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INSTABILITY OF SEDIMENTING BIDISPERSE SUSPENSIONS

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Abstract—When an initially homogeneous suspension consisting of two species of spherical particles (with differing size and/or density) suspended in a fluid (i.e. a bidisperse suspension) is allowed to sediment it is known that variations in the concentrations of the particles spontaneously occur due to some instability. Vertical columns containing one or the other species of particle are formed, with the resulting induced motion causing augmented particle sedimentation rates. A theory is developed here which explains the instability and column formation in terms of macroscopic equations derived by considering the effect of two-particle collisions which are assumed to involve some non-hydrodynamic process (such as physical contact) between the particles. Based on the derived macroscopic equations, conditions for instability of the suspension are obtained. These results are then compared with existing experiments and theory.

Key Words: suspensions, sedimentation, particles, viscous flow

1. INTRODUCTION

Whitmore (1955) noted that in a monodisperse suspension of sedimenting small solid spherical particles the settling speed would be *increased* by adding neutrally buoyant particles to the suspension. This observation was explained as being the consequence of structure formation in the suspension in which the sedimenting heavier particles would gather together to form vertical columns which, being more dense on the average than the surrounding suspension, would set up convection currents causing the heavier particles to settle more rapidly. This structure formation was further investigated by Weiland and co-workers (Weiland & McPherson 1979; Fessas & Weiland 1981, 1982, 1984; Weiland *et al.* 1984) for more general bidisperse suspensions of spheres (i.e. suspensions in a liquid of two species of spherical particles which differ from each other in either size and/or density). They found again structure formation in which each species of particle would gather together and form vertical columns. Experiments performed by Batchelor & Janse van Rensburg (1986) have indicated that initially the instability of the suspension seems to produce "blobs" of each species of sphere which grow in size and then under some circumstances (but not always) develop into vertical streaming columns.

In the present paper a theory is developed which gives a possible explanation for the initial instability and the column formation in a sedimenting bidisperse suspension. This is done by considering the horizontal displacement of particles due to two-particle collisions in which some non-hydrodynamic effect (such as particle–particle contact of the colliding particles) is taken into account (sections 3 and 4). By considering a situation in which the particle concentrations and also the velocity of the suspension (which is assumed to be vertical) vary in only one horizontal direction, the probability distribution of horizontal particle displacements is found in section 5. This is then used in section 6 to obtain equations for the macroscopic behaviour of the sedimenting bidisperse suspension. These results are further investigated and given physical interpretation in section 7. The conditions for instability of an initially homogeneous sedimenting bidisperse suspension are then obtained in section 8. The conclusion (section 9) gives a comparison between the present theory for bidisperse suspension instability (in which horizontal variations of particle concentration are considered) and the theory given by Batchelor & Janse van Rensburg (1986) (in which vertical variations of particle concentration are considered).

2. PROBLEM CONSIDERED

A bidisperse suspension consists of a Newtonian fluid of viscosity μ and density ρ in which two distinct species of monodisperse particles are suspended. These two species of particle will be labelled 1 and 2, with particle species 1 being uniform spheres of radius a_1 and density ρ_1 and particle species 2 being uniform spheres of radius a_2 and density ρ_2. Assuming that the sizes of the particles are sufficiently small that all relevant Reynolds numbers are so small that fluid inertia effects may be neglected and also that the particle sizes are sufficiently large for Brownian motion to be negligible, we obtain the downward sedimentation velocities V_1 and V_2 of isolated particles of species 1 and 2 as

$$V_1 = \frac{2(\rho_1 - \rho)ga_1^2}{9\mu} \quad \text{and} \quad V_2 = \frac{2(\rho_2 - \rho)ga_2^2}{9\mu} \tag{1}$$

by balancing the Stoke's drag with the gravity and buoyancy forces on a particle.

It will be assumed that the particle sizes and the mean distance between neighbouring particles are extremely small compared to some macroscopic experimental length scale L so that the suspension may be considered as a continuum with well-defined volume concentrations c_1 and c_2 of particles 1 and 2 which vary with position with this length scale L. Then c_1 and c_2 are related to the numbers n_1 and n_2 per unit volume of spheres 1 and 2 by

$$c_1 = \frac{4\pi a_1^3 n_1}{3} \quad \text{and} \quad c_2 = \frac{4\pi a_2^3 n_2}{3}, \tag{2}$$

so that the total number of particles N per unit volume is

$$N = \frac{3}{4\pi}\left(\frac{c_1}{a_1^3} + \frac{c_2}{a_2^3}\right). \tag{3}$$

The above conditions may therefore be written as

$$\frac{a_1}{L} \ll 1, \quad \frac{a_2}{L} \ll 1 \tag{4}$$

and

$$c_1\left(\frac{L}{a_1}\right)^3 + c_2\left(\frac{L}{a_2}\right)^3 \gg 1. \tag{5}$$

On the macroscopic length scale L the suspension may possess a macroscopic velocity $\mathbf{U}$ which may be a gravitationally driven flow resulting from mean density variations of the suspension on the macroscopic scale or may be externally imposed (by, for example, moving boundaries).

We avoid problems concerning the complex nature of particle motion in a concentrated suspension by assuming that the concentrations c_1 and c_2 are small, i.e.

$$c_1 \ll 1 \quad \text{and} \quad c_2 \ll 1. \tag{6}$$

Then the total solids concentration $c = c_1 + c_2$ is also very small.

We now define a set of dimensional cartesian axes x_1, x_2, x_3 with arbitrary origin and with the x_3-axis vertically downwards. In order to examine motion on the macroscopic scale it is convenient to define a macroscopic dimensionless position vector $\tilde{\mathbf{r}} = (\tilde{x}_1, \tilde{x}_2, \tilde{x}_3)$, where

$$\tilde{x}_1 = \frac{x_1}{L}, \quad \tilde{x}_2 = \frac{x_2}{L}, \quad \tilde{x}_3 = \frac{x_3}{L}. \tag{7}$$

By investigating particle motion on the microscopic scale we will derive equations which determine the macroscopic quantities c_1, c_2 and $\mathbf{U}$. Since the general three-dimensional problem is still complex (despite the above assumptions [4]–[6]), we assume that $\mathbf{U}$ is in the vertical direction and that c_1, c_2 and $\mathbf{U}$ vary only in the horizontal x_1-direction on the macroscopic length scale L. Thus we write

$$c_1 = c_1(\tilde{x}_1), \quad c_2 = c_2(\tilde{x}_1) \quad \text{and} \quad \mathbf{U}/U_0 = (0, 0, \tilde{U}(\tilde{x}_1)), \tag{8}$$

where U_0 is the characteristic magnitude of the velocity field $\mathbf{U}$. The quantity $\tilde{U}(\tilde{x}_1)$ is therefore dimensionless and of order unity in magnitude. The concentrations and flow given by [8] have approximately the form required to represent the vertical fingers observed by Weiland *et al.* (1984) in their experiments.

3. TWO-PARTICLE COLLISIONS

Since at low concentrations, the number of two-particle collisions occurring at any instant is very much larger than the number of collisions involving three particles (the former being proportional to the concentration and the latter to the square of the concentration), we expect the effect of two-particle collisions to dominate.

Consider the hydrodynamic interaction between a sphere of species 1 and a sphere of species 2 resulting from their sedimentation in an undisturbed fluid flow $\mathbf{U}$ of the form [8]. The fluid velocity $\mathbf{u}$ and dynamic pressure p in the neighbourhood of the spheres satisfy the creeping flow equations

$$\mu\nabla^2\mathbf{u} - \nabla p = \mathbf{0}, \quad \nabla\cdot\mathbf{u} = 0 \qquad [9]$$

with the no-slip boundary condition on the sphere surfaces.

The velocity and angular velocity of the spheres are then determined by the requirements that the hydrodynamic force on spheres 1 and 2 be $F_1 = (4\pi/3)(\rho_1 - \rho)ga_1^3$ and $F_2 = (4\pi/3)(\rho_2 - \rho)ga_2^3$, respectively, upwards and that the hydrodynamic torque on each sphere about its centre be zero. Suppose that at some instant during the interaction the spheres have the positions shown in figure 1a, the velocity of the centre of sphere 1 is (v_1, v_2, v_3) relative to a stationary observer. If all velocities are reversed, then by making use of the linearity of [9], it is seen that we have the situation shown in figure 1b. A reflection of the system in the x_1x_2-plane then yields the situation of figure 1c. Thus, a change in sphere configuration from that in figure 1a to that in figure 1c causes the velocity of the centre of sphere 1 to change from (v_1, v_2, v_3) to $(-v_1, -v_2, v_3)$. It is therefore seen that the orbit of the centre of sphere 1 relative to a stationary observer is its own mirror image about the x_1x_2-plane at the level where the line joining the sphere centres becomes horizontal during the interaction (see figure 2). It therefore follows that the x_1 and x_2 coordinates of the centre of sphere 1 after the interaction are identical to their values before the interaction. In other words, sphere 1 is not displaced horizontally by its interaction with sphere 2. Similarly, sphere 2 is also not displaced horizontally by the interaction (see figure 2). In a similar manner it is seen that spheres are not displaced horizontally during other types of two-particle collisions (between sphere 1 and sphere 1 or sphere 2 and sphere 2). Thus, it follows that there can be no horizontal flux of spheres 1 or of spheres 2 in a suspension in which interactions between the spheres are purely hydrodynamic and the effects of only two-particle collisions are taken into account. Since the phenomenon observed by Weiland *et al.* (1984) involves horizontal fluxes of the two species of spheres, it cannot be explained by purely hydrodynamic two-particle interactions. The observed phenomenon must therefore result from either

(a) interactions between three or more particles

or

(b) interactions between two spheres involving an effect other than the purely hydrodynamic inertialess flow considered above.

While individual spheres are not displaced horizontally in purely hydrodynamic two-sphere interactions, it is probable that they are in purely hydrodynamic three-sphere interactions [see (a) above] due to the lack of symmetry for such collisions. However, it is not clear whether this will (or under what circumstances this will) cause horizontal fluxes of each of the species of spheres. What is certain is that if all sphere interactions (whether between two, three or more spheres) are purely hydrodynamic, then the behaviour of the suspension is reversible in the sense that if after a certain period of time, during which the particles move under the influence of gravity

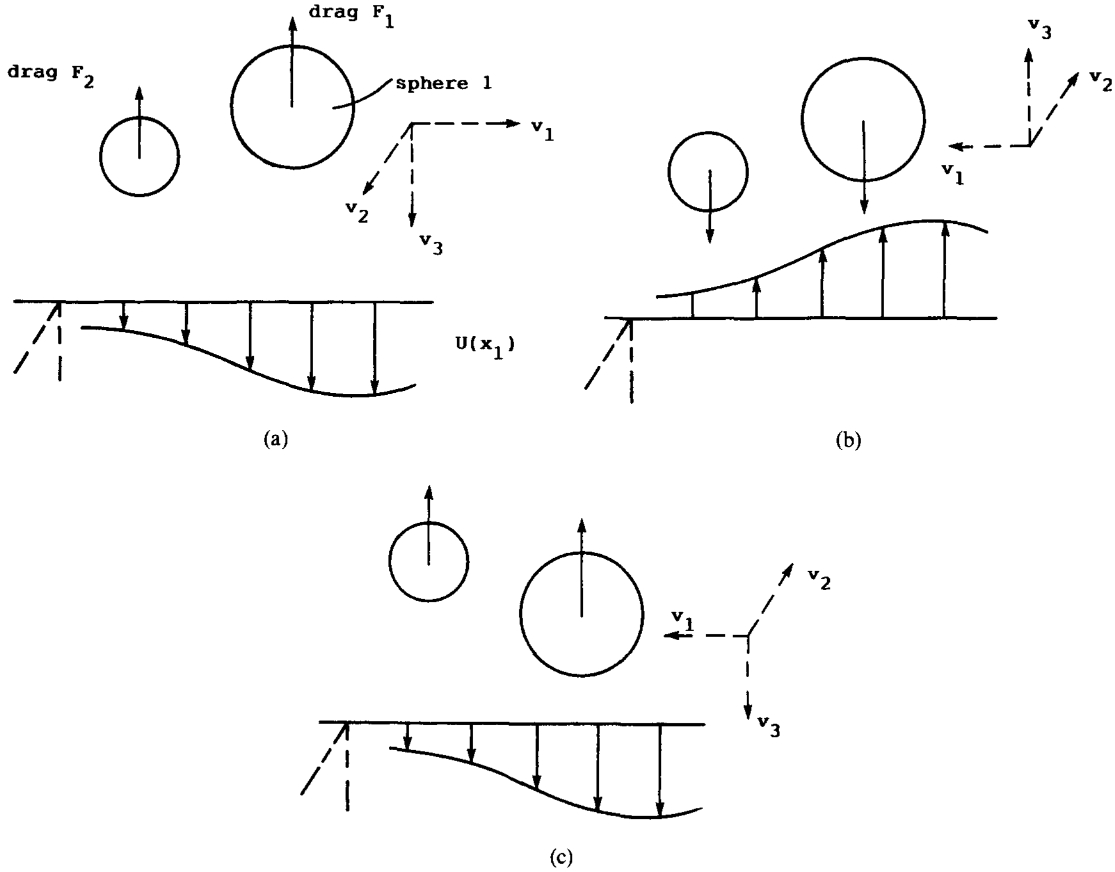


Figure 1. (a) Due to gravity forces on the spheres and to the flow field $U(x_1)$ the velocity of sphere 1 is (v_1, v_2, v_3). (b) Situation obtained from figure 1a by reversing all velocities (and stresses). (c) Situation obtained from figure 1b by reflection in x_1x_2-plane.

and superimposed flow $U(x_1)$, the direction of gravity and of $U(x_1)$ are reversed, then each individual particle in the suspension will retrace its original motion in the reverse direction. That this is so is due to the linearity of [9] and boundary conditions which determine the motion [see Slattery (1964)]. Thus, the macroscopic behaviour of the suspension on reversing the direction of gravity and $U(x_1)$ is the same as that of reversing the direction of time. Furthermore, such a suspension possesses an unfading infinitely long memory, in that the effects of any such gravity and $U(x_1)$ reversal are felt equally strongly however distant in the past such a reversal occurred. As mentioned above, it is not certain whether purely hydrodynamic interactions can be responsible for the finger formation observed by Weiland *et al.* (1984) but *if* they can, then a reversal of the direction of gravity after the fingers have formed would cause the fingers to disappear (as if time were reversed) and the homogeneous suspension to be regained. Then at a still later time fingers should reappear.

Non-linear effects acting during the collision of a pair of spheres [see (b) above] can cause the individual spheres to be displaced horizontally. For example, if the sphere surfaces are rough they may make actual physical contact if at their position of closest approach (position A in figure 2) the predicted gap between the spheres is less than the height of the dimensional surface roughness Δ^*. Such a roughness would physically push the spheres apart so that their orbits would be as shown in figure 3. Thus, starting at position B, the spheres will move under purely hydrodynamic interactions until position C, where the predicted gap between the spheres takes the value Δ^*. Then, assuming that the roughness prevents relative motion of the sphere surfaces at the contact point, the spheres will rotate as if rigidly attached to each other until the sphere centres are on the same horizontal level (position D). Taking the gap between the spheres at this position as Δ^*, the subsequent motion may be calculated by again assuming purely hydrodynamic interactions between the spheres. The roughness of the spheres will in general be of great importance: one need

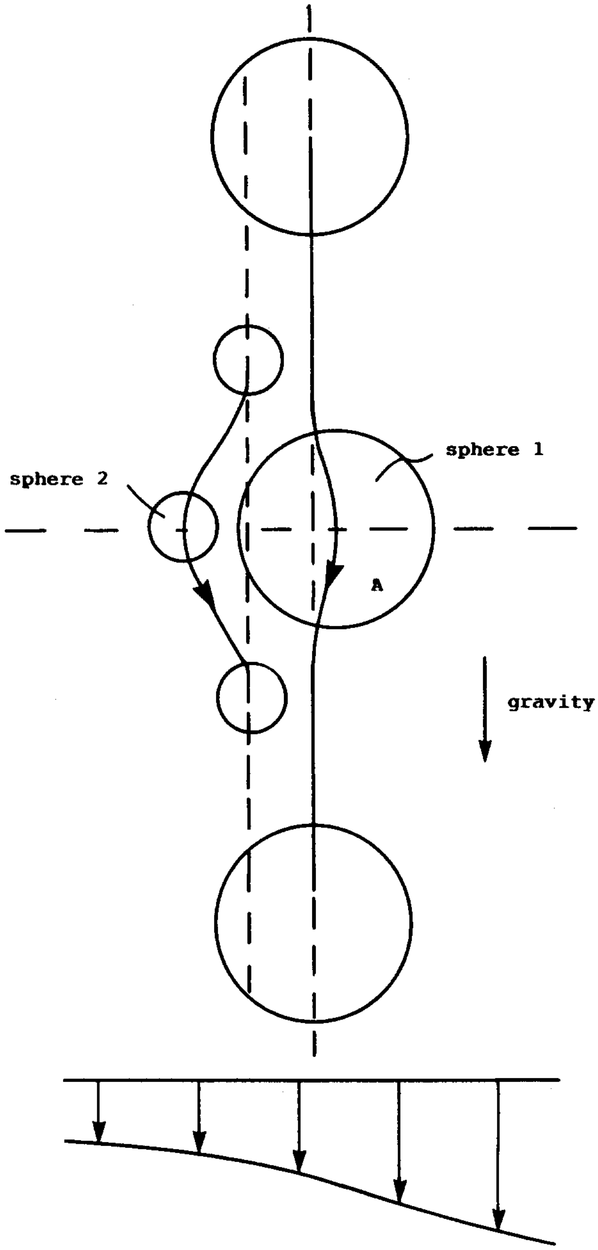


Figure 2. Orbits of the centres of spheres 1 and 2 during a purely hydrodynamic collision.

only note that for the case of neutrally buoyant equal-sized spheres $a_1 = a_2 (= a$ say) in shear flow, $U(x_1) = \gamma x_1$, if the initial horizontal separation between sphere centres is $0.316a$ in the x_1-direction well before the collision [corresponding to $C = 0.1$, $B = 0$ in Arp & Mason (1977)] then for roughness of the spheres to be important Δ^*/a must be at least 9.5×10^{-5} [see Table V in Arp & Mason (1977)], which for the spheres of radius 68 μm used by Weiland *et al.* (1984) gives a remarkably small value for Δ^* of 6.2 nm.

Another non-linear effect which may be of importance, particularly for small particles, is the repulsion which may exist between particles due to electrostatic double-layer forces. This should have an effect very similar to that of the surface roughness in pushing the particles apart horizontally as they collide with each other. Additional non-linear effects which may possibly be important during the collision of a pair of spheres include non-Newtonian and non-continuum effects in the liquid, cavitation and elastic deformation of the spherical particles. Should the colliding particles be immiscible drops instead of solid spheres, then non-linearity is introduced due to the deformation of the drop surfaces during the collision process.

In the present paper we consider only horizontal displacements of the spheres resulting from two-sphere collisions with some non-linearity present. It will, however, be shown that under the assumed condition of low solids concentration that the effects of hydrodynamic three-sphere interactions are much smaller than those produced by the above two-sphere collisions and may therefore be neglected.

4. HORIZONTAL DISPLACEMENT DUE TO A COLLISION

Consider the motion relative to a fixed observer of a sphere of species i ($i = 1, 2$) which interacts with a sphere of species j ($j = 1, 2$), the centre of the latter sphere having a position vector relative to the centre of the former which possesses horizontal components $(\delta x_1, \delta x_2)$ before the interaction takes place (i.e. when both spheres are moving vertically under gravity and the flow $U(x_1)$).

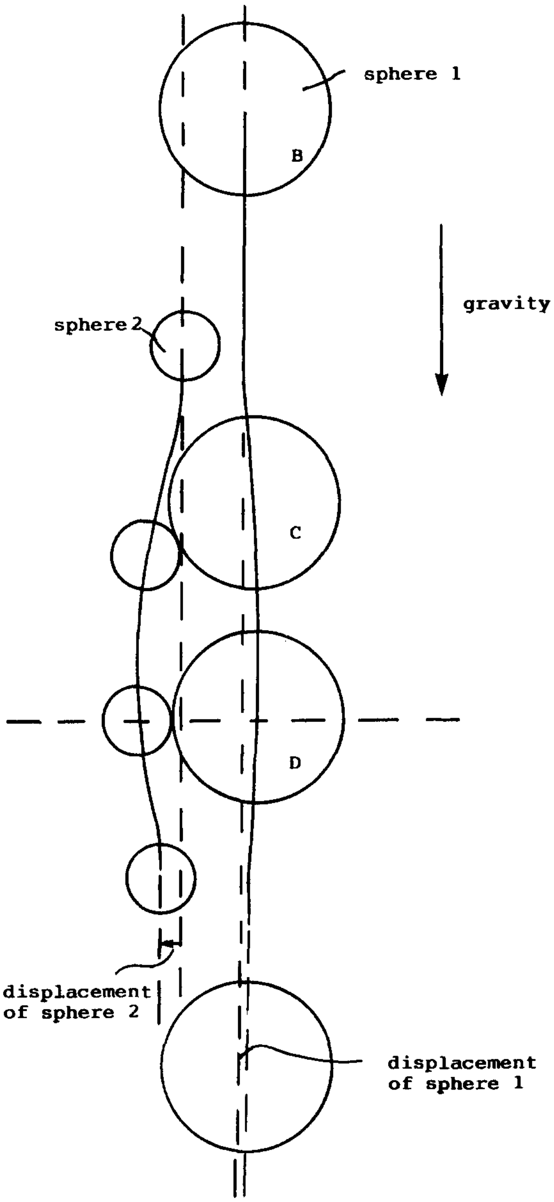


Figure 3. Orbits of the centres of spheres 1 and 2 during a collision of rough spheres where physical contact is made between the spheres from position C to position D.

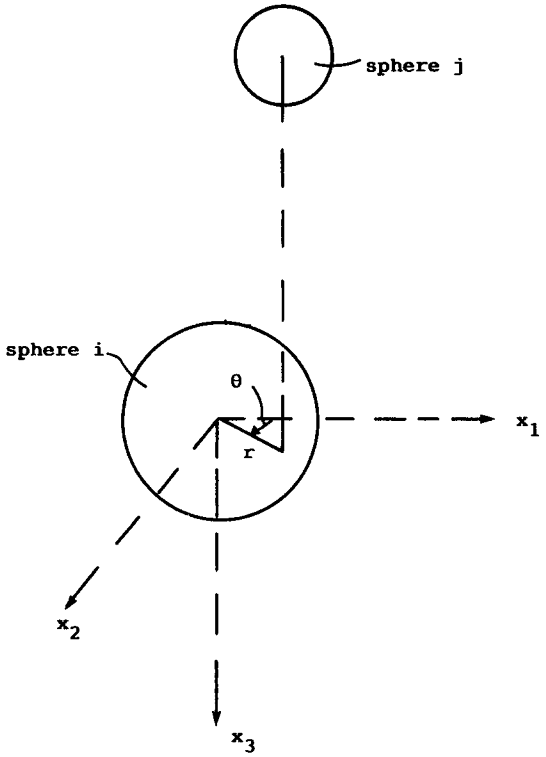


Figure 4. Positions of two spheres prior to a collision.

Define the polar coordinates

$$\delta x_1 = r\cos\theta, \quad \delta x_2 = r\sin\theta, \qquad [10]$$

as shown in figure 4.

As a result of the surface roughness of the spheres (or of some other non-linear effect), the centre of sphere i after the interaction (when both spheres are again moving vertically) will be displaced a distance Δx_1 in the x_1-direction, which will be denoted by F_{ij}. If $\hat{x}_1$ is the value of x_1 at the initial position (before the collision) of the centre of the sphere i we may expand the undisturbed flow field $U(x_1)$ in the neighbourhood of the spheres as

$$U(x_1) = U(\hat{x}_1) + \left\{\frac{\mathrm{d}U}{\mathrm{d}x_1}(\hat{x}_1)\right\}(x_1 - \hat{x}_1) + \frac{1}{2!}\left\{\frac{\mathrm{d}^2U}{\mathrm{d}x_1^2}(\hat{x}_1)\right\}(x_1 - \hat{x}_1)^2 + \cdots. \qquad [11]$$

Thus, F_{ij} for $i = 1, 2$ and $j = 3 - i$ must be a function of

$$r, \theta, \Delta^*, a_1, a_2, V_1, V_2, \frac{\mathrm{d}U}{\mathrm{d}x_1}, \frac{\mathrm{d}^2U}{\mathrm{d}x_1^2}, \ldots.$$

The uniform velocity $U(\hat{x}_1)$ does not appear since it may be eliminated by translating the observer with that velocity. We now use dimensional analysis and also make use of

$$\frac{\mathrm{d}U}{\mathrm{d}x_1} = \frac{U_0}{L}\frac{\mathrm{d}\tilde{U}}{\mathrm{d}\tilde{x}_1}, \quad \frac{\mathrm{d}^2U}{\mathrm{d}x_1^2} = \frac{U_0}{L^2}\frac{\mathrm{d}^2\tilde{U}}{\mathrm{d}\tilde{x}_1^2} \quad \text{etc.,} \qquad [12]$$

obtained from [7] and [8], in order to obtain the functional form for Δx_1 shown in table 1. Here $(V_2 - V_1)$ rather than V_1 or V_2 has been used to non-dimensionalize $\tilde{U}'$ since, by doing so, the subsequent analysis is simplified. For the interaction between a pair of spheres due to sedimentation alone (with $U(x_1) = 0$), the value of Δx_1 is denoted by F_{ijsd} and has the functional form shown in table 1. Results are not given for F_{ijsd} for $i = j = 1$ or $i = j = 2$, since identical spheres on sedimenting keep their same relative positions (Goldman *et al.* 1966) so no collision will occur.

Table 1. Functional form of Δx_1 for sedimentation and flow and also for sedimentation alone†

i	j	Form of Δx_1 for sedimentation and flow
1	2	$\frac{\Delta x_1}{a_1+a_2}=F_{12}\left(\frac{r}{a_1+a_2},\theta,\frac{\Delta^*}{a_1+a_2},\frac{a_2}{a_1},\frac{V_2}{V_1},\frac{(a_1+a_2)U_0}{L(V_2-V_1)}\tilde{U}',\frac{(a_1+a_2)^2U_0}{L^2(V_2-V_1)}\tilde{U}'',\ldots\right)$
2	1	$\frac{\Delta x_1}{a_1+a_2}=F_{21}=F_{12}\left(\frac{r}{a_1+a_2},\theta,\frac{\Delta^*}{a_1+a_2},\frac{a_1}{a_2},\frac{V_1}{V_2},-\frac{(a_1+a_2)U_0}{L(V_2-V_1)}\tilde{U}',-\frac{(a_1+a_2)^2U_0}{L^2(V_2-V_1)}\tilde{U}'',\ldots\right)$
1	1	$\frac{\Delta x_1}{2a_1}=F_{11}=F_{12}\left(\frac{r}{2a_1},\theta,\frac{\Delta^*}{2a_1},+1,+1,\infty,\infty,\ldots\right)$
2	2	$\frac{\Delta x_1}{2a_2}=F_{22}=F_{12}\left(\frac{r}{2a_2},\theta,\frac{\Delta^*}{2a_2},+1,+1,\infty,\infty,\ldots\right)$
i	j	Form of Δx_1 for sedimentation alone
1	2	$\frac{\Delta x_1}{a_1+a_2}=F_{12sd}=F_{12}\left(\frac{r}{a_1+a_2},\theta,\frac{\Delta^*}{a_1+a_2},\frac{a_2}{a_1},\frac{V_2}{V_1},0,0,\ldots\right)$
2	1	$\frac{\Delta x_1}{a_1+a_2}=F_{21sd}=F_{12}\left(\frac{r}{a_1+a_2},\theta,\frac{\Delta^*}{a_1+a_2},\frac{a_1}{a_2},\frac{V_1}{V_2},0,0,\ldots\right)$

†$\tilde{U}'=\frac{d\tilde{U}}{d\tilde{x}}$, $\tilde{U}''=\frac{d^2\tilde{U}}{d\tilde{x}_1^2}$ etc.

For pure sedimentation, the collision between a sphere 1 and a sphere 2 must, be symmetry, result in each sphere being displaced in the radial r-direction so that Δx_1 for each sphere must be proportional to $\cos\theta$. Thus, F_{12sd} must be of the form

$$F_{12sd}=\cos\theta\cdot G_{12sd}\left(\frac{r}{a_1+a_2},\frac{\Delta^*}{a_1+a_2},\frac{a_2}{a_1},\frac{V_2}{V_1}\right). \qquad [13]$$

5. PROBABILITY DISTRIBUTION OF Δx_1

Assuming that the sizes of the two species of sphere are of the same order of magnitude (i.e. $|a_2/a_1|$ is neither very small nor very large compared with unity), we note that the typical time for a collision between spheres $[=\min(a/|V_2-V_1|,\ 1/|U'|)]$ is very much smaller than the typical time between collisions $[=\min(ac^{-1/3}/|V_2-V_1|,\ c^{-1/3}/|U'|)]$. Here and elsewhere, a and c denote characteristic values of particle radius and concentration and primes denote differentiation with respect to x_1. Thus, in any time interval Δt satisfying

$$\min\left(\frac{a}{|V_2-V_1|},\frac{1}{|U'|}\right)\ll\Delta t\ll\min\left(\frac{ac^{-1/3}}{|V_2-V_1|},\frac{c^{-1/3}}{|U'|}\right) \qquad [14]$$

the probability of any sphere undergoing more than one collision is very small, so it will be assumed that a sphere undergoes either no collision or just one collision. In addition, any collision that occurs may be considered complete so that sphere displacements in the x_1-direction are as described in the previous section.

We consider now a sphere of species 1 with centre initially at position $x_1=\hat{x}_1$ and calculate the probability distribution $p(\Delta x_1)$ of its displacement Δx_1 in the x_1-direction in the time Δt. In this time interval the sphere either undergoes no collision (giving $\Delta x_1=0$) or it collides with a sphere 2 or with another sphere 1. However, it will be assumed that the concentrations c_1 and c_2 of the two species of sphere are of the same order of magnitude (i.e. c_2/c_1 is neither very small nor very large compared with unity) and that in some sense (to be discussed later) the effects of particle sedimentation dominate over that of the shear $U'(x_1)$. Since spheres of the same species have no relative motion when undergoing pure sedimentation it is seen that under the above assumptions the velocity of approach to the sphere 1 by another sphere 1 is very much lower than that of a sphere 2. It therefore follows that in the time Δt there is a much greater probability that the sphere 1 under consideration will collide with a sphere 2 rather than another sphere 1. It is this situation which we will consider in the present paper. Thus, we assume that either the sphere 1 undergoes no collision or a collision with a sphere 2 in the time Δt. The inclusion of the effects of collisions with another sphere 1 (which would be important for $c_1 \gg c_2$ or for large $U'(x_1)$) will not be considered here. It is shown in the appendix that under the conditions of low concentration considered here that the horizontal movement of particles due to purely hydrodynamic interactions between three or more spheres may be neglected.

If the centre of the sphere 1 is at $x_1 = \hat{x}_1$ before the collision with a sphere 2, the centre of the sphere 2 will be at $x_1 = \hat{x}_1 + r\cos\theta$, $x_2 = r\sin\theta$, as shown in figure 4. The velocity of the centre of the sphere 1 before the collision will therefore be $\{U(\hat{x}_1) + V_1\}$ downwards, while that of the centre of the sphere 2 will be $\{U(\hat{x}_1 + r\cos\theta) + V_2\}$. However, since U should be considered having the form [8], we should write the velocity of sphere 1 as $\{U_0\tilde{U}(\hat{\tilde{x}}_1) + V_1\}$ and that of sphere 2 as $\{U_0\tilde{U}(\hat{\tilde{x}}_1 + r/L\cos\theta) + V_2\}$, where $\hat{\tilde{x}}_1 = \hat{x}_1/L$. The magnitude of the velocity of the sphere 2 relative to the sphere 1 is thus $|U_0\{\tilde{U}(\hat{\tilde{x}}_1 + r/L\cos\theta) - \tilde{U}(\hat{\tilde{x}}_1)\} + V_2 - V_1|$. The number density of the spheres 2 at $x = \hat{x}_1 + r\cos\theta$ is $n_2(\hat{\tilde{x}}_1 + r/L\cos\theta)$ so that in the time Δt the number of spheres crossing a horizontal plane ($\tilde{x}_3$ fixed) and approaching the sphere 1 for r, θ lying in the range $(r, r + \mathrm{d}r)$ and $(\theta, \theta + \mathrm{d}\theta)$ is

$$p(r,\theta)\,\mathrm{d}r\,\mathrm{d}\theta = n_2\left(\hat{\tilde{x}}_1 + \frac{r}{L}\cos\theta\right)\left|U_0\left\{\tilde{U}\left(\hat{\tilde{x}}_1 + \frac{r}{L}\cos\theta\right) - \tilde{U}(\hat{\tilde{x}}_1)\right\} + V_2 - V_1\right| r\,\Delta t\,\mathrm{d}r\,\mathrm{d}\theta. \quad [15]$$

As a result of such a collision the displacement Δx_1 of the sphere 1 in the x_1-direction is given by

$$\frac{\Delta x_1}{a_1 + a_2} = F_{12}\left(\frac{r}{a_1 + a_2}, \theta, \ldots\right), \quad [16]$$

as indicated in section 4. If for any fixed θ, we solve this equation for $r/(a_1 + a_2)$ in terms of $\Delta x_1/(a_1 + a_2)$, we will obtain

$$\frac{r}{a_1 + a_2} = F^*_{12}\left(\frac{\Delta x_1}{a_1 + a_2}, \theta, \ldots\right). \quad [17]$$

Changing from the (r, θ) variables in [15] to (η, θ) variables, where

$$\eta = \frac{\Delta x_1}{(a_1 + a_2)}, \quad [18]$$

so that $r = (a_1 + a_2)F^*_{12}(\eta, \theta, \ldots)$, we obtain the probability distribution $p(\eta, \theta)$ of η and θ as

$$p(\eta,\theta) = n_2\left[\hat{\tilde{x}}_1 + \left(\frac{a_1 + a_2}{L}\right)F^*_{12}\cos\theta\right]\left|U_0\left\{\tilde{U}\left[\hat{\tilde{x}}_1 + \left(\frac{a_1 + a_2}{L}\right)F^*_{12}\cos\theta\right] - \tilde{U}(\hat{\tilde{x}}_1)\right\} + V_2 - V_1\right|(a_1 + a_2)^2 F^*_{12}\left|\frac{\partial F^*_{12}}{\partial\eta}\right|\Delta t, \quad [19]$$

where we have used the result

$$\left|\frac{\partial(r,\theta)}{\partial(\eta,\theta)}\right| = (a_1 + a_2)\left|\frac{\partial F^*_{12}}{\partial\eta}\right|. \quad [20]$$

If this is integrated with respect to θ, we obtain the probability distribution $p(\eta)$ corresponding to the change Δx_1 in x_1 for the sphere 1 in the time interval Δt as

$$p(\eta) = \int_0^{2\pi} n_2\left[\hat{\tilde{x}}_1 + \left(\frac{a_1 + a_2}{L}\right)F^*_{12}\cos\theta\right]\left|U_0\left\{\tilde{U}\left[\hat{\tilde{x}}_1 + \left(\frac{a_1 + a_2}{L}\right)F^*_{12}\cos\theta\right] - \tilde{U}(\hat{\tilde{x}}_1)\right\} + V_2 - V_1\right|(a_1 + a_2)^2 F^*_{12}\left|\frac{\partial F^*_{12}}{\partial\eta}\right|\Delta t\,\mathrm{d}\theta. \quad [21]$$

Since by the assumptions [4], $(a_1 + a_2)/L$ is very small, we may expand terms in the integrand of [21] as

$$n_2\left[\hat{\tilde{x}}_1 + \left(\frac{a_1 + a_2}{L}\right)F^*_{12}\cos\theta\right] \sim n_2(\hat{\tilde{x}}_1) + \left(\frac{a_1 + a_2}{L}\right)F^*_{12}\,n_2'(\hat{\tilde{x}}_1)\cos\theta + O\left(\frac{a_1 + a_2}{L}\right)^2 \quad [22]$$

and

$$\left|U_0\left\{\tilde{U}\left[\hat{x}_1+\left(\frac{a_1+a_2}{L}\right)F^*_{12}\cos\theta\right]-\tilde{U}(\hat{x}_1)\right\}+V_2-V_1\right|$$

$$\sim|V_2-V_1|+U_0\left(\frac{a_1+a_2}{L}\right)F^*_{12}\cos\theta\,\tilde{U}'(\hat{x}_1)\mathrm{sgn}(V_2-V_1)+O\left(\frac{a_1+a_2}{L}\right)^2, \quad [23]$$

where, for [23] to be valid, we require that

$$\frac{(a_1+a_2)U_0}{L|V_2-V_1|}\ll 1, \quad [24]$$

where U_0 is a characteristic value of the velocity $U(x_1)$. Substituting the expansions [22] and [23] in [21] we obtain

$$\rho(\eta)=(a_1+a_2)^2\,\Delta t f_1(\hat{x}_1)g_1(\eta)+\frac{(a_1+a_2)^3}{L}\Delta t[f_2(\hat{x}_1)+f_3(\hat{x}_1)]g_2(\eta)+O\left(\frac{(a_1+a_2)^4}{L^2}\Delta t\right), \quad [25]$$

where

$$f_1(\hat{x}_1)=n_2(\hat{x}_1)|V_2-V_1|,$$
$$f_2(\hat{x}_1)=U_0n_2(\hat{x}_1)\tilde{U}'(\hat{x}_1)\mathrm{sgn}(V_2-V_1),$$
$$f_3(\hat{x}_1)=n_2'(\hat{x}_1)|V_2-V_1|, \quad [26]$$

and

$$g_1(\eta)=\int_0^{2\pi}F^*_{12}\left|\frac{\partial F^*_{12}}{\partial\eta}\right|\mathrm{d}\theta$$

and

$$g_2(\eta)=\int_0^{2\pi}(F^*_{12})^2\left|\frac{\partial F^*_{12}}{\partial\eta}\right|\cos\theta\,\mathrm{d}\theta \quad [27]$$

As a result of the assumptions [4] and [24] it is seen that F_{12}, given in table 1, may be expanded to give

$$\eta=F_{12\mathrm{sd}}\left(\frac{r}{a_1+a_2},\theta,\ldots\right)+\frac{(a_1+a_2)U_0}{L(V_2-V_1)}\tilde{U}'\tilde{F}_{12}\left(\frac{r}{a_1+a_2},\theta,\ldots\right)+O\left(\frac{(a_1+a_2)^2U_0}{L^2(V_2-V_1)}\right) \quad [28]$$

or, by [13],

$$\eta=\cos\theta\,G_{12\mathrm{sd}}\left(\frac{r}{a_1+a_2},\ldots\right)+\frac{(a_1+a_2)U_0}{L(V_2-V_1)}\tilde{U}'\tilde{F}_{12}\left(\frac{r}{a_1+a_2},\theta,\ldots\right)+\cdots. \quad [29]$$

The first term in [29] represents the sphere displacement due to sedimentation alone, while the second term gives the correction due to the shear flow. As a result of the linearity of the creeping flow equations [9] and symmetry it may be shown (see figure 5) that the additional sphere displacement due to the shear is the same if θ is replaced by $(\pi-\theta)$. Thus, $\tilde{F}_{12}$ in [29] is unchanged if θ is replaced by $(\pi-\theta)$, i.e.

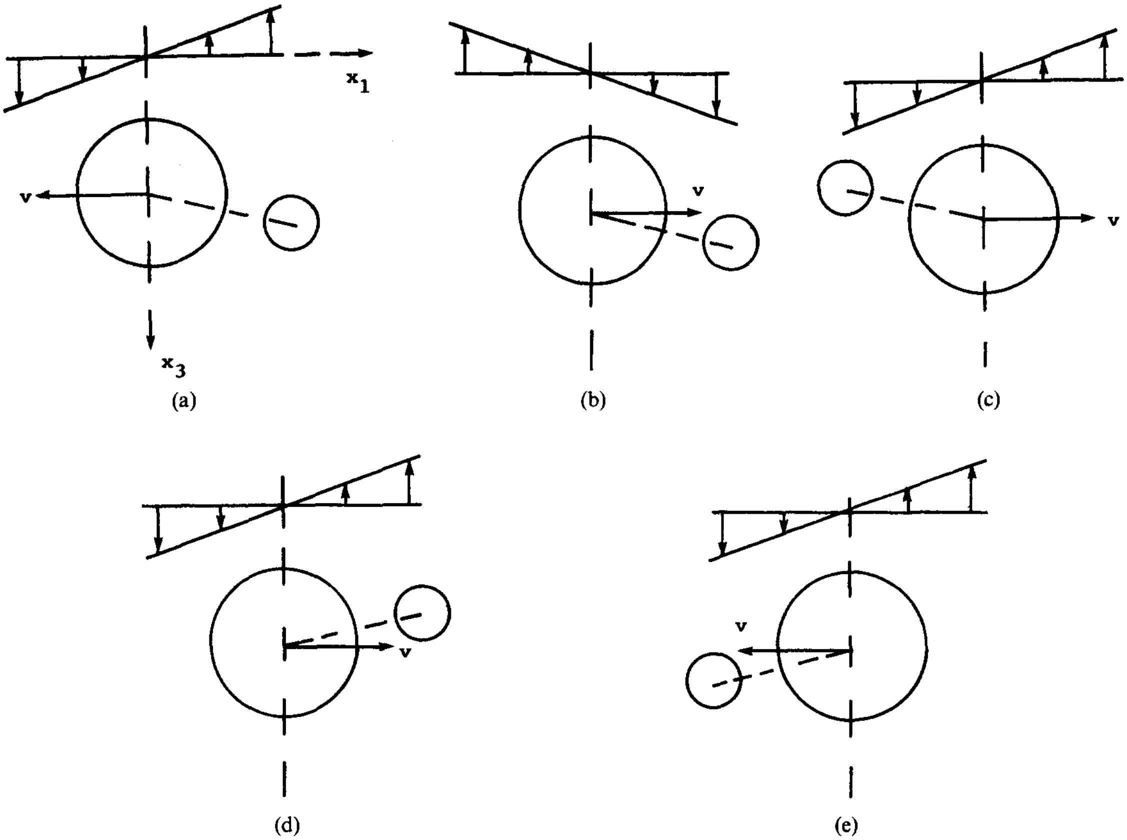
$$\tilde{F}_{12}\left(\frac{r}{a_1+a_2},\pi-\theta,\ldots\right)=\tilde{F}_{12}\left(\frac{r}{a_1+a_2},\theta,\ldots\right). \quad [30]$$

Due to sedimentation alone

$$\frac{\eta}{\cos\theta}=G_{12\mathrm{sd}}\left(\frac{r}{a_1+a_2},\ldots\right), \quad [31]$$

which we suppose may be solved for $r/(a_1+a_2)$ to give

$$\frac{r}{a_1+a_2}=G^*_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta},\ldots\right). \quad [32]$$



Figures 5(a)–(e). The figures show the behaviour of the motion of the sphere 1 as it interacts with the sphere 2 in the presence of the shear. (a) Due to the shear flow suppose the additional velocity of sphere 1 is v as shown. (b) Situation obtained from figure 5(a) by reversing all velocities (and stresses). (c) Situation obtained from figure 5(a) by rotating through π about the x_2-axis. (d) Situation obtained from figure 5(b) by reflection in the x_1x_2-plane. (e) Situation obtained from figure 5(d) by rotating through π about the x_2-axis.

Then solving [29] for $r/(a_1+a_2)$, we obtain the value of F^*_{12} correct to order $(a_1+a_2)U_0/L(V_2-V_1)$ as

$$F^*_{12} = G^*_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right) - \frac{(a_1+a_2)U_0}{L(V_2-V_1)}\frac{U'}{\cos\theta}\tilde{F}_{12}\left(G^*_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right),\theta,\ldots\right)G^{*\prime}_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right)+\cdots. \quad [33]$$

We thus obtain, by expanding $|\partial F^*_{12}/\partial\eta|$ in terms of $[(a_1+a_2)U_0]/[L(V_2-V_1)]$,

$$\left|\frac{\partial F^*_{12}}{\partial\eta}\right| = \frac{\left|G^{*\prime}_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right)\right|}{|\cos\theta|} - \frac{(a_1+a_2)U_0}{L(V_2-V_1)}\frac{\tilde{U}'}{\cos\theta}\frac{\mathrm{sgn}\left\{G^{*\prime}_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right)\right\}}{\mathrm{sgn}(\cos\theta)}$$
$$\times\frac{\mathrm{d}}{\mathrm{d}\eta}\left[\tilde{F}_{12}\left(G^*_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right),\theta,\ldots\right)G^{*\prime}_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right)\right]+\cdots. \quad [34]$$

In [33] and [34] above primes denote differentiation with respect to the first variable listed so that

$$G^{*\prime}_{12\mathrm{sd}}(\bar{\eta},\ldots) = \frac{\partial G^*_{12\mathrm{sd}}}{\partial\bar{\eta}}.$$

By substituting the values of F^*_{12} and $|\partial F^*_{12}/\partial\eta|$ given by [33] and [34] into [27], we obtain

$$g_1(\eta) = g_{10}(\eta) + \frac{(a_1+a_2)U_0}{L(V_2-V_1)}\tilde{U}'g_{11}(\eta)+\cdots \quad [35]$$

and

$$g_2(\eta) = g_{20}(\eta) + O\left(\frac{(a_1 + a_2)U_0}{L(V_2 - V_1)}\right), \tag{36}$$

where

$$g_{10}(\eta) = \int_0^{2\pi} \frac{\left|G^{*\prime}_{12\mathrm{sd}}\left(\dfrac{\eta}{\cos\theta}\right)\right|}{|\cos\theta|} G^{*}_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right) \mathrm{d}\theta, \tag{37}$$

$$g_{11}(\eta) = -\int_0^{2\pi} \frac{\operatorname{sgn}\left(G^{*\prime}_{12\mathrm{sd}}\left(\dfrac{\eta}{\cos\theta}\right)\right)}{|\cos\theta|}$$

$$\times \frac{\mathrm{d}}{\mathrm{d}\eta}\left[G^{*}_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right) G^{*\prime}_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right) \tilde{F}_{12}\left(G^{*}_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right), \theta, \ldots\right)\right] \mathrm{d}\theta \tag{38}$$

and

$$g_{20}(\eta) = \int_0^{2\pi} \left\{G^{*}_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right)\right\}^2 \left|G^{*\prime}_{12\mathrm{sd}}\left(\frac{\eta}{\cos\theta}\right)\right| \operatorname{sgn}(\cos\theta)\, \mathrm{d}\theta. \tag{39}$$

Thus, the value of the probability distribution $p(\eta; \hat{\tilde{x}}_1)$ of η (corresponding to a displacement Δx_1 in a time interval Δt) for a sphere 1 with centre initially at $\tilde{x}_1 = \hat{\tilde{x}}_1$ is given by [25], [35] and [36] as

$$p(\eta; \hat{\tilde{x}}_1) = (a_1 + a_2)^2 \Delta t \left\{ f_1(\hat{\tilde{x}}_1) g_{10}(\eta) + \frac{(a_1 + a_2)U_0}{L(V_2 - V_1)} \tilde{U}'(\hat{\tilde{x}}_1) f_1(\hat{\tilde{x}}_1) g_{11}(\eta) \right.$$
$$\left. + \left(\frac{a_1 + a_2}{L}\right) [f_2(\hat{\tilde{x}}_1) + f_3(\hat{\tilde{x}}_1)] g_{20}(\eta) + \cdots \right\}. \tag{40}$$

If we write

$$\hat{\eta} = -\eta, \quad \hat{\theta} = \pi - \theta$$

and change from the (η, θ) variables to $(\hat{\eta}, \hat{\theta})$ variables in [37]–[39] and make use of result [30], we obtain

$$g_{10}(-\eta) = g_{10}(\eta), \tag{41}$$

$$g_{11}(-\eta) = -g_{11}(\eta) \quad \text{and} \quad g_{20}(-\eta) = -g_{20}(\eta), \tag{42}$$

so that g_{10} is an even function of η, while g_{11} and g_{20} are odd functions of η.

6. MACROSCOPIC EQUATIONS

In order to obtain the macroscopic equations for the volume concentration $c_1(\tilde{x}_1)$ of the spheres of species 1, we consider the transport of spheres 1 into and out of a volume of the suspension in the form of a thin slab $\hat{\tilde{x}}_1 < \tilde{x}_1 < \hat{\tilde{x}}_1 + \mathrm{d}\hat{\tilde{x}}_1$ with unit cross-sectional area in the $\tilde{x}_2\tilde{x}_3$-plane. In a time Δt the number of spheres of species 1 leaving such a volume is

$$n_1(\hat{\tilde{x}}_1)\, \mathrm{d}\hat{\tilde{x}}_1 \int_{\eta=-\infty}^{+\infty} p(\eta; \hat{\tilde{x}}_1)\, \mathrm{d}\eta, \tag{43}$$

while the number of spheres 1 entering the volume from outside is

$$\int_{\eta=-\infty}^{+\infty} p\left(\eta; \hat{\tilde{x}}_1 - \left(\frac{a_1 + a_2}{L}\right)\eta\right) n_1\left[\hat{\tilde{x}}_1 - \left(\frac{a_1 + a_2}{L}\right)\eta\right] \mathrm{d}\hat{\tilde{x}}_1\, \mathrm{d}\eta. \tag{44}$$

Thus, the increase $\Delta n_1(\hat{\tilde{x}}_1)$ of $n_1(\hat{\tilde{x}}_1)$ in the time Δt is given by

$$\Delta n_1(\hat{\tilde{x}}_1) = \int_{\eta=-\infty}^{+\infty} \left\{ p\left(\eta; \hat{\tilde{x}}_1 - \left(\frac{a_1+a_2}{L}\right)\eta\right) n_1\left[\hat{\tilde{x}}_1 - \left(\frac{a_1+a_2}{L}\right)\eta\right] - p(\eta; \hat{\tilde{x}}_1) n_1(\hat{\tilde{x}}_1)\right\} \mathrm{d}\eta. \quad [45]$$

Since $(a_1 + a_2)/L \ll 1$, the integrand may be expanded to give

$$\begin{aligned} \Delta n_1(\hat{\tilde{x}}_1) = &-\left(\frac{a_1+a_2}{L}\right)\Bigg[n_1(\hat{\tilde{x}}_1)\int_{\eta=-\infty}^{\infty} \eta \frac{\partial p}{\partial \tilde{x}_1}(\eta; \hat{\tilde{x}}_1)\,\mathrm{d}\eta \\ &+ n_1'(\hat{\tilde{x}}_1)\int_{\eta=-\infty}^{\infty} \eta p(\eta; \hat{\tilde{x}}_1)\,\mathrm{d}\eta\Bigg] \\ &+\frac{1}{2}\left(\frac{a_1+a_2}{L}\right)^2\Bigg[n_1(\hat{\tilde{x}}_1)\int_{\eta=-\infty}^{\infty} \eta^2 \frac{\partial^2 p}{\partial \tilde{x}_1^2}(\eta; \hat{\tilde{x}}_1)\,\mathrm{d}\eta \\ &+ 2n_1'(\hat{\tilde{x}}_1)\int_{\eta=-\infty}^{\infty} \eta^2 \frac{\partial p}{\partial \tilde{x}_1}(\eta; \hat{\tilde{x}}_1)\,\mathrm{d}\eta \\ &+ n_1''(\hat{\tilde{x}}_1)\int_{\eta=-\infty}^{\infty} \eta^2 p(\eta; \hat{\tilde{x}}_1)\,\mathrm{d}\eta\Bigg] + O\left(\frac{a_1+a_2}{L}\right)^3, \end{aligned} \quad [46]$$

where, for example $(\partial p/\partial \tilde{x}_1)(\eta; \hat{\tilde{x}}_1)$ means the partial derivative of $p(\eta; \tilde{x}_1)$ with respect to $\tilde{x}_1$ evaluated at $\tilde{x}_1 = \hat{\tilde{x}}_1$. If the value of $p(\eta; \hat{\tilde{x}}_1)$ given by [40] is substituted into [46], we obtain an equation, which when divided by Δt and the limit $\Delta t \to 0$ taken, yields

$$\begin{aligned} \frac{\partial n_1}{\partial t} = \frac{(a_1+a_2)^4}{L^2}\frac{\partial}{\partial \tilde{x}_1}\Bigg[&-\left(\frac{U_0}{V_2-V_1}\right) n_1 \tilde{U}' f_1 \int_{\eta=-\infty}^{\infty} \eta g_{11}\,\mathrm{d}\eta \\ &- n_1(f_2+f_3)\int_{\eta=-\infty}^{\infty} \eta g_{20}\,\mathrm{d}\eta + \tfrac{1}{2}(n_1 f_1' + n_1' f_1)\int_{\eta=-\infty}^{\infty} \eta^2 g_{10}\,\mathrm{d}\eta + \cdots\Bigg], \end{aligned} \quad [47]$$

where we have used the result, obtained from [41], that

$$\int_{\eta=-\infty}^{\infty} \eta g_{10}\,\mathrm{d}\eta = 0. \quad [48]$$

In [47] we have also replaced $\hat{\tilde{x}}_1$ by the general value $\tilde{x}_1$ of the coordinate. Substitution of the values of f_1, f_2 and f_3 given by [26] then yields the macroscopic equation for $n_1(\tilde{x}_1, t)$ as

$$\begin{aligned} \frac{\partial n_1}{\partial t} = \frac{(a_1+a_2)^4}{L^2}\frac{\partial}{\partial \tilde{x}_1}[&(\alpha_1^* + \beta_1^*) U_0 \tilde{U}' n_1 n_2 \operatorname{sgn}(V_2 - V_1) \\ &+ \beta_1^* n_1 n_2' |V_2 - V_1| + \tfrac{1}{2}\gamma_1^*(n_1 n_2' + n_1' n_2)|V_2 - V_1| + \cdots], \end{aligned} \quad [49]$$

where

$$\alpha_1^* = -\int_{\eta=-\infty}^{\infty} \eta g_{11}\,\mathrm{d}\eta, \quad \beta_1^* = -\int_{\eta=-\infty}^{\infty} \eta g_{20}\,\mathrm{d}\eta \quad \text{and} \quad \gamma_1^* = +\int_{\eta=-\infty}^{\infty} \eta^2 g_{10}\,\mathrm{d}\eta. \quad [50]$$

If we write [49] in terms of the volume concentrations c_1 and c_2 of the two sphere species using [2] and change to dimensional variables using [7] and [8], we obtain

$$\begin{aligned} \frac{\partial c_1}{\partial t} = k_1 \frac{\partial}{\partial x_1}\Bigg[&(\alpha_1^* + \beta_1^*) c_1 c_2 \frac{\partial U}{\partial x_1} \operatorname{sgn}(V_2 - V_1) + \beta_1^* c_1 \frac{\partial c_2}{\partial x_1}|V_2 - V_1| \\ &+ \tfrac{1}{2}\gamma_1^*\left(c_1 \frac{\partial c_2}{\partial x_1} + c_2 \frac{\partial c_1}{\partial x_1}\right)|V_2 - V_1| + \cdots\Bigg], \end{aligned} \quad [51]$$

where $k_1 = 3(a_1 + a_2)^4/4\pi a_2^3$ is a constant. The values of α_1^*, β_1^* and γ_1^*, obtained by substituting the values of g_{10}, g_{11} and g_{20} given by [37]–[39] into [50], are

$$\alpha_1^* = \int_{\eta=-\infty}^{\infty}\int_{\theta=0}^{2\pi} \frac{\eta\,\mathrm{sgn}\left(G_{12sd}^{*\prime}\left(\frac{\eta}{\cos\theta}\right)\right)}{|\cos\theta|} \times \frac{\mathrm{d}}{\mathrm{d}\eta}\left[G_{12sd}^{*}\left(\frac{\eta}{\cos\theta}\right)G_{12sd}^{*\prime}\left(\frac{\eta}{\cos\theta}\right)\tilde{F}_{12}\left(G_{12sd}^{*}\left(\frac{\eta}{\cos\theta}\right),\theta,\ldots\right)\right]\mathrm{d}\eta\ \mathrm{d}t,$$

$$\beta_1^* = -\int_{\eta=-\infty}^{\infty}\int_{\theta=0}^{2\pi}\eta\left\{G_{12sd}^{*}\left(\frac{\eta}{\cos\theta}\right)\right\}^2\left|G_{12sd}^{*\prime}\left(\frac{\eta}{\cos\theta}\right)\right|\mathrm{sgn}(\cos\theta)\,\mathrm{d}\eta\ \mathrm{d}\theta$$

and

$$\gamma_1^* = \int_{\eta=-\infty}^{\infty}\int_{\theta=0}^{2\pi}\frac{\eta^2\left|G_{12sd}^{*\prime}\left(\frac{\eta}{\cos\theta}\right)\right|}{|\cos\theta|}G_{12sd}^{*}\left(\frac{\eta}{\cos\theta}\right)\mathrm{d}\eta\ \mathrm{d}\theta, \qquad [52]$$

which may be further simplified by changing the integration variables from (η, θ) to (ξ, θ), where ξ is defined by

$$\xi = G_{12sd}^{*}\left(\frac{\eta}{\cos\theta}\right)$$

or

$$\frac{\eta}{\cos\theta} = G_{12sd}(\xi), \qquad [53]$$

and thus represents the quantity $r/(a_1 + a_2)$. Thus, we obtain

$$\alpha_1^* = -\int_{\xi=0}^{\infty}\int_{\theta=0}^{2\pi}\xi\tilde{F}_{12}(\xi,\theta)\,\mathrm{d}\xi\ \mathrm{d}\theta, \qquad [54]$$

$$\beta_1^* = -\pi\int_{\xi=0}^{\infty}\xi^2 G_{12sd}(\xi)\,\mathrm{d}\xi \qquad [55]$$

and

$$\gamma_1^* = +\pi\int_{\xi=0}^{\infty}\xi[G_{12sd}(\xi)]^2\,\mathrm{d}\xi, \qquad [56]$$

where integration by parts with respect to ξ has been used in order to yield [54]. By interchanging the roles of the spheres 1 and 2, we see that the macroscopic equation for c_2 in dimensional variables is

$$\frac{\partial c_2}{\partial t} = k_2\frac{\partial}{\partial x_1}\left[(\alpha_2^* + \beta_2^*)c_1c_2\frac{\partial U}{\partial x_1}\mathrm{sgn}(V_1 - V_2) + \beta_2^* c_2\frac{\partial c_1}{\partial x_1}|V_2 - V_1| + \tfrac{1}{2}\gamma_2^*\left(c_2\frac{\partial c_1}{\partial x_1} + c_1\frac{\partial c_2}{\partial x_1}\right)|V_2 - V_1| + \cdots\right], \qquad [57]$$

where $k_2 = 3(a_1 + a_2)^4/4\pi a_1^3$ is a constant and where

$$\alpha_2^* = -\int_{\xi=0}^{\infty}\int_{\theta=0}^{2\pi} \xi \tilde{F}_{21}(\xi, \theta)\, d\xi\, d\theta, \tag{58}$$

$$\beta_2^* = -\pi \int_{\xi=0}^{\infty} \xi^2 G_{21sd}(\xi)\, d\xi \tag{59}$$

and

$$\gamma_2^* = +\pi \int_{\xi=0}^{\infty} \xi [G_{21sd}(\xi)]^2\, d\xi, \tag{60}$$

and from table 1 (and [29])

$$G_{21sd}\left(\xi, \frac{\Delta^*}{a_1 + a_2}, \frac{a_1}{a_2}, \frac{V_1}{V_2}\right) = G_{12sd}\left(\xi, \frac{\Delta^*}{a_1 + a_2}, \frac{a_2}{a_1}, \frac{V_2}{V_1}\right) \tag{61}$$

and

$$\tilde{F}_{21}\left(\xi, \theta, \frac{\Delta^*}{a_1 + a_2}, \frac{a_1}{a_2}, \frac{V_1}{V_2}\right) = -\tilde{F}_{12}\left(\xi, \theta, \frac{\Delta^*}{a_1 + a_2}, \frac{a_2}{a_1}, \frac{V_2}{V_1}\right). \tag{62}$$

In addition to the two non-linear coupled diffusion equations, [51] and [57], which we have derived for c_1 and c_2, we must also consider a vertical momentum equation since the variation of c_1 and c_2 with x_1 will in general result in mean density variations of the suspension in the x_1-direction which would affect the flow field $U(x_1, t)$. A unit volume of the suspension contains n_1 spheres 1, each of mass $4\pi a_1^3 \rho_1/3$, and n_2 spheres 2, each of mass $4\pi a_2^3 \rho_2/3$. Thus, the macroscopic density of the suspension is

$$\rho + \tfrac{4}{3}\pi a_1^3(\rho_1 - \rho)n_1 + \tfrac{4}{3}\pi a_2^3(\rho_2 - \rho)n_2,$$

which by [1] and [2] may be written as

$$\rho + \frac{9\mu}{2g}\left(\frac{c_1 V_1}{a_1^2} + \frac{c_2 V_2}{a_2^2}\right).$$

Thus, if P is the pressure, the momentum equations on the macroscopic scale are

$$\frac{\partial^2 U}{\partial x_1^2} - K + \frac{9}{2}\left(\frac{c_1 V_1}{a_1^2} + \frac{c_2 V_2}{a_2^2}\right) = \frac{1}{\nu}\frac{\partial U}{\partial t} \tag{63}$$

and

$$\frac{\partial P}{\partial x_1} = 0, \tag{64}$$

where

$$K = \frac{1}{\mu}\left(\frac{\partial P}{\partial x_3} - \rho g\right)$$

and $\nu = \mu/\rho$ is the kinematic viscosity of the fluid. Since, by [64], P is independent of x_1 it follows that in [63] K can be only a function of time t. The inertia term has been included in [63] since the Reynolds number based on the macroscopic length scale may possibly be of order unity or larger.

The diffusion equations [51] and [57] together with the momentum equation [63] constitute a set of three partial differential equations for the concentrations c_1 and c_2 and the velocity field U. It should be noted that in deriving [51] and [57] it was assumed that the time for a particle collision was very much smaller than the time over which macroscopic quantities vary. That this is indeed so in general, is seen by noting that the particle collision time (or order $a/|V_2 - V_1|$) is very much smaller than the characteristic times predicted from [51] and [57]. However, shorter times could occur in the flow field (given by [63]), particularly if such a flow were to be produced by the impulsive motion of solid boundaries.

7. PHYSICAL INTERPRETATION OF THE RESULTS

From the macroscopic diffusion equation [51] for c_1, it is observed that the flux J_1 (i.e. the volume of spheres crossing unit area per unit time) of spheres 1 in the positive x_1-direction is

$$J_1 = -k_1\left[(\alpha_1^* + \beta_1^*)c_1 c_2 \frac{\partial U}{\partial x_1}\operatorname{sgn}(V_2 - V_1) + \beta_1^* c_1 \frac{\partial c_2}{\partial x_1}|V_2 - V_1| + \tfrac{1}{2}\gamma_1^*\left(c_1\frac{\partial c_2}{\partial x_1} + c_2\frac{\partial c_1}{\partial x_1}\right)|V_2 - V_1| + \cdots\right]. \quad [65]$$

In the light of the derivation of this expression, given in sections 5 and 6, we will give a physical interpretation of each of the terms appearing in [65]:

(i) The flux $-k_1[\alpha_1^* c_1 c_2(\partial U/\partial x_1)\operatorname{sgn}(V_2 - V_2)]$ is a *convective* flux of the spheres 1 resulting from the adjustment to the orbits due to the small shear flow which is present. This is shown in figure 6, from which it is observed that, at least for the case shown, F_{12} is negative, giving (see [54]) $\alpha_1^* > 0$, with the flux being in the negative x_1-direction (for $V_2 > V_1$ and $\partial U/\partial x_1 > 0$). However, there may possibly be situations for which α_1^* is negative.

(ii) The flux $-k_1[\beta_1^* c_1 c_2(\partial U/\partial x_1)\operatorname{sgn}(V_2 - V_1)]$ is a convective flux of spheres 1 resulting from sedimentation alone and is due to the fact that the collision

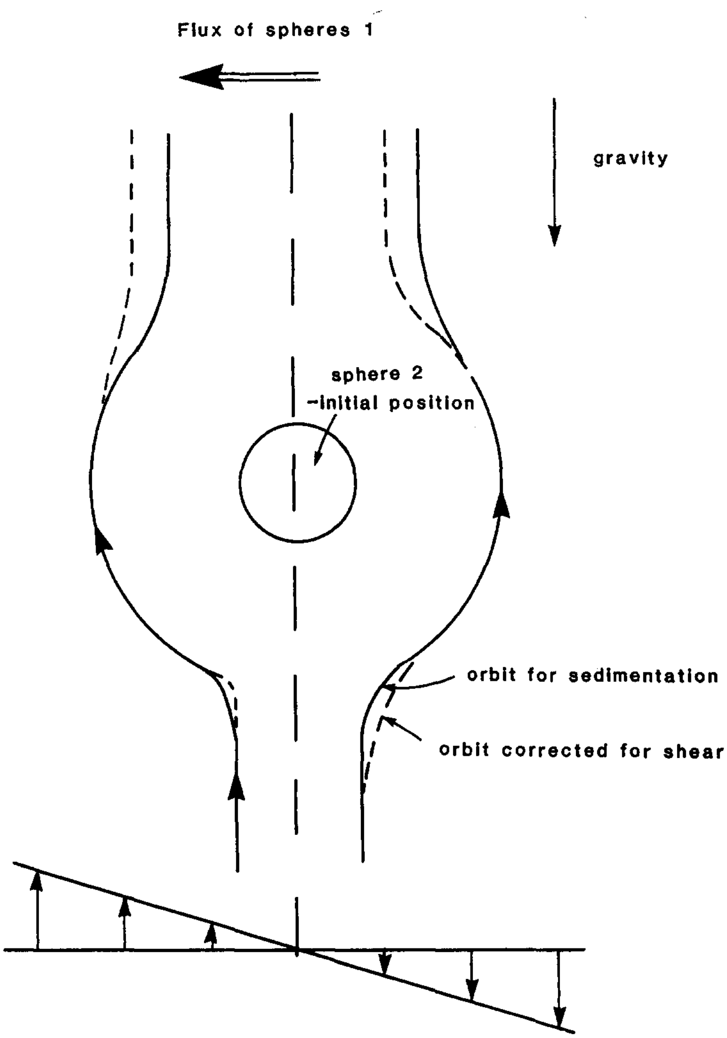


Figure 6. Flux of spheres 1 resulting from the disturbance of the orbits due to the shear flow.

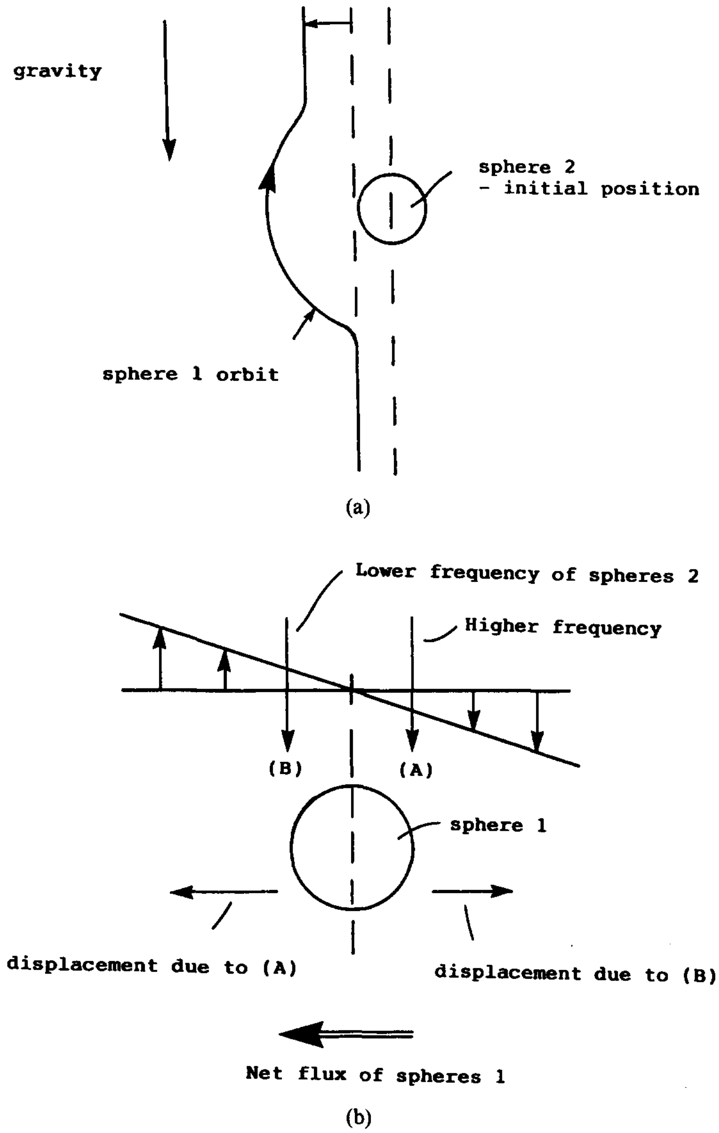


Figure 7. (a) The orbit of sphere 1 due to sedimentation alone. (b) Flux of spheres 1 resulting from a higher collision frequency with spheres 2 on the downflow side (if $V_2 > V_1$).

frequency with spheres 2 is higher on the upflow side (if sphere 1 is the less dense sphere, i.e. $V_2 > V_1$) (see figure 7).

(iii) The flux $-k_1[(\beta_1^* + \frac{1}{2}\gamma_1^*)c_1(\partial c_2/\partial x_1)|V_2 - V_1|]$ is a flux of the spheres resulting from sedimentation alone and is due to the fact that the collision frequency with spheres 2 is higher on that side of sphere 1 where the concentration of spheres 2 is higher (see figure 8).

(iv) The flux $-k_1[\frac{1}{2}\gamma_1^* c_2(\partial c_1/\partial x_1)|V_2 - V_1|]$ is a diffusive flux of spheres 1 from high to low concentrations of the spheres 1 (since, by [56], γ_1^* is strictly positive) and is due to the random displacements they undergo as a result of their collisions with spheres 2 as they sediment.

While γ_1^* is always positive, the signs of α_1^* and β_1^* are uncertain. Although the situation indicated in figure 7 shows $G_{12sd}(\xi)$ as being negative (and hence the value of β_1^* from [55] as positive), the interacting particles will in general experience a lift force which can result in a net horizontal displacement of the particles (especially if V_1 and V_2 are of the same sign) for which $G_{12sd}(\xi)$ may be positive (leading to a negative value of β_1^*).

It is observed from [65] that a flux of spheres 1 can be generated by either (a) a shear flow, (b) a gradient in concentration of spheres 2 or (c) a gradient in concentration of spheres 1. While the flux due to the gradient in concentration of spheres 1 is always from high to low concentrations of spheres 1, the flux due to a gradient in velocity or to a gradient in concentration of spheres 2 may be in either direction (depending on the values of α_1^*, β_1^* and γ_1^*).

For the special situation of equal-sized spheres ($a_1/a_2 = +1$) which sediment with equal speeds but in opposite directions ($V_1/V_2 = -1$) and which are very rough [$\Delta^*/(a_1 + a_2)$ is not too small], the interaction between a sphere 1 and a sphere 2 can be approximated by the motion shown in figure 9 in which sphere 1, on reaching the same horizontal level as sphere 2, continues to move vertically as if sphere 2 has a negligible influence. Since, by symmetry, the horizontal displacement of sphere 1 in space is one-half its horizontal displacement relative to sphere 2, and since (r, θ) describes the horizontally projected initial position of the centre of sphere 2 relative to that of sphere 1, it follows that

$$\tilde{F}_{12}(\xi, \theta) = 0,$$

$$G_{12sd}(\xi) = \begin{cases} -\frac{1}{2}(1-\xi) & \text{for } 0 \leqslant \xi \leqslant 1, \\ 0 & \text{otherwise.} \end{cases} \qquad [66]$$

Then from [54]–[56],

$$\alpha_1^* = 0, \quad \beta_1^* = \frac{\pi}{24} \quad \text{and} \quad \gamma_1^* = \frac{\pi}{48}. \qquad [67]$$

In addition, from [61] and [62], we see that $G_{21sd} = G_{12sd}$ and $\tilde{F}_{21} = -\tilde{F}_{12}$ so that equations [58]–[60] yield

$$\alpha_2^* = 0, \quad \beta_2^* = \frac{\pi}{24} \quad \text{and} \quad \gamma_2^* = \frac{\pi}{48}. \qquad [68]$$

This means that for $V_2 > V_1$ there is, due to a velocity gradient, a flux of spheres 1 (the slower sedimenting particles) towards the upflow and a flux of spheres 2 (the faster sedimenting particles)

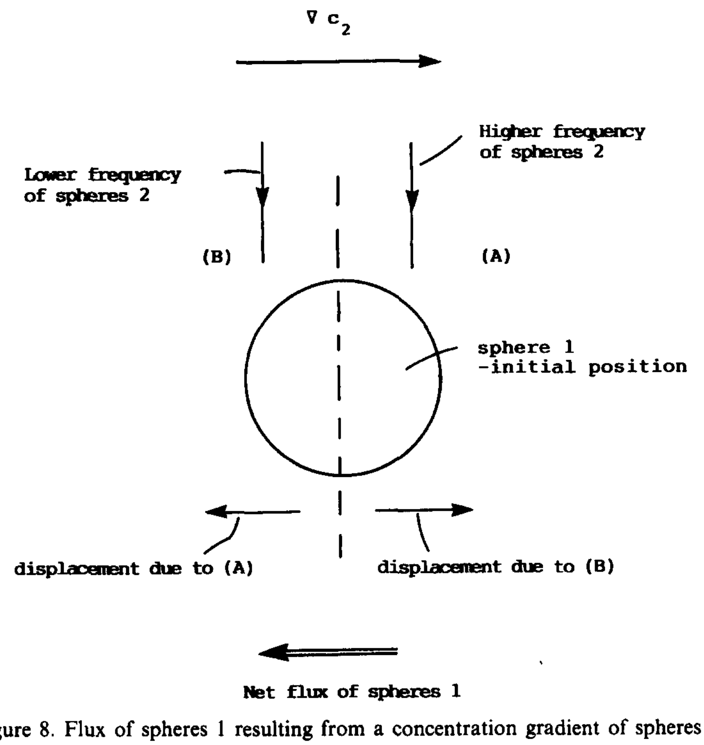


Figure 8. Flux of spheres 1 resulting from a concentration gradient of spheres 2.

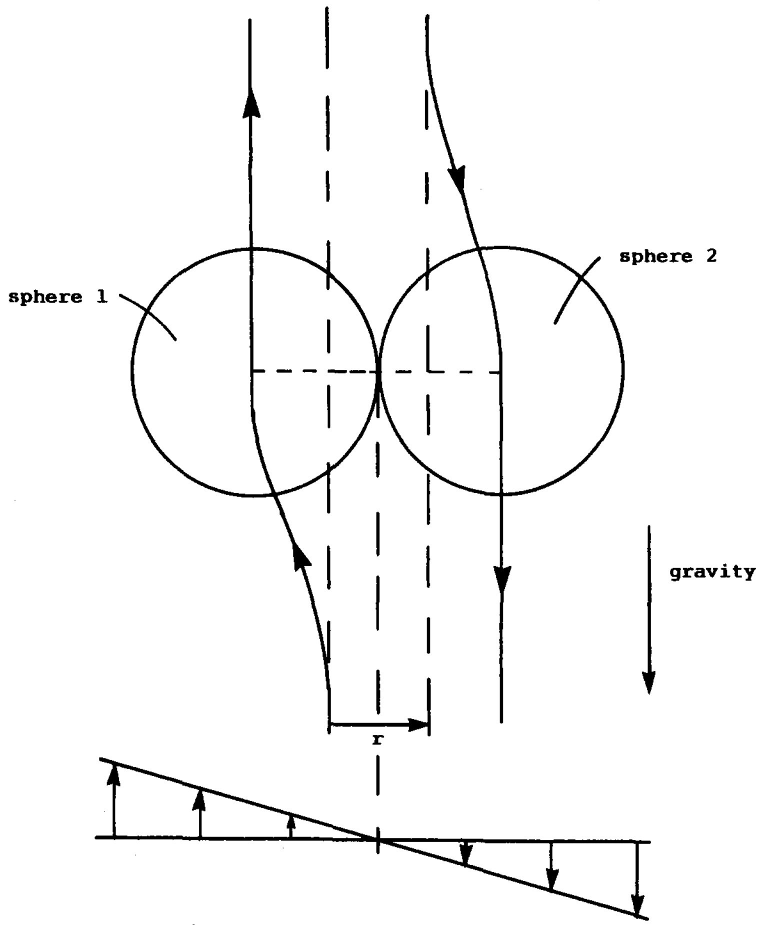


Figure 9. Approximate motion of very rough spheres for the situation where $a_1/a_2 = +1$ and $V_1/V_2 = -1$.

towards the downflow. In addition, due to a concentration gradient of spheres 2, there is a flux of spheres 1 from high to low concentrations of spheres 2.

8. SUSPENSION STABILITY

We consider a bidisperse suspension which is initially at rest and contains a volume concentration c_{10} of spheres 1 and a volume concentration c_{20} of spheres 2 and examine its stability to a small sinusoidal disturbance to the concentrations in the horizontal x_1-direction. As a result of such a disturbance a small sinusoidal flow $U(x_1)$ will be generated. Thus, we take

$$c_1 = c_{10} + \epsilon c_{11}\, e^{\bar{p}t}\, e^{i\bar{k}x},$$

$$c_2 = c_{20} + \epsilon c_{21}\, e^{\bar{p}t}\, e^{i\bar{k}x},$$

$$U = \epsilon U_1\, e^{\bar{p}t}\, e^{i\bar{k}x}, \qquad [69]$$

where $\bar{k}$ is the wavenumber, $\bar{p}$ is the growth rate of the disturbance and ϵ is a small parameter. Substituting these values into [51], [57] and [63] for c_1, c_2 and U and neglecting terms of order ϵ^2 and smaller, we obtain a set of three homogeneous linear algebraic equations for c_{11}, c_{21} and U_1. The condition for a non-trivial solution then yields the characteristic equation

$$A\bar{k}^{-2}\left(\frac{\bar{p}}{|V_2 - V_1|}\right)^2 + (B + C\bar{k}^{-2})\left(\frac{\bar{p}}{|V_2 - V_1|}\right) + (D\bar{k}^2 + E) = 0 \qquad [70]$$

for the value of $\bar{p}$, where

$$A=\frac{1}{k_1k_2c_{10}c_{20}},\quad B=\frac{1}{2}\left(\frac{\gamma_1^*}{k_2c_{10}}+\frac{\gamma_2^*}{k_1c_{20}}\right),$$

$$C=\frac{9}{2(V_2-V_1)}\left[\frac{V_1}{k_2a_1^2}(\alpha_1^*+\beta_1^*)-\frac{V_2}{k_1a_2^2}(\alpha_2^*+\beta_2^*)\right],\quad D=-(\beta_1^*\beta_2^*+\tfrac{1}{2}\beta_1^*\gamma_2^*+\tfrac{1}{2}\beta_2^*\gamma_1^*)$$

and

$$E=\frac{9}{2(V_2-V_1)}\left\{\frac{V_1c_{10}}{a_1^2}[(\alpha_2^*+\beta_2^*)(\beta_1^*+\tfrac{1}{2}\gamma_1^*)+\tfrac{1}{2}\gamma_2^*(\alpha_1^*+\beta_1^*)]\right.$$
$$\left.-\frac{V_2c_{20}}{a_2^2}[(\alpha_1^*+\beta_1^*)(\beta_2^*+\tfrac{1}{2}\gamma_2^*)+\tfrac{1}{2}\gamma_1^*(\alpha_2^*+\beta_2^*)]\right\}. \quad [71]$$

For simplicity the inertia term involving $\partial U/\partial t$ in [63] has been omitted. Solving [70] for the growth rate $\bar{p}$, we obtain

$$\frac{\bar{p}}{|V_2-V_1|}=\frac{\bar{k}^2}{2A}[-B-C\bar{k}^{-2}\pm\{(B^2-4AD)+\bar{k}^{-2}(2BC-4AE)+\bar{k}^{-4}C^2\}^{1/2}], \quad [72]$$

with the suspension being stable for disturbances of the type [69] if the real parts of both values of $\bar{p}$ are negative $\forall\bar{k}>0$. Otherwise, if the real part of either value of $\bar{p}$ is positive for any $\bar{k}>0$, the suspension is unstable. From an examination of the values of $\bar{p}$ given by [72], it is therefore observed that, since A and B are always positive, the suspension is stable iff

$$C>0,\quad D>0\quad\text{and}\quad E>0. \quad [73]$$

For the example given in section 7, for which $a_1/a_2=+1$ and $V_1/V_2=-1$, if the values of $\alpha_1^*, \beta_1^*, \gamma_1^*$ etc. are as given by [67] and [68], then

$$A=\frac{\pi^2}{144a_1^2c_{10}c_{20}},\quad B=\frac{\pi^2}{1152a_1}\left(\frac{1}{c_{10}}+\frac{1}{c_{20}}\right),$$
$$C=-\frac{\pi^2}{64a_1^3},\quad D=-\frac{\pi^2}{384}$$

and

$$E=-\frac{3\pi^2}{512a_1^2}(c_{10}+c_{20}), \quad [74]$$

so that the suspension is unstable with growth rate $\bar{p}$ given by

$$\bar{P}=\tfrac{1}{16}\bar{K}\{-s+18\bar{K}^{-1}+[(s^2+96)+180s\bar{K}^{-1}+324\bar{K}^{-2}]^{1/2}\}, \quad [75]$$

where

$$\bar{P}=\frac{a_1\bar{p}}{c_{10}c_{20}|V_2-V_1|},\quad \bar{K}=\frac{(a_1\bar{k})^2}{\sqrt{c_{10}c_{20}}}\quad\text{and}\quad s=\frac{c_{10}+c_{20}}{\sqrt{c_{10}c_{20}}}. \quad [76]$$

For the bidisperse suspension used in the experiments performed by Weiland *et al.* (1984), for which $a_1=a_2=70\,\mu$m, $V_1=-V_2\simeq 0.06$ cm s^{-1} and $c_{10}\simeq c_{20}\simeq 0.17$, we see that for $\bar{K}<6$ (corresponding to $a_1\bar{k}<1$) the growth rate $\bar{p}$ does not change very much with wavenumber $\bar{k}$, having a value for all disturbance wavelengths of approx. 1.0 s^{-1}. This would seem to agree with the experimental observations that irregular columns were observed at times of order 3 s after the start of sedimention. However, it should be mentioned that for wavelengths $\gtrsim$0.3 cm, for the fluid used ($\nu=0.05$ cm^2 s^{-1}) the results would be modified by the inertia term in [63] which was neglected.

9. CONCLUSION

By taking into account two-particle interactions in which the particles make physical contact with each other, it is possible to examine the sedimentation of particles in a bidisperse suspension consisting of two different species of solid spheres suspended in a fluid. Such particle interactions can result in the particles being moved horizontally as they sediment. By writing continuity equations for each species of particle it is possible to obtain the macroscopic equations [51] and [57] for the volume concentrations c_1 and c_2 of the two species of particles. Only the situation in which c_1, c_2 and the mean vertical velocity U of the suspension vary with one horizontal coordinate x_1 and with time t has been considered. These equations, [51] and [57], are macroscopic non-linear diffusion equations in which a horizontal flux of one species can be generated by either a horizontal concentration gradient of that species, a horizontal concentration gradient of the other species or by a horizontal gradient of the vertical velocity U. In addition, to solve for c_1, c_2 and U one must use the macroscopic vertical momentum equation [63].

The stability of an initially quiescent homogeneous bidisperse suspension was then examined on the basis of these equations, [51], [57] and [63]. The growth rate $\bar{p}$ was determined for a small sinusoidal disturbance (in c_1, c_2 and U) with wavenumber $\bar{k}$ in the horizontal x_1-direction. In this manner, the necessary and sufficient conditions for stability ([73]) were obtained. An approximate analysis was done for a specific example for which it was shown instability should occur. The order of magnitude of the growth rate obtained for this example agree with the experimental results of Weiland *et al.* (1984). In addition, it should be noted that since the growth rate was found to be of order $c_1 c_2$ it would be difficult to observe the instability at the low concentrations for which the present theory is valid. Indeed, an analysis by Batchelor & Janse van Rensburg (1986) of experimental results indicated observed instability only for $c_1 c_2 > 6 \times 10^{-3}$. Should the radius a_1 of one type of particle, species 1, be much larger than that of the other, species 2, i.e. $a_1/a_2 \gg 1$, then the particle of species 2 will merely move along the streamlines around the particle of species 1. Thus, no solid–solid particle contact will occur, resulting in C, D and E in [71] all being zero, implying that the suspension would be neutrally stable. This agrees with the experimental results of Batchelor & Janse van Rensburg (1986) of no observable instability for such a case.

An alternative explanation of the instability of sedimenting bidisperse suspensions was presented by Batchelor & Janse van Rensburg (1986), in which they considered variations of c_1 and c_2 in only the vertical direction and wrote continuity equations for each particle species, assuming certain relationships between the sedimentation velocities and the particle concentrations c_1 and c_2. In this manner they were able to show that the suspension, under certain conditions, could be unstable to small variations of c_1 and c_2 in the vertical direction.

At the present time, it does not appear to be possible to say whether the mechanism considered in the present theory or that considered by Batchelor & Janse van Rensburg (or possibly some other mechanism) is responsible for the initial instability of a bidisperse suspension. The experiments of Weiland *et al.* (1984) indicate the formation of wide vertical streaming columns, whereas those of Batchelor & Janse van Rensburg (1986) indicate a fine-scale grainy appearance immediately after the start of sedimentation. The latter grainy appearance would then slowly develop into either "blobs" or into vertical streaming columns, depending on the system being examined. Thus it would appear, at least in some situations, that the initial instability is of a three-dimensional nature. However, the present theory does present mechanisms [particularly mechanism (ii) described in section 7] which can result in the maintenance and amplification of the vertical streaming columns once they are formed, despite diffusive effects [such as mechanism (iv) described in section 7].

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APPENDIX

For the close collisions of spheres with non-hydrodynamic effects (such as solid–solid contact) considered here, the time between collisons is of order (a/cV) whilst the displacement of spheres due to such a collision is of order a. (Here a is a characteristic particle size, V is a characteristic sedimentation velocity of one particle relative to another and c is a characteristic particle concentration.) This results in a horizontal velocity V_h and diffusivity D of the particles, where

$$V_h \simeq cV \quad \text{and} \quad D \simeq caV. \qquad [A.1]$$

However, spheres can also be transported horizontally by three-particle hydrodynamic collisions. Such encounters of three particles in which they are of distances of order a apart occur for a given particle only at times of order (a/c^2V) and result in a displacement of order a. Hence they result in values of V_h and D of order

$$v_h \simeq c^2V \quad \text{and} \quad D \simeq c^2aV. \qquad [A.2]$$

If the three-particle interaction is such that two of the particles (A and B say) are at a distance of order a apart whilst the third particle (C say) is at a distance of order $l = c^{-1/3}a$ from particles A and B (l being the mean interparticle distance in the suspension), then A and B induce a velocity at C (and vice versa) of order $Va/l = c^{1/3}V$ for a time of order a/V (the interaction time for particles A and B). Thus, displacements of order $c^{1/3}a$ are produced in times of order (a/cV) (the time between close interactions for two particles). Hence, such interactions result in values of v_h and D of order

$$v_h \simeq c^{4/3}V \quad \text{and} \quad D \simeq c^{5/3}aV. \qquad [A.3]$$

Should the three-particle interactions be such that particles A, B and C are at a distance of order l apart, then particle B experiences an additional shear flow of order (Va/l^2) due to the disturbance flow of particle A. Thus at sphere C, this produces an additional disturbance velocity due to particle B of order $(a^4V/l^4) = c^{4/3}V$. Thus, in the interaction time of order $(l/V) = c^{-1/3}a/V$, particle B moves a distance ca. Hence, such interactions results in values of v_h and D of order

$$v_h \simeq c^{4/3}V \quad \text{and} \quad D \simeq c^{7/3}aV. \qquad [A.4]$$

We note that the values of v_h and D given by [A.2]–[A.4] for three-particle hydrodynamic interactions are all much smaller (for $c \ll 1$) than the values given by [A.1] for two-particle interactions with non-hydrodynamic effects and, as such, it is valid at lowest order to neglect the effects of three-particle hydrodynamic interactions.