipsoid is then obtained as

$$D_{ij} = C_{ijkl}e_{kl} - \left\{\tfrac{8}{3}(F-F')\frac{b^2q_jr_i - c^2q_ir_j}{b^2+c^2} + \text{two similar terms}\right\}, \qquad (6.4)$$

where

$$\frac{C_{ijkl}}{abc} = \frac{J_1(p_ip_j - \frac{1}{3}\delta_{ij})\,p_kp_l + \ldots + \ldots}{\frac{3}{4}(J_1J_2 + J_2J_3 + J_3J_1)} + \left\{\frac{(q_ir_j + q_jr_i)(q_kr_l + q_lr_k)}{\frac{3}{2}I_1} + \ldots + \ldots\right\}. \qquad (6.5)$$

In these expressions a, b, c are the semi-diameters of the ellipsoid, and I_1, I_2, I_3 and J_1, J_2, J_3 are integrals like

$$I_1 = \int_0^\infty \frac{abc(b^2+c^2)\,d\lambda}{\Delta(b^2+\lambda)(c^2+\lambda)}, \quad J_1 = \int_0^\infty \frac{abc\lambda\,d\lambda}{\Delta(b^2+\lambda)(c^2+\lambda)}, \qquad (6.6)$$

where $\quad \Delta^2 = (a^2+\lambda)(b^2+\lambda)(c^2+\lambda).$

(In Jeffery's paper, $I_1/abc(b^2+c^2)$ and J_1/abc are denoted by α_0' and α_0'' respectively; I_1 and J_1 are dimensionless quantities depending only on the shape of the ellipsoid, which is convenient.)

Jeffery gives the relation between the angular velocity of the ellipsoid and the couple imposed on it (his equation (36)—but note that his couple (L, M, N) is minus the vector couple $\mathbf{L}$ used here), from which we find first the zero-couple relation

$$\Gamma_i^{(s)} = \left(\frac{b^2-c^2}{b^2+c^2}p_iq_jr_k + \ldots + \ldots\right)e_{jk}, \qquad (6.7)$$

and then the rotational resistance coefficient

$$k_{ij} = \frac{16\pi abc\mu}{3}\left\{\frac{p_ip_j}{J_1 + 2I_1b^2c^2(b^2+c^2)^{-2}} + \ldots + \ldots\right\}. \qquad (6.8)$$

He also shows that

$$F - F' = \frac{3}{32\pi\mu}L_k\,p_k, \quad G - G' = \frac{3}{32\pi\mu}L_kq_k, \quad H - H' = \frac{3}{32\pi\mu}L_kr_k. \qquad (6.9)$$

It is immediately clear that the antisymmetrical part of D_{ij}, as given by (6.4), is indeed of the expected form (5.13). The symmetrical part likewise has the expected form (5.15), and we have

$$C_{ijk} = -\frac{b^2-c^2}{b^2+c^2} p_k(q_i r_j + q_j r_i) + \text{two similar terms.} \tag{6.10}$$

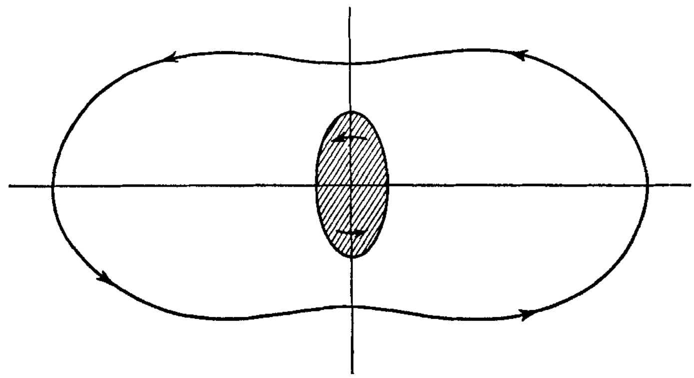

FIGURE 2. Far-field streamline in a principal plane of an ellipsoid forced to rotate in fluid at rest at infinity. The angular velocity is normal to the plane of the figure. The equation for the streamlines in the far field is

$$\ln \sigma = \text{const.} - \frac{3}{4}\frac{a^2-b^2}{a^2+b^2}\cos 2\phi,$$

where σ, ϕ are polar co-ordinates in the plane and $\phi = 0$ in the direction of the principal diameter $2a$; and all far-field streamlines are geometrically similar to the one shown.

It is now not difficult to appreciate that the contribution to the symmetrical part of the particle stress due to the imposed couple is associated with the radial motion of the fluid, at some distance from the particle, caused by the rotation of a non-spherical particle. The disturbance velocity far from an ellipsoid on which a couple N parallel to the third principal axis acts, in the absence of a bulk deformation, has a radial component

$$-\frac{3}{2}\left(\frac{1}{8\pi\mu} C_{ijk} L_k\right)\frac{x_i x_j}{r^4}, \qquad = \frac{3N}{8\pi\mu}\frac{a^2-b^2}{a^2+b^2}\frac{\bar{x}\bar{y}}{r^4},$$

where $\bar{x}$, $\bar{y}$ are components of $\mathbf{x}$ in the directions of the first two principal diameters. There is outflow in the first and third quadrants of the $(\bar{x}, \bar{y})$-plane, when $a > b$, and inflow in the second and fourth quadrants in the lee of the ends of the ellipsoid. The point is also made by figure 2, which shows the form of the streamlines, far from a rotating ellipsoid, in one of the principal planes. The circumferential component represented by these streamlines is responsible for the antisymmetrical part of the particle stress, and the radial component generates a contribution to the symmetrical part of the particle stress; and, contrary to what one might be inclined to suppose, the radial component of disturbance velocity approaches zero no more rapidly, as $r \to \infty$, than the circumferential component. The angular pattern of the symmetrical part of the particle stress may be visualized by noting that it is as in figure 1 (*a*), when the long diameter of the ellipsoid has the angular position shown in that figure.

In the case of a suspension of couple-free ellipsoids which are similar in shape and similarly oriented, C_{ijkl}/abc has the same value for all the particles; then, from (5.16) we have

$$\Sigma_{ij}^{(p)} = 3\mu e_{kl}\frac{C_{ijkl}}{abc}\frac{\Sigma\frac{4}{3}\pi abc}{V}, \tag{6.11}$$

in which C_{ijkl}/abc is given by (6.5) and $\Sigma\frac{4}{3}\pi abc/V$ is simply the concentration of particles by volume.

As a partial check on these formulae we may note that for the special case of a sphere, $a = b = c$, both $\Gamma_i^{(s)}$ and C_{ijk} vanish, as expected (a pure straining motion cannot make a sphere rotate, and a couple applied to a sphere cannot produce any radial motion far from the sphere), and (6.8) yields the known formula

$$L_i = 8\pi\mu a^3\,\Gamma_i^{(c)}. \tag{6.12}$$

Also (6.5) reduces to

$$C_{ijkl} = \tfrac{5}{3}a^3(\tfrac{1}{2}\delta_{ik}\delta_{jl}+\tfrac{1}{2}\delta_{il}\delta_{jk}-\tfrac{1}{3}\delta_{ij}\delta_{kl}), \tag{6.13}$$

which yields a Newtonian form of the bulk stress tensor with the Einstein value of the increase in the effective viscosity (viz. $\frac{5}{2}\mu$ by the volume concentration).

Jeffery (1922) showed that the action of a bulk pure straining motion without rotation is to turn a couple-free ellipsoidal particle so that its largest diameter is ultimately parallel to the greatest principal rate of extension and its smallest diameter is ultimately parallel to the least principal rate of extension. It is not difficult to see that, in a dilute suspension of either prolate or oblate spheroids which is subjected to an *axisymmetric* bulk pure straining motion, the bulk stress in that ultimate state has the same axial symmetry and so has the Newtonian form. Thus in this particular case it is possible to describe the particle stress in terms of an additional viscosity and to obtain its value from the change in the rate of dissipation due to the presence of the particles, as has been done by Takserman-Krozer & Ziabicki (1963).

Another circumstance in which the bulk stress has the Newtonian form occurs when couple-free particles are subjected to such strong Brownian motion that their orientations are randomly distributed with uniform probability. The suspension here has a microscopic structure which is statistically isotropic, and an average of the expression (6.5) for C_{ijkl} over all particle orientations yields a tensor form like (6.13), although with a different value of the effective viscosity. It may be shown with a little trouble that in this case

$$\Sigma_{ij}^{(p)} = 2\mu e_{ij}\frac{4\pi}{3V}\Sigma\, abc\left\{\frac{4(J_1+J_2+J_3)}{15(J_1J_2+J_2J_3+J_3J_1)}+\frac{2}{5}\left(\frac{1}{I_1}+\frac{1}{I_2}+\frac{1}{I_3}\right)\right\}, \tag{6.14}$$

the increase in the bulk viscosity due to the particles being the coefficient of $2e_{ij}$. So far as I know, this expression has not been given before, although it is latent in Jeffery's work. It could be used to investigate numerically the effect of particle shape on the effective viscosity, for this case of dominant Brownian motion; however, the results available for spheroids (for which the values of I_1, J_1, etc. can be calculated more readily) are probably adequate for this purpose.

Appendix. The place of surface tension in the analysis of bulk stress

In any consideration of the distribution of stress over a region which includes an interfacial surface, surface tension must be regarded as a singularity in the stress distribution with certain integral properties. A volume integral of the stress over such a region is then improper, strictly speaking, and must be interpreted appropriately. It is helpful for some purposes to regard the interface as a layer of small thickness ϵ, and to take the limit $\epsilon \to 0$ subsequently. The stress components for which both suffixes correspond to directions in the tangent plane of the interface are of large magnitude in the layer, and the correct integral properties may be obtained by supposing this singular part of the stress tensor in the layer to be of the form $T(\delta_{ij} - n_i n_j)/\epsilon$, where T is the coefficient of surface tension and $\mathbf{n}$ is the unit normal to the interfacial surface. The contribution to the volume integral $\int \sigma_{ij} dV$ from the portion of the interfacial surface A lying within the integration volume V is then

$$\int T(\delta_{ij} - n_i n_j)\, dA. \tag{A 1}$$

One-half of the integral (A 1) for a closed surface A_0 has been termed the surface-energy tensor, since its trace equals the surface energy TA_0 and it is found to play a role in 'virial theorems' which is analogous to that of other energy tensors such as $\int \frac{1}{2}\rho u_i u_j dV$ (Rosenkilde 1967).

Then, with V_0+ denoting the volume of a particle bounded by the closed surface A_0+ on the outer side of the interfacial layer, and V_0- and A_0- the corresponding quantities for the inner side of the same interfacial layer, we have

$$\left(\int_{V_0+} - \int_{V_0-}\right) \sigma_{ij} dV = \int_{A_0} T(\delta_{ij} - n_i n_j)\, dA. \tag{A 2}$$

Rosenkilde shows (by applying Stokes's theorem for a closed surface to the quantity $\epsilon_{kil} x_j n_l$, regarded as a vector with components given by the different values of k) that an alternative expression for the right-hand side is

$$\int_{A_0} T x_j n_i \operatorname{div} \mathbf{n}\, dA.$$

But since $\operatorname{div} \mathbf{n}$ is equal to the sum of the curvatures of any two orthogonal sections of the interfacial surface which contain the local normal $\mathbf{n}$, $Tn_i \operatorname{div} \mathbf{n}$ is simply the jump in normal stress across the interface due to surface tension. It follows that

$$\left(\int_{V_0+} - \int_{V_0-}\right) \sigma_{ij} dV = \left(\int_{A_0+} - \int_{A_0-}\right) \sigma_{ik} x_j n_k dA, \tag{A 3}$$

which confirms explicitly the statement in the text that the volume integral on the right-hand side of (4.3) has the same value for V_0+ and for V_0-.

It will be noted that in the case of a suspension of spherical fluid drops which is stationary everywhere, the bulk stress is unaffected by the existence of surface tension (the contribution to the volume average from the excess pressure within a drop being cancelled by the contribution from the interfacial layer) and is identical with the stress in the ambient fluid. In general, surface tension affects the bulk stress only through its influence on the shape and motion of the particle.

When an interface is deformed, work is done, and the volume integral used to evaluate the average rate of dissipation Φ (see (4.8)) contains a contribution from surface tension. The contribution to the integral $\int \frac{1}{2}\partial u_i/\partial x_j\,(\sigma_{ij}+\sigma_{ji})\,dV$ from the interfacial surface A_0 lying within the integration volume V is

$$\int_{A_0} T\frac{\partial u_i}{\partial x_j}(\delta_{ij}-n_i n_j)\,dA, \tag{A4}$$

which equals the rate of increase of surface energy TA_0 when T is uniform over A_0. But the deformation of a surface element is a reversible process, and changes in the surface energy should not be counted as 'dissipation'. It is therefore necessary to modify the relations (4.8) and (4.9) by replacing the average rate of dissipation Φ by $\Phi + d\Psi/dt$, where

$$\Psi = (1/V)\,\Sigma\, TA_0$$

is the surface energy per unit volume of the suspension. In the case of couple-free particles and negligible inertia forces, (4.9) then reduces to

$$\frac{\partial U_i}{\partial x_j}\Sigma_{ij} = \Phi + \frac{d\Psi}{dt}, \tag{A5}$$

which provides an indication that when the surface energy is changing the bulk stress has some features characteristic of an elastic medium.

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