The stress generated in a non-dilute suspension of elongated particles by pure straining motion

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In a pure straining motion, elongated rigid particles in suspension are aligned parallel to the direction of the greatest principal rate of extension, provided the effect of Brownian motion is weak. If the suspension is dilute, in the sense that the particles are hydrodynamically independent, each particle of length 2l makes a contribution to the bulk deviatoric stress which is of roughly the same order of magnitude as that due to a rigid sphere of radius l. The fractional increase in the bulk stress due to the presence of the particles is thus equal to the concentration by volume multiplied by a factor of order l^2/b^2 , where 2b is a measure of the linear dimensions of the particle cross-section. This suggests that the stress due to the particles might be relatively large, for volume fractions which are still small, with interesting implications for the behaviour of polymer solutions. However, dilute-suspension theory is not applicable in these circumstances, and so an investigation is made of the effect of interactions between particles. It is assumed that, when the average lateral spacing of particles (h) satisfies the conditions $b \ll h \ll l$, the disturbance velocity vector is parallel to the particles and varies only in the cross-sectional plane. The velocity near a particle is found to have the same functional form as for an isolated particle, and the modification to the outer flow field for one particle is determined by replacing the randomly placed neighbouring particles by an equivalent cylindrical boundary. The resulting expression for the contribution to the bulk stress due to the particles differs from that for a dilute suspension only in a minor way, viz. by the replacement of $\log 2l/b$ by $\log h/b$, and the above suggestion is confirmed. The relative error in the expression for the stress is expected to be of order $(\log h/b)^{-1}$. Some recent observations by Weinberger of the stress in a suspension of glass-fibre particles for which 2l/h = 7.4 and h/2b = 7.8 do show a particle stress which is much larger than the ambient-fluid stress, although the theoretical formula is not accurate under these conditions.

1. Introduction

The bulk properties of suspensions of elongated particles are of direct interest, since some natural particles, such as paper-pulp fibres, are of this form. There is also reason to believe that in certain circumstances the flexible macromolecules of polymeric materials are greatly drawn out in one direction and act hydrodynamically as elongated particles. Analytically, elongated particles are of course convenient for study, since the extensive results of slender-body theory for Stokes flow (Burgers 1938; Tuck 1964; Tillet 1970; Cox 1970; Batchelor 1970b) may be called on, and exact results are available for the particular case of an ellipsoid with one relatively large principal diameter. Observations of the flow properties of synthetic suspensions in the laboratory mostly refer to spherical particles, but elongated cylindrical or rod-like particles seem to be next in order of simplicity and some observations for suspensions of such particles are now available (Goldsmith & Mason 1967; Weinberger 1970).

A suspension of straight elongated rigid particles has the striking property that, when subjected to a steady pure straining motion, all the particles take up the orientation in which they individually make their greatest contribution to the bulk stress. It may be shown, moreover, that the contribution to the bulk stress due to the presence of parallel rigid particles increases rapidly with the length-to-breadth ratio, for a given volume fraction, in the case of a dilute suspension for which the particles are hydrodynamically independent. However, the range of values of the volume fraction for which a suspension is 'dilute' in this sense decreases as the particle length-to-breadth ratio increases, and the particle stress in a dilute suspension is never more than a perturbation of the stress due to the ambient fluid alone. The formula for the bulk stress in a dilute suspension is suggestive about what happens when the concentration is too large for particles to be independent, but one cannot say more than that.

It has been known for some time that various polymer solutions may exhibit a very much larger apparent viscosity in a pure straining motion than in a simple shearing motion, and experimental data concerning the stress levels are beginning to be available (Metzner & Metzner 1970). It is also evident that a long flexible chain-like macromolecule will be extended in one direction by the action of a pure straining motion of the solvent, to an extent which depends on the relative strengths of the frictional pull of the ambient fluid and the tendency due to Brownian motion for one part of the macromolecule to wander randomly relative to any other part. Calculations of the steady statistical configuration of an isolated macromolecule in a steady axisymmetric pure straining motion have been made on the basis of various assumptions about the mechanical and hydrodynamic properties of a macromolecule, from which one can make estimates of the bulk stress in a solution of non-interacting macromolecules (Takserman-Krozer 1963; Peterlin 1966). Both the experiments and the calculations indicate stress components which increase with the rate of strain more rapidly than linearly. However, the available theory suffers from the shortcoming that only isolated macromolecules are considered, whereas the large stress levels observed in polymer solutions in straining motions undoubtedly refer to macromolecules in an extended form in which the hydrodynamic interactions between particles are significant.

The primary purpose of this paper is to consider the effect of hydrodynamic interaction of parallel elongated particles in a pure straining motion on the bulk stress. We consider only the case of effectively rigid particles on which no external force or couple acts. All effects of Brownian motion are neglected. Application of the results to polymer solutions would involve the further problem of incorporating the effect of the hydrodynamic interaction between particles into calculations of the statistical shape of a macromolecule.

2. The stress in a suspension of elongated particles

We consider a suspension of rigid particles in an ambient Newtonian fluid characterized by viscosity μ . Each particle is free to translate and rotate with the surrounding fluid, and will be assumed to be so small that the effect of inertia forces in the relative motion near it is negligible.

In these circumstances, the contribution to the bulk or average stress due to the presence of the particles is given without further approximation by

$$\Sigma_{ij}^{(p)} = \frac{1}{V} \sum \int_{\mathcal{A}_0} \sigma_{ik} x_j n_k dA, \qquad (2.1)$$

where the integral is taken over the surface A_0 of a particle and the summation is over the many particles in a volume V in which conditions are statistically homogeneous (see Batchelor 1970*a*); $\sigma_{ik}n_k$ is the force per unit area exerted on the particle surface by the ambient fluid, at a point where the unit normal is **n**, and is to be determined as a property of the relative motion of the ambient fluid near the particle.

In the case of an elongated particle of length 2l whose surface is at a distance of order b from the straight line through the two ends of the particle (the particle 'axis'), where $b \ll l$, we have

$$\int_{A_0} \sigma_{ik} x_j n_k dA = -p_j l^2 \int_{-1}^1 F_i s \, ds + O(b/l), \tag{2.2}$$

where sl denotes distance along the particle axis from its centre, **p** is a unit vector parallel to this axis, and **F** is the force per unit length exerted by the particle on the ambient fluid at station s. The component of **F** normal to the particle length makes zero contribution to the integral for a particle on which no external force or couple acts, and so we have the approximate relation

$$\Sigma_{ij}^{(p)} = -\frac{1}{V} \sum p_i p_j l^2 \int_{-1}^{1} \mathbf{p} \cdot \mathbf{F} s \, ds \tag{2.3}$$

for a suspension of elongated particles.

In general \mathbf{p} will be different for different particles, but in some circumstances the effect of the bulk motion is to cause each particle to take up the same preferred orientation. In that event, (2.3) becomes

$$\Sigma_{ij}^{(p)} = -\frac{p_i p_j}{V} \Sigma l^2 \int_{-1}^{1} \mathbf{p} \cdot \mathbf{F} s \, ds, \qquad (2.4)$$

representing a particle stress system which is symmetrical about the common direction \mathbf{p} of the particle axes.

Only the deviatoric part of the expression (2.3) or (2.4) for the particle stress tensor is significant (since the isotropic part contributes to the bulk pressure which, in an incompressible fluid, is not dependent only on local quantities and must be determined from field equations) and the factor $p_i p_j$ may be imagined to be replaced by $p_i p_j - \frac{1}{3}\delta_{ij}$ here and in later formulae.

The complete expression for the bulk stress in a suspension of parallel particles in a bulk pure straining motion represented by e_{ij} is then

$$\Sigma_{ij} = -P\delta_{ij} + 2\mu e_{ij} + (\frac{1}{3}\delta_{ij} - p_i p_j) \frac{1}{V} \sum l^2 \int_{-1}^{1} \mathbf{p} \cdot \mathbf{F} s \, ds, \qquad (2.5)$$

where $P(=-\frac{1}{3}\Sigma_{ii})$ is the bulk pressure. We shall need later to compare the magnitudes of the contributions to the bulk deviatoric stress due to the presence of the particles and due to the pure ambient fluid. These two contributions to the bulk stress are not of exactly the same tensorial form unless the bulk straining motion is symmetrical about the direction **p**, so that a straight comparison of their magnitudes is not strictly possible in general. However, the two stress systems differ in a known and simple way, and there is no ambiguity in a comparison of the two contributions to the quantity

$$\Sigma_{11} - \frac{1}{2}(\Sigma_{22} + \Sigma_{33}), \quad = \frac{3}{2}(\Sigma_{11} + P), \tag{2.6}$$

where, for simplicity of symbolism, we have (temporarily) chosen the x_1 -axis to be in the direction **p**. This quantity is determined wholly by the deviatoric part of the bulk stress and is equal to $3\mu e_{11}$ for a Newtonian fluid.[†] The ratio of the contribution due to particles all parallel to the x_1 -axis to that for the pure ambient fluid is

$$\frac{\Sigma_{11}^{(p)} - \frac{1}{2} (\Sigma_{22}^{(p)} + \Sigma_{33}^{(p)})}{3\mu e_{11}} = -\frac{1}{3\mu e_{11} V} \Sigma l^2 \int_{-1}^{1} F_1 s \, ds.$$
(2.7)

We shall be concerned in particular to ask whether this ratio can be much larger than unity.

All these formulae hold for arbitrary values of the volume concentration of the particles. In the next section the special forms which apply in the absence of particle interactions will be described, as a preliminary to consideration of interaction effects.

3. Formulae for a dilute suspension

When the particles are sufficiently far apart from each other, the relative motion of the fluid near one particle is unaffected by the presence of the others. We adopt this state of hydrodynamic independence of the particles as the defining property of a 'dilute' suspension.

† In the case of a circular cylindrical column of Newtonian liquid at whose surface the normal stress is zero everywhere and which is being extended uniformly in the axial direction at the rate e_{11} , we have $\Sigma_{22} = -P + 2\mu e_{22} = 0$, giving $P = 2\mu e_{22} = -\mu e_{11}$ and showing that the ratio of axial tension to axial rate of extension is $\Sigma_{11}/e_{11} = 3\mu$. The quantity 3μ is known as the 'Trouton viscosity' (after Trouton (1906), who made measurements of this ratio for highly viscous materials such as wax and pitch). However, now that the tensor representation of stress and rate of strain is customary, it is preferable to avoid the use of a special name for a quantity which is only a multiple of the shear viscosity.

In a dilute suspension each particle is effectively immersed in fluid whose velocity gradient tends to a given constant value at infinity. The elongated rigid particle translates and rotates with the fluid, but it cannot take up the imposed straining motion. To a good approximation for a slender particle, the force component $\mathbf{p} \cdot \mathbf{F}$ exerted by unit length of the particle is determined wholly by, and is proportional to (in view of the linearity of the governing equations), the imposed rate of extension in the instantaneous direction of the particle length. We may therefore write

$$\mathbf{p} \cdot \mathbf{F} = -\mu p_k p_l e_{kl} l G(s), \tag{3.1}$$

where e_{kl} is the rate-of-strain tensor for the motion at infinity, and G(s) is a non-dimensional function of distance along the particle axis. Then (2.3) becomes

$$\Sigma_{ij}^{(p)} = \frac{\mu e_{kl}}{V} \sum p_i p_j p_k p_l l^3 \int_{-1}^{1} G(s) \, s \, ds.$$
(3.2)

This relation is in a form which displays a strong dependence on the particle length, but of course l has been used formally as a non-dimensionalizing factor and it remains to be seen how G depends on the thickness ratio b/l.

The function G(s) may be determined approximately by the methods of slender-body theory for Stokes flow, and results are available for particles of different cross-sectional shape and different kinds of variation of the cross-section along the particle length (Batchelor 1907b). In the crudest approximation, for which the relative error is of order $(\log l/b)^{-1}$, the particle shape is irrelevant and

$$G(s) = \frac{2\pi s}{\log\left(l/b\right)},\tag{3.3}$$

the definition of the constant b here being arbitrary to the extent of a factor of order unity. The influence of particle shape enters in the next approximation, which may be written as

$$G(s) = 2\pi s \epsilon \frac{1 - \epsilon \log \frac{(1 - s^2)^{\frac{1}{2}}}{R_s/R_0}}{1 - \epsilon (K + \frac{3}{2})},$$
(3.4)

where $\epsilon = (\log 2l/R_0)^{-1}$, $2\pi R_s$ is the perimeter of the particle cross-section at station s, $R_s = R_0$ at the central section s = 0, and K is a cross-sectional shape parameter (K = 0 for a circle) which may vary with s. The absolute error for this approximation is of order ϵ^3 in general, and a procedure for working out better approximations is available. In the case of a particle for which $(1-s^2)^{\frac{1}{2}}/R_s$ and K are both independent of s, the error in (3.4) is smaller than any power of ϵ .

Evaluation of the integral in (3.2) with any but the crudest approximation for G requires a knowledge of the particle shape. We may write the expression for the particle stress in general as

$$\Sigma_{ij}^{(p)} = \frac{4\pi}{3V} \mu e_{kl} \sum p_i p_j p_k p_l l^3 e Q(\epsilon), \qquad (3.5)$$
$$Q(\epsilon) = \frac{3}{4\pi\epsilon} \int_{-1}^{1} G(s) s \, ds$$

where

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is a shape factor which is unity with an error of order ϵ for any particle. One useful particular result is that

$$eQ(e) = \frac{1}{\log \frac{4l}{b_0 + c_0} - \frac{3}{2}}$$
(3.6)

for an ellipsoidal particle with semi-diameters l, b_0, c_0 , the error in this expression being smaller than any power of ϵ . And in the case of a cylindrical particle, for which the expression for $G(\epsilon)$ can be taken to the term of order ϵ^3 without difficulty (Batchelor 1970*b*),

$$Q(\epsilon) = \frac{1 + 0.640\epsilon}{1 - \epsilon(K + \frac{3}{2})} + \epsilon^2 \{ 0.699 + 0.640(K + \frac{3}{2}) \} + O(\epsilon^3).$$
(3.7)

The orientation of a particle in a dilute suspension is determined by the velocity gradient tensor describing the bulk flow and also by the effect of Brownian motion. In the case of a bulk simple shear flow, it is known from the work of Jeffery (1922) that in the absence of Brownian motion an elongated rigid spheroid executes a periodic orbit which is a member of a one-parameter family, and that for all these orbits the particle spends most of the orbit time near to the orientation in which **p** is normal to the direction in which the bulk velocity varies. This is an orientation in which the particle makes minimum disturbance of the fluid in its neighbourhood, and a time-average of the contribution to the bulk stress due to this particle (which is equivalent to an average over a number of identical particles with random phase in the same orbit) is of smaller magnitude than would be inferred from the appearance of the factor l^3 in (3.5).

The case of a bulk pure straining motion (or an 'extensional flow', as it is often termed in the literature on polymer solutions) is considerably simpler. Here the effect of the bulk motion on any isolated straight elongated rigid particle (such as the long ellipsoid considered by Jeffery (1922)) is to make its direction approach that of the greatest of the principal rates of extension. (The case in which two principal rates of extension are positive and equal is excluded.) On the other hand the effect of the rotational diffusion due to Brownian motion couples acting on particles is to spread the particle directions about the preferred orientation. The steady distribution of particle directions resulting from the balance between these two effects in a dilute suspension of rigid spheroids has been calculated by Takserman-Krozer & Ziabicki (1963) for various values of the ratio of e_{11} to the rotational diffusivity. The rotational diffusivity for a slender rigid particle is proportional to l^{-2} approximately, and so is small compared with e_{11} for sufficiently long particles. In order to bring out more clearly the effect of particle alignment here, we shall ignore all effects of Brownian motion in the explicit calculations while acknowledging their relevance to the behaviour of macromolecules. Then provided that the bulk pure straining motion remains steady for a time of order e_{11}^{-1} , where e_{11} is the greatest principal rate of extension and occurs in the direction of the x_1 -axis, all the elongated particles in a dilute suspension become approximately parallel to the x_1 -axis, in which case (3.5) gives

$$\Sigma_{ij}^{(p)} = \delta_{i1}\delta_{j1}\mu e_{11}\frac{4\pi}{3V}\sum \frac{l^3Q(\epsilon)}{\log 2l/R_0}.$$
(3.8)

The factor eQ(e) in (3.8) is a slowly varying function of l/R_0 , and as a rough approximation the contribution of each particle to the bulk stress is proportional to l^3 . It is common practice in the literature of suspension mechanics to specify the total number of similarly shaped particles in terms of their concentration by volume. However, this is not an appropriate choice for elongated particles since the particle stress depends only very weakly on the dimensions of the cross-section of the particle. (And the particle stress is determined more by the perimeter of the cross-section than by its area, as noted in an earlier paper (Batchelor 1970b).) The particle length has the dominant influence, and it enters the formula (3.8) approximately in the form of the volume of the (smallest) sphere that circumscribes the particle. Thus we could usefully write the expression for the particle stress in a dilute suspension of parallel elongated particles in a pure straining motion as

$$\Sigma_{ij}^{(p)} = \delta_{i1}\delta_{j1}\mu e_{11}\alpha, \qquad (3.9)$$

where

$$\alpha = \frac{1}{V} \sum_{3}^{4} \pi l^3 \epsilon Q(\epsilon) \tag{3.10}$$

is the 'volume' fraction of the particles regarded as spheres of radius $l(eQ)^{\frac{1}{2}}$.

It appears that in the case of a dilute suspension of aligned elongated particles the ratio (2.7) representing the relative magnitudes of the contributions to the bulk stress due to the particles and due to the pure ambient fluid is simply $\frac{1}{3}\alpha$. It is possible to find values of l and R_0 and of the number density of the particles such that $\frac{1}{3}\alpha$ takes values much larger than unity even though the true volume fraction of the particles is small. This makes one wonder whether dramatically large magnitudes of the bulk stress might be generated in a pure straining motion by elongated particles which occupy only a small fraction of the total volume. An affirmative answer is being suggested in this paper, although not simply on the basis of formulae for a dilute suspension. These formulae hold only when there is no hydrodynamic interaction between particles, and we shall now show that the condition for validity of the formulae is violated unless the value of $\frac{1}{3}\alpha$ is *small* compared with unity.

A slender rigid body immersed in a pure straining motion acts as a force doublet (of strength λ , say) so far as the disturbance motion at large distances is concerned, and the disturbance velocity is of magnitude $\lambda/\mu r^2$ at a large distance r from the particle. The rate of strain associated with the disturbance motion due to a particle is thus asymptotically of magnitude $\lambda/\mu r^3$, and the condition that a second particle in this position will not be affected hydrodynamically by the presence of the first particle is

$$|\lambda|/\mu r^3 \ll e_{11},$$

where e_{11} is a measure of the imposed bulk rate of strain. The number density n of particles whose centres are separated from each other by an average distance r is of order r^{-3} , so the condition for hydrodynamic independence of identical particles may be written as

$$n|\lambda| \ll \mu e_{11}$$

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But the doublet strength for a particle parallel to the x_1 -axis is

$$\lambda = l^2 \int_{-1}^{1} F_1(s) \, s \, ds = -\frac{4}{3} \pi \mu e_{11} l^3 \epsilon Q(\epsilon),$$

and so the condition becomes effectively

$$\alpha \ll 1$$
, or $nl^3 \epsilon \ll 1$, (3.11)

since the shape factor Q is of order unity. The volume of a rigid sphere which in a pure straining motion gives rise to a distant disturbance velocity of the same magnitude as that due to an elongated particle of length 2l is evidently of order $l^{3}c$, and the condition for the suspension to be 'dilute' is that the volume fraction of these equivalent spheres must be small compared with unity.

The conclusion is that dilute-suspension theory cannot predict particle stresses which are more than a perturbation of the stress due to the ambient fluid alone.

4. The velocity distribution in a suspension of close parallel particles subjected to pure straining motion

Limited though the range of applicability of the above formulae for a dilute suspension may be, it is clear first that a particle which is elongated makes a much larger contribution to the particle stress than one of the same volume and nearly spherical shape, and second that the contributions from different elongated particles are of the same sign when all the particles have the same orientation. These conclusions are likely to remain valid when the particles are not so far apart as to be hydrodynamically independent, although the magnitude of the particle stress will no doubt be affected by the interactions. We need new analysis which will take account of the interactions between particles and which will confirm the expectation of a potentially large particle stress and put it in quantitative form.

We suppose the suspension to be subjected to a bulk pure straining motion which is approximately steady for a time at least as large as the reciprocal of the strain rate. Brownian motion effects will again be ignored. In these circumstances an isolated elongated particle tends to become parallel to the direction of the greatest principal rate of extension, and it is a plausible assumption that each particle takes up this same orientation approximately even when neighbouring particles are not hydrodynamically independent. We consider the suspension in this state in which all the particles are approximately parallel to the x-axis, their positions being random. Now if the number of particles per unit volume is n, and if they all have about the same length 2l, the average number of particles which intersect unit area of any plane normal to the x-axis is 2nl. The average distance between neighbouring intersections is thus a length of order

$$(2nl)^{-\frac{1}{2}}, = h$$
 say. (4.1)

The ratio l/h determines the importance of interactions between the particles, and the case of a 'dilute' suspension corresponds to $l/h \ll 1$ (or, more precisely, as may be seen from (3.11), to $lc^{\frac{1}{2}}/h \ll 1$).

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Analysis which applies for an arbitrary value of l/h appears to be difficult, but there are some simplifying features in the case

$$l \gg h \gg b, \tag{4.2}$$

where 2b is, as before, a measure of the diameter of a particle cross-section. In this case the ambient fluid is moving in the space between parallel needle-like particles whose lateral spacing is small compared with their length, and we may suppose (1) that the difference between the local velocity and the bulk or average velocity is a vector approximately in the x-direction, and (2) that spatial gradients of the fluid velocity in the x-direction are small relative to those in the (y, z)plane. Inertia forces are negligible in the relative motion between adjoining particles, and it follows that the pressure gradient $\partial p/\partial x$ must be approximately constant, and equal to zero in view of the (statistical) homogeneity of conditions in the x-direction. The equation for the x-component of velocity in the ambient fluid is then approximately

$$\frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2} = 0. \tag{4.3}$$

It is known that this is the governing equation in the region within a distance from an elongated particle which is small compared with l (except of course near blunt ends of a particle) when the particle is isolated in a pure straining motion (Batchelor 1970*b*), and the new result is that, since there is no point in the fluid which is *not* within a distance from a particle which is small compared with lwhen the condition $l \ge h$ is satisfied, equation (4.3) holds everywhere in the ambient fluid under this condition.

The boundary conditions to be satisfied by u, regarded as a function of y and z, are the no-slip conditions at the surfaces of each of the particles intersected by a plane normal to the x-axis. We shall suppose that the velocity of each particle is equal to the value of the bulk velocity at the position of the centre of the particle. Then if we choose the axes of reference so that the bulk velocity is zero at the origin, the distribution of u in the (y, z)-plane through the origin satisfies the conditions

$$u = e_{11} x_c^{(m)}$$

on the curve A_m in which the surface of the *m*th particle, whose centre is at $x = x_c^{(m)}$, intersects the plane x = 0; e_{11} is the bulk rate of extension in the direction of the particle lengths as before. $x_c^{(m)}$ is a random quantity with zero mean which takes values between -l and +l with uniform probability, and the position of the cross-section of particle *m* in the plane x = 0 is also a random quantity. Figure 1 illustrates the problem for identical particles whose cross-sections are circular. One might think of the dependent variable *u* as being represented as the normal distance from the plane of figure 1 (*b*), giving a surface which rises or falls to given positions at the curves A_m ; and it is a consequence of equation (4.3) that this surface has the same shape as a soap film stretched between the particles.

Near any one particle, the shape of the particle cross-section will affect the distribution of u. But the velocity distribution becomes circularly symmetric at a distance of many diameters from the particle surface, irrespective of the

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cross-sectional shape, and u there has the same value as if the particle crosssection were circular with a certain equivalent radius and the same force per unit length of the particle were exerted on the fluid (Batchelor 1970b). We can write this equivalent radius as kR_s , where $2\pi R_s$ is the actual perimeter of the local cross-section and the shape parameter k is related to the quantity K in (3.4) by $K = \log k$. The value of k is known for several specific shapes of the crosssection of a particle which is locally approximately cylindrical, and lies between 0.785 and 1.0 for simple shapes which are convex everywhere. This allows us to pose the above problem in terms of particles of circular cross-section, of radius b



FIGURE 1. Sketch showing parallel elongated particles of length 2l and breadth 2b in a pure straining motion, with the greatest principal rate of strain in the x-direction. The average lateral spacing h satisfies the condition $l \ge h \ge b$. Figure 1(b) shows the intersections of particles with the transverse plane normal to the particle lengths at x = 0.

say, on the understanding that the radius has the appropriate effective value when the real particle cross-section is not circular.

The mathematical problem is then to find a solution of Laplace's equation which satisfies the conditions

$$u = e_{11} x_c^{(m)}$$
 at $|\boldsymbol{\sigma} - \boldsymbol{\sigma}^{(m)}| = b^{(m)}$, (4.4)

where σ is the two-dimensional vector with components (y, z), $\sigma^{(m)}$ specifies the position of the centre of the (effective) circle in which the *m*th particle intersects the plane x = 0 and $b^{(m)}$ is its radius. Near the intersection with *m*th particle the solution is of the form

$$u = e_{11} x_c^{(m)} \frac{\log \{ |\boldsymbol{\sigma} - \boldsymbol{\sigma}^{(m)}| / d^{(m)} \}}{\log b^{(m)} / d^{(m)}},$$
(4.5)

where $d^{(m)}$ is an unknown length which may vary with m. The force per unit particle length that the mth particle exerts on the fluid (at x = 0), which is what we want to find, is then

$$F_1 = -2\pi\mu \frac{e_{11}x_c^{(m)}}{\log b^{(m)}/d^{(m)}}.$$
(4.6)

 $d^{(m)}$ and F_1 are determined by the way in which the 'inner' flow (4.5) joins the outer flow, at $|\boldsymbol{\sigma} - \boldsymbol{\sigma}^{(m)}| \ge b^{(m)}$, where the direct influence of neighbouring particles is important.

It is not likely that an explicit solution of equation (4.3) with the random boundary conditions (4.4) could be found. The best that I can think of doing is to exploit the two known primary features of the random boundary conditions, viz.

(a) the average of $e_{11}x_c^{(m)}$ over the different particles is zero, which implies that the average value of u over the plane x = 0 is zero, and

(b) the average distance between neighbouring particles is h. These facts show that u decreases from $e_{11}x_c^{(m)}$ at $|\boldsymbol{\sigma}-\boldsymbol{\sigma}^{(m)}| = b^{(m)}$ to values which are zero on average at distances $|\boldsymbol{\sigma}-\boldsymbol{\sigma}^{(m)}|$ which are of order h. Then if we ignore the existence of fluctuations about the average and simply require that u be zero at a distance h from the centre of the mth particle, we find

$$d^{(m)} = h. \tag{4.7}$$

The corresponding expression for the force per unit length exerted by a particle of local radius b, at a distance x from the particle centre, is

$$F_1 = -\frac{2\pi\mu e_{11}x}{\log h/b}.$$
 (4.8)

The solution (4.7) would be correct if the various particles surrounding the *m*th particle were replaced by a single circular boundary at $|\boldsymbol{\sigma} - \boldsymbol{\sigma}^{(m)}| = h$ where the velocity is zero. We are thus making a kind of 'cell model' of the flow, as has been done in the case of some other problems of interaction between particles with random positions (see Happel & Brenner 1965). Cell models do not always give accurate results, and the above argument cannot be said to be beyond question. However, it seems unlikely here that the average value of $d^{(m)}$ could differ from h by more than a factor of order unity, since $d^{(m)}$ is a parameter of the 'outer' flow field for the *m*th particle and the average spacing of the particles is the only length, or at any rate the only obvious length, occurring in the statistical specification of the geometrical arrangement of the particles.

The above result is given a little further support by an exact solution for just two particles, with circular cross-sections of radius b and centres on the y-axis distance H apart, and equal and opposite velocities in the x-direction ($\mp W$). The well-known solution of Laplace's equation that describes (among other physical applications) the stream function for two-dimensional irrotational flow due to two point vortices of equal and opposite strength in unbounded fluid is known to yield streamlines which are all circular, and on adapting this solution appropriately we obtain

$$u = \frac{1}{2}W\log\frac{(y+\frac{1}{2}H-\beta)^2+z^2}{(y-\frac{1}{2}H+\beta)^2+z^2} / \log\frac{H+b-\beta}{b+\beta},$$

$$2\beta = H - (H^2 - 4b^2)^{\frac{1}{2}}.$$
(4.9)

where

When $H/b \ge 1$, we have $\beta/b \ll 1$, and the solution near either particle boundary is seen to be of the form (4.5) with $d^{(m)} \approx H$; and when making a transition from two particles to a random array with uniform mean number density it is reasonable to regard the average spacing h as corresponding to the determinate spacing H of the two particles.

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This same solution (4.9) shows also that when two circular particles are nearly touching, corresponding to H/2b being near unity, the frictional forces per unit length exerted by the particles are approximately

$$\mp 24\pi\mu W \left(\frac{b}{H-2b}\right)^{\frac{1}{2}},\tag{4.10}$$

of which the denominator has small magnitude. Consequently, in an exact solution of (4.3) for a random distribution of particles over the whole plane x = 0 with the boundary condition (4.4), the force per unit length exerted by those particles that happen to be very close to a neighbour would be found to be much larger in magnitude than the expression (4.8). However, our theoretical model of the suspension is not realistic for nearly touching particles, because we have



FIGURE 2. Definition sketch for particles which are nearly touching and are separating with relative velocity 2W.

assumed each particle to be moving (in the x-direction) with a velocity equal to the bulk velocity at the position of its centre, whereas in reality the velocity of a particle is determined by the condition that no net force acts on it. If two circular cylinder particles each of length 2l are parallel, with distance H between their axes, and nearly touching over a portion $2(l-x_c)$ (corresponding to particle centres at $x = \mp x_c$, see figure 2), and are moving with velocities $\mp W$, each particle exerts on the other a retarding force

$$4\pi\mu(l-x_c)\;W\left(\frac{b}{H-2b}\right)^{\frac{1}{2}}$$

This force is balanced by the pull exerted on the free portion of a particle, which we may estimate as $f^{l+x_e} 2\pi \mu(e-x-W)$

$$W = \frac{\int_{l-x_c}^{l+x_c} \frac{2\pi\mu(e_{11}x - W)}{\log h/b} dx,}{\frac{le_{11}x_c(H-2b)^{\frac{1}{2}}}{b^{\frac{1}{2}}(l-x_c)\log h/b + x_c(H-2b)^{\frac{1}{2}}}}.$$
(4.11)

whence we obtain

This speed of separation of two very close particles is small compared with $e_{11}x_c$, and substitution of (4.11) in (4.10) shows that the force per unit length on the nearly-touching portion of each particle is now of the *same* order of magnitude as the expression (4.8). Hence there is no reason to think that the occurrence of a certain number of very close pairs of particles (or groups of more than two) will invalidate the estimate (4.8).

Finally, we note that the relative error in the expression (4.8) for F_1 may be

expected to be of order $(\log h/b)^{-1}$. The numerical accuracy of the theory is consequently very poor, and is comparable with that obtained from the first approximation to the particle stress in a dilute suspension in which all features of the particle shape other than its length to breadth ratio are irrelevant (Batchelor 1970*b*).

5. The bulk stress in a suspension of close particles

We may now substitute (4.8) in the relation (2.4) and evaluate the particle stress for a suspension being subjected to a bulk straining motion for which e_{11} is the greatest principal rate of strain and occurs in the direction of the x-axis. Strictly speaking the (effective) radius b of the particle cross-section is a function of position along the length of the particle; however, the approximation already made about the outer boundary condition implies that h is arbitrary to the extent of a factor or order unity, and we may therefore take b as a constant length and choose it for convenience to be R_0 , where $2\pi R_0$ is the perimeter of the central cross-section of the particle. We then find

$$\Sigma_{ij}^{(p)} = \delta_{j1} \delta_{j1} \mu e_{11} \frac{4\pi}{3V} \sum \frac{l^3}{\log h/R_0},$$
(5.1)

where $h = (2nl)^{-\frac{1}{2}}$.

This expression for the bulk stress is applicable when $l \ge h \ge R_0$, and it is a consequence of this condition that the volume fraction of the particles is small compared with unity, although not so small that the suspension is dilute in the sense of negligible particle interactions.

The similarity of form of this expression and the expression (3.8) found for a dilute suspension of parallel particles is striking. In particular, the same strong dependence on particle length is present in (3.8) and (5.1). If we regard the function $Q(\epsilon)$ in (3.8) as being unity, for the purpose of comparing (3.8) with a less accurate formula, the sole difference between the two expressions is that $\log 2l/R_0$ in (3.8) is replaced by $\log h/R_0$ in (5.1). This similarity of the two expressions for the particle stress results of course from the fact that the particle interactions considered in the previous section simply modify the 'outer flow' for one particle and lead to the outer boundary condition of zero disturbance at infinity in all three dimensions (for a dilute suspension) being replaced by zero velocity disturbance at the cylindrical boundary $(y^2 + z^2)^{\frac{1}{2}} = h$. We note that the two formulae would give the same values for the particle stress at $n = (2l)^{-3}$, when h/2l would be unity, which lies outside the range of validity of either formulae; interpolation between the two formulae in the range h/l = O(1) is thus possible.

It appears then that a pure straining motion can generate a bulk deviatoric stress which is of much larger magnitude than that for the pure ambient fluid, even when the volume fraction of the particles (c) is small compared with unity. We may see this explicitly by taking the case of a suspension of identical cylindrical particles of circular cross-section, for which

$$c = 2\pi R_0^2 ln$$
 and $\frac{h}{R_0} = \left(\frac{\pi}{c}\right)^{\frac{1}{2}}$.

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The conditions (4.2) for validity of the close-particles formula imply that

$$c^{\frac{1}{2}} \ll 1$$
 and $c^{\frac{1}{2}}l/R_0 \gg 1$.

Under these conditions the ratio of the contribution to $\Sigma_{11} - \frac{1}{2}(\Sigma_{22} + \Sigma_{33})$ due to the presence of the particles to that due to the pure ambient fluid is

$$\frac{\Sigma_{11}^{(p)} - \frac{1}{2} (\Sigma_{22}^{(p)} + \Sigma_{33}^{(p)})}{3\mu e_{11}} = \frac{4}{9} c \frac{l^2 / R_0^2}{\log \pi / c}.$$
(5.2)

This ratio exceeds 700 when c = 0.01 and $l/R_0 = 10^3$. In the case of particles whose cross-sections are flattened, the volume fraction needed to produce the same magnitude of particle stress is even smaller.

It is instructive to show on the one diagram how the deviatoric particle stress varies over the whole range of values of the number density (n) for a suspension of identical particles. In a dilute suspension the particle stress is proportional to n, and in the close-particles range there is only a weak departure from linearity. A convenient quantity to plot is therefore

$$\frac{\sum_{11}^{(p)} - \frac{1}{2}(\sum_{22}^{(p)} + \sum_{33}^{(p)})}{3\mu e_{11}nl^3},$$
(5.3)

which can be regarded as a non-dimensional form of (one component of the) deviatoric particle stress per particle in unit volume. In the close-particles range (5.1) shows that the expression (5.3) is equal to

$$\frac{4\pi/9}{\log h/R_0} \quad \text{or} \quad \frac{4\pi/9}{\log 2l/R_0 - \frac{1}{2}\log 8nl^3},\tag{5.4}$$

which is plotted in figure 3 for several values of l/R_0 , and in the range corresponding to a dilute suspension (3.8) shows that (5.3) reduces to

$$\frac{4\pi Q(\epsilon)}{9\log 2l/R_0}.$$
(5.5)

The first approximation to $Q(\epsilon)$ when $l/R_0 \ge 1$ is unity for particles of any shape, but the curves in figure 3 have been calculated from the expression

$$Q(\epsilon) = \{1 - \frac{3}{2}(\log 2l/R_0)^{-1}\}^{-1},$$

which is correct to the order of any power of ε for a spheroidal particle and is likely to be more accurate for a particle of unknown shape than the first approximation for Q.

Figure 3 makes it clear that the formula obtained for a dilute suspension continues to give reasonably accurate results when $8l^3n$ (or 2l/h) is of order unity, which is well above the values for which the particles are hydrodynamically independent. In other types of bulk flow, such as a simple shearing motion, the effect of hydrodynamic interaction of the particles might lead to a more radical change in the expression for the particle stress.

It is obviously possible to construct interpolation formulae which behave like

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(5.4) when $h \ll l$ and like (5.5) when $h \gg l$. One such formula for the deviatoric particle stress is

$$\frac{\Sigma_{11}^{(p)} - \frac{1}{2} (\Sigma_{22}^{(p)} + \Sigma_{33}^{(p)})}{3\mu e_{11} n l^3} = \frac{4\pi/9}{\log \frac{2l}{R_0} - \log \frac{h+2l}{h} - 1.5},$$
(5.6)

which reduces to the dilute-suspension formula for spheroidal particles when $h \ge l$ and has the same asymptotic form, as $h/l \to 0$ and $R_0/h \to 0$, as the close-particles formula.



FIGURE 3. A non-dimensional form of the deviatoric particle stress, per particle in unit volume, as a function of the number density of identical elongated particles, for a pure straining motion with e_{11} as the greatest principal rate of strain. The solid curves in the range $l \ll h$, for which the suspension is dilute, are those found theoretically for spheroidal particles. The solid curves in the range $l \gg h$ were obtained from the theoretical relation (5.1). The broken curves are sketched interpolations. The black circle denotes the experimental result obtained by Weinberger (1970) for a suspension of identical glass-fibre rods.

6. Comparison with observation by Weinberger

Observations of the bulk stress in a synthetic suspension of glass-fibre rods subjected to a pure straining motion have recently been made and described in a Ph.D. dissertation by Weinberger (1970), and so far as I know these are the only available measurements of this kind. The rods were uniformly of length 0.2 mmand of circular cross-section with diameter 0.0035 mm, and the volume fraction (in each of two liquids, 'Indopol' and 'Silicone') was 0.013; the corresponding length ratios are $l/R_0 = 57$, $h/2R_0 = 7.8$, 2l/h = 7.4. These numbers are not large enough for the close-particles formula to be expected to give accurate results, but a comparison with the theory nevertheless has some value.

The method of observation of the stress and the detailed results will be described in a paper and submitted for publication. Meanwhile Dr Weinberger has kindly allowed me to quote his result. His pure straining motion was axisymmetric, with a rate of extension e_{11} along the axis of symmetry, and he found that the observations of the deviatoric stress $\sum_{11} -\frac{1}{2}(\sum_{22} + \sum_{33})$ as a function of e_{11} could be fitted by a straight line. The coefficient in this linear relation was found to be equivalent to an increase in the viscosity of each of the two ambient liquids by a factor of about 9[†], that is to say, the particle deviatoric stress was about 8 times as large as the deviatoric stress for the pure ambient fluid. This observational result is shown in figure 3.

For circular-cylinder particles with c = 0.013 and $l/R_0 = 57$, the value of the ratio of the two contributions to the deviatoric stress obtained from the closeparticles formula (5.2) is 3.5.1 This is not close to the observed value 8, but better agreement cannot be expected since the experiments do not satisfy adequately the conditions assumed in the theory. As mentioned above, the relative error in the close-particles formula for the particle stress is likely to be of order $(\log h/R_0)^{-1}$, which for these experiments is 0.36, too large for an asymptotic formula to be accurate. If in the next approximation to $(5\cdot 1)$ the factor $\log h/R_0$ were replaced, for instance, by $\log (h/R_0) - 2 \cdot 14$, which is the modification to the factor $\log 2l/R_0$ found to be needed for a dilute suspension of circular cylinders, the theoretical prediction of the stress ratio for Weinberger's suspension would be 16 instead of 3.5. And if the interpolation formula (5.6) is used, the calculated stress ratio for Weinberger's suspension is 8.6, which is quite close to the observed value. Similar experiments with particles having much larger values of $h/2R_0$ and 2l/hare needed for a decisive check on the close-particles theory. Meanwhile the formula (5.6) appears to provide the best available basis for quantitative predictions.

The theory for closely spaced particles and Weinberger's observation both show that the magnitude of the bulk deviatoric stress in a suspension of elongated particles subjected to pure straining motion may be much larger than that for pure ambient fluid, despite the fact that the volume fraction of the particles is small compared with unity.

[†] This number is obtained from the straight lines drawn by Weinberger in his figures 4.28 and 4.29 to give the best visual fit with the data.

 \ddagger In his dissertation Weinberger says that my close-particles formula gives 2.19 for the suspension used in his experiment. He explains that he used the abbreviated version of the formula recorded in the summary of a lecture I gave at a symposium in March 1969 (see *J. Fluid Mech.* **39**, 1969, p. 397) and that he inserted numerical factors to make the formula consistent with that for a dilute suspension. These inserted numerical factors are not the correct ones (nor is there any need for consistency of the dilute-suspension and close-particles formulae since they apply to non-overlapping ranges of values of n).

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