# Break-up of a falling drop containing dispersed particles

By J. M. NITSCHE<sup>1</sup> AND G. K. BATCHELOR<sup>2</sup>

<sup>1</sup>Department of Chemical Engineering, State University of New York at Buffalo, Buffalo, NY 14260, USA

<sup>2</sup>Department of Applied Mathematics and Theoretical Physics, University of Cambridge, Silver Street, Cambridge CB3 9EW, UK

(Received 25 August 1996 and in revised form 22 January 1997)

The general purpose of this paper is to investigate some consequences of the randomness of the velocities of interacting rigid particles falling under gravity through viscous fluid at small Reynolds number. Random velocities often imply diffusive transport of the particles, but particle diffusion of the conventional kind exists only when the length characteristic of the diffusion process is small compared with the distance over which the particle concentration is effectively uniform. When this condition is not satisfied, some alternative analytical description of the dispersion process is needed. Here we suppose that a dilute dispersion of sedimenting particles is bounded externally by pure fluid and enquire about the rate at which particles make outward random crossings of the (imaginary) boundary. If the particles are initially distributed with uniform concentration within a spherical boundary, we gain the convenience of approximately steady conditions with a velocity distribution like that in a falling spherical drop of pure liquid. However, randomness of the particle velocities causes some particles to make an outward crossing of the spherical boundary and to be carried round the boundary and thence downstream in a vertical 'tail'. This is the nature of break-up of a falling cloud of particles.

A numerical simulation of the motion of a number of interacting particles (maximum 320) assumed to act as Stokeslets confirms the validity of the above picture of the way in which particles leak away from a spherical cluster of particles. A dimensionally correct empirical relation for the rate at which particles are lost from the cluster involves a constant which is indeed found to depend only weakly on the various parameters occurring in the numerical simulation. According to this relation the rate at which particles are lost from the blob is proportional to the fall speed of an isolated particle and to the area of the blob boundary. Some photographs of a leaking tail of particles in figure 5 also provide support for the qualitative picture.

#### 1. Introduction

It is a feature of flowing dispersions of particles suspended in viscous fluid (normally a liquid) that the particles make random displacements arising from hydrodynamic interactions with their neighbours, and that these displacements generate stochastic particle trajectories. This process of hydrodynamic dispersion has an origin entirely distinct from that of the Brownian diffusion of very small particles (which respond dynamically to collisions with molecules of the surrounding solvent), and it occurs

with comparatively large particles for which Brownian agitation may be negligible as a force causing transport. The investigation of *hydrodynamic* dispersion of interacting spherical particles normal to the streamlines of an annular Couette flow by Eckstein, Bailey & Shapiro (1977) appears to have been the first in an essentially new field. Since then considerable experimental and theoretical work has addressed hydrodynamic diffusion of particles undergoing bulk shear flow or sedimentation. (See Davis 1996 for a report on the developments presented at an international symposium in 1995.)

Many of these studies have been restricted to dispersions in which the particle concentration is uniform or is a slowly varying function of position. In these circumstances it may be possible to define a hydrodynamic diffusivity of the particles as the particle flux divided by the concentration gradient. In some other cases, however, there is a jump in the value of the particle concentration across an interface. In these circumstances a diffusivity defined as the ratio of the particle flux to the concentration gradient does not have physical meaning. Transfer of particles due to velocity fluctuations still occurs, but is not describable in terms of a diffusivity, just as one would not expect the continuum equations for diffusion of a gas to be applicable when – as in the interior of a shock wave for instance – the characteristic length scale of the diffusive process is not small compared with distances over which the particle concentration is effectively uniform.

We shall present a numerical investigation of hydrodynamic dispersion in a system containing an interface which separates a random dispersion of prescribed particle concentration on one side from clear fluid on the other side. Specifically, we consider the motion under gravity of particles within a blob (a convenient term for a finite volume of a dispersion of particles in liquid) comprising a large number N of particles initially distributed randomly in liquid with uniform mean concentration within a prescribed closed surface, and inquire as to its subsequent time evolution. The particles will tend to spread out from each other, and questions of interest are therefore: do particles leave the blob, and if so how, and what is the lifetime of the blob as a cohesive entity? A spherical blob shape is especially well suited to a study of random particle migration across interfaces because the gravity-driven flow system maintains essentially constant form. Thus, the migration process can be observed without the complication of significant deformation of the blob as a whole. As noted above, it is not possible to specify the flux of particles across such an interface in terms of a particle diffusivity of the conventional kind. Some alternative analytical description of the dispersion process at the interface is required.

Insofar as the blob can be regarded as an effective continuum having a density higher than that of the surrounding clear fluid, the flow system under consideration is closely related to the fall of a drop of heavy fluid through an infinite expanse of lighter fluid, for which the velocity field is given – in the case of a spherical blob – by well-known formulae (Batchelor 1967) attributed to Hadamard and Rybczyński (designated here as H-R). A spherical blob containing particles possesses the additional physical feature that the suspended particles, which contribute excess mass to the blob, fall relative to the fluid in which they are immersed. A generalization of the H-R solution appropriate to this circumstance has been given in another context by Batchelor (1974), who found that a low-Reynolds-number spherical sedimenting drop may carry with it a circulating 'halo' of clear fluid, and that the compound structure as a whole may fall without change of form.

The sequence of problems considered may thus be described as follows: (i) the H-R solution refers to a spherical drop of viscous fluid falling through a second viscous fluid of smaller density; (ii) the Batchelor (1974) solution refers to a spherical drop

containing small dispersed particles assumed to fall with uniform speed relative to the fluid locally; and (iii) in the problem studied herein the particle velocities vary randomly as a consequence of particle interactions, and the particles disperse.

Our consideration of the dispersion of particles in a blob also has elements in common with some previous experimental studies of the break-up of particulate blobs in simple shear and extensional flows (Kao & Mason 1975; Powell & Mason 1982). Despite the differences between externally imposed shear and gravity-driven flow, we shall find that the rates at which particles leave the blob are correlated by similar mass-transfer expressions in these two cases.

Our paper is organized as follows. Section 2 formulates the equations governing the particle and fluid motions, and identifies restrictions on the circumstances to which these equations apply. The equations of motion are solved numerically in §3 to determine the time evolution of the blob, and, in particular, to quantify the outward flux of particles across its boundary. An empirical correlation for this flux is introduced and discussed in §4. Section 5 describes some experimental observations related to the calculations.

# 2. Governing equations for the particles and fluid

We suppose that initially N spherical particles of radius a and density  $\rho_p$  are distributed randomly and with uniform mean concentration within a spherical region of radius R in an infinite expanse of fluid having density  $\rho$  and viscosity  $\mu$ . The particles move under the influence of gravity and the viscous resistance exerted by the fluid, and our immediate objective is to formulate the equations governing their motions

The terminal settling velocity of an isolated particle will be denoted by  $u_0$ , and we shall suppose that the particle Reynolds number  $Re = 2a\rho u_0/\mu$  is small, in which case  $u_0$  has the Stokes value  $2a^2(\rho_p - \rho)g/9\mu$ . We shall also suppose that the Stokes number  $S = \tau u_0/R$  (representing the ratio of the viscous relaxation time  $\tau = m/6\pi\mu a$  of a particle to the time needed for it to fall the relevant distance R) is small, so that the effects of particle inertia are negligible. Under these circumstances the equation of motion for particle i takes the form

$$dx_i/dt = v_i, \quad 0 = -(m - m_0)gk + h_i + f_i, \quad (2.1)$$

where  $m = 4\pi a^3 \rho_p/3$  denotes the mass of a particle,  $m_0$  the mass of fluid displaced by a particle, k the unit vector pointing upwards, g the gravitational acceleration,  $h_i$  the total hydrodynamic force exerted by all particles k ( $k \neq i$ ) upon particle i, and  $f_i$  any additional force exerted upon particle i. The hydrodynamic force  $h_i$  is given by the solution for Stokes flow in a system with i0 torque-free rigid spherical boundaries centred at the points i1 moving with velocities i2. The present flow system is unusual in the respect that, even though there is no imposed ambient flow, the forces i2 moving i3 collectively generate a systematic flow (namely, a toroidal circulation essentially identical to that in a falling drop – see below). The Reynolds number of this collective motion is also assumed to be small. The additional force i3 is zero for particles moving solely under the influence of gravity and the viscous resistance exerted by the fluid.

Exact representation of the particle interactions would require the solution of the N-body Stokes-flow problem, which is inaccessible analytically and time-consuming to calculate accurately if N is large (cf. Kim & Karrila 1991). Our primary focus is on dilute dispersions, for which the typical interparticle spacing  $|x_i - x_j|$  is much

larger than a. In this limit it is appropriate to approximate the fluid velocity as a superposition of Stokeslet disturbances. The hydrodynamic forces approximated on this basis and denoted by  $h'_i$  are given by expressions of the form

$$\mathbf{h}_i' = -6\pi \mu a(\mathbf{v}_i - \mathbf{u}_i),\tag{2.2}$$

where  $u_i$  denotes the ambient fluid velocity evaluated at the position of particle *i*. The  $u_i$  are calculable from the equations

$$\mathbf{u}_i = \sum_{j \neq i} \mathbf{w}(\mathbf{x}_i; \mathbf{x}_j) \cdot 6\pi \mu a(\mathbf{v}_j - \mathbf{u}_j) \quad (i = 1, \dots N),$$
 (2.3)

in which  $\mathbf{w}(\mathbf{x}; \mathbf{x}_0)$  denotes the Stokeslet tensor with singularity at  $\mathbf{x}_0$ . Equation (2.3) expresses the fact that each ambient velocity  $\mathbf{u}_i$  is a superposition of point-force disturbances  $\mathbf{u}_i$  arising from the gravitational slip of particles relative to the fluid. It can be shown that equations (2.2) and (2.3) are valid approximately in the sense that for prescribed particle velocities  $\mathbf{v}_i$ , the forces  $\mathbf{h}_i'$  approximate the true forces  $\mathbf{h}_i$  correctly up to second order in  $a/|\mathbf{x}_i - \mathbf{x}_j|$ . In general, for prescribed  $\mathbf{v}_i$ , equation (2.3) represents a system of linear equations for the  $\mathbf{u}_i$  requiring simultaneous solution. Under the present circumstances, however, the local slip velocities  $\mathbf{v}_i - \mathbf{u}_i$  are known a priori from the force balance in (2.1).

Our truncation of the multipole expansion for the fluid velocity after the first order (equations (2.2) and (2.3)) represents a simple approximation that does not prevent particles from overlapping. Overlaps may produce an unrealistically large settling speed of a pair of particles, because if the centre-to-centre distance happens to be very small then, according to equation (2.3), each particle induces a large ambient fluid velocity at the position of the other. This phenomenon may, in turn, lead to unrealistically large velocity fluctuations.

Overlaps can, in principle, be prevented from occurring in the mathematical model by pairwise consideration of the particle interactions in the following way. For each particle i the nearest neighbour (particle k, say) is identified, and the ambient velocity  $\hat{u}_i$  incident upon both i and k is computed by excluding j = k from the summation in (2.3):

$$\hat{\boldsymbol{u}}_i = \sum_{i \neq i, k} \boldsymbol{w}(\boldsymbol{x}_i; \boldsymbol{x}_j) \cdot 6\pi \mu a(\boldsymbol{v}_j - \boldsymbol{u}_j). \tag{2.4}$$

Thereafter, an improved approximation  $\mathbf{h}_{i}^{"}$  to the hydrodynamic force  $\mathbf{h}_{i}$  is calculable from the equation

$$\boldsymbol{h}_{i}^{"} = -\mu \boldsymbol{K}_{ii} \cdot (\boldsymbol{v}_{i} - \hat{\boldsymbol{u}}_{i}) - \mu \boldsymbol{K}_{ik} \cdot (\boldsymbol{v}_{k} - \hat{\boldsymbol{u}}_{i}), \tag{2.5}$$

in which  $K_{ii}$  and  $K_{ik}$  denote the well-known two-sphere resistance dyadics giving the force on particle i due to translation of particle i and particle k, respectively (see Kim & Karrila 1991). Equations (2.4) and (2.5) are consistent with the Stokeslet approximation in the sense that, if particle i is far from all other particles ( $|x_i-x_j|\gg a$  for all  $j\neq i$ ), then  $h_i''$  reduces to  $h_i'$ . Equation (2.5) is similar to the pairwise addition of near-field hydrodynamic forces in 'Stokesian dynamics' simulations (see e.g. Brady & Bossis 1988). Unfortunately, it seems in practice that prohibitively small time steps are needed to prevent overlaps by this method (cf. Bossis & Brady 1984; Durlofsky & Brady 1989).

We have therefore resorted to the expedient of staying with the simple hydrodynamic approximation embodied in equations (2.2) and (2.3), and introducing in equation (2.1) an artificial short-range repulsive force acting between all pairs of

$$\mathbf{f}_{i} = \sum_{j \neq i} C_{1} 6\pi \mu a u_{0} \frac{\mathbf{x}_{i} - \mathbf{x}_{j}}{|\mathbf{x}_{i} - \mathbf{x}_{j}|} \exp\left(-\frac{|\mathbf{x}_{i} - \mathbf{x}_{j}| - 2a}{C_{2}a}\right). \tag{2.6}$$

Here  $C_1$  and  $C_2$  are parameters determining the strength and range of the repulsive force, and are usually set to the reasonable but arbitrary numerical values of 10 and 0.1, respectively. Introduction of an extra repulsive force to prevent overlaps (cf. e.g. Da Cunha 1994) is not unrealistic because forces acting between particles in nature and in laboratory practice are often repulsive.

We emphasize that our purpose here is not to perform detailed calculations of particle interactions. Rather, we aim to explore the physical processes giving rise to dispersion and its consequences. Our ultimate conclusion (see below) that Stokeslet interactions suffice for the production of random particle migration across an interface is significant, because it represents an identification of the minimum physics needed to reproduce this phenomenon. Our use of equations (2.2)–(2.3) is therefore not merely expedient, but also serves to reveal this important conclusion.

It is advantageous now to introduce the dimensionless variables

$$X_i = R^{-1} x_i, \quad T = t u_0 / R, \quad V_i = u_0^{-1} v_i, \quad U_i = u_0^{-1} u_i,$$
 (2.7)

in terms of which equations (2.1), (2.2), (2.3) and (2.6) become

$$dX_i/dT = V_i, \qquad 0 = -\mathbf{k} - (V_i - U_i) + \mathbf{F}_i, \tag{2.8}$$

$$U_i = \sum_{i \neq i} \mathbf{W}(X_i; X_j) \cdot 6\pi \epsilon(V_j - U_j), \tag{2.9}$$

with  $\epsilon = a/R$ ,

$$\mathbf{W}(X;X_0) = \frac{1}{8\pi} \left[ \frac{1}{|X - X_0|} + \frac{(X - X_0)(X - X_0)}{|X - X_0|^3} \right]$$
(2.10)

and

$$F_{i} = \sum_{j \neq i} C_{1} \frac{X_{i} - X_{j}}{|X_{i} - X_{j}|} \exp\left(-\frac{|X_{i} - X_{j}|\epsilon^{-1} - 2}{C_{2}}\right).$$
(2.11)

#### 3. Time evolution of the blob

For any given number of particles N, equations (2.8)–(2.11) constitute a dimensionless system of 3N coupled first-order nonlinear ODEs, of which the solution is approximated numerically by fourth-order Runge–Kutta integration. Simulations are performed with blobs typically comprising N=80, 160 or 320 particles, and their time evolution is usually tracked over the time interval  $0 \le T \le 10$ . The initial distribution of particle centres is random with uniform probability throughout a sphere of unit radius centred at the origin, with the provision that particle overlaps are excluded. The initial particle volume fraction is kept small (4% or less) so that the simple Stokeslet approximation for hydrodynamic interactions is reasonable. A spot check with 160 particles of relative size  $\epsilon=0.05$  indicates that halving the time step of the numerical integration  $\Delta T$  from 0.001 to 0.0005 leads to changes in the computed individual particle positions smaller than about 0.02 at time T=10. We therefore use a time step  $\Delta T=0.001$  and regard this as yielding adequate convergence of the

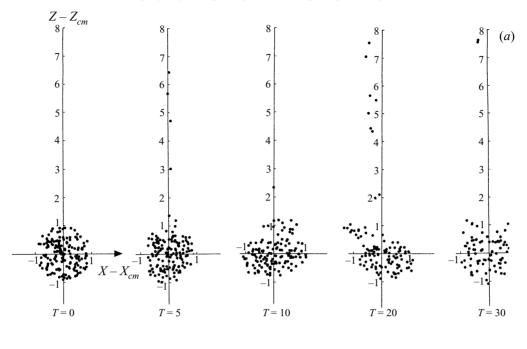
particle paths. Possible random errors resulting from use of a larger time step seem unlikely to invalidate the computed statistical properties of the blob, even if they do affect the individual trajectories. We therefore refrain from performing detailed checks for convergence on all the computed particle paths. (It is worth noting that the present system appears to be chaotic in the sense that any errors in the computed particle positions, no matter how slight, seem to be magnified exponentially with time. Thus, owing to computer round-off and finite accuracy of the numerical integration, it would be impossible in practice to determine the individual particle positions at arbitrarily long times.) Computed position coordinates of all the particles are written to a file at regular intervals in T (usually 0.5, or every 500 time steps), which is analysed later to extract or compile any desired information. Spot checks indicate that the particles do not overlap. Since the blob maintains its cohesion and size (see below), a good estimate of the position  $X_{cm}$  of the blob's centre of mass at each time T is provided by averaging the position coordinates of all particles having heights not exceeding that of the lowest particle Z-coordinate by more than a fixed number; this number should be somewhat greater than the initial diameter (2) to allow for small random deviations from the spherical shape, and is usually taken to be 3. Particles are considered to have leaked away from the blob if  $|X_i - X_{cm}| > 2$ . The overall conclusions of the analysis are insensitive to the precise numerical criterion according to which a particle is classified as belonging to the blob or having leaked away, and our criterion seems to be reasonable.

Figure 1 illustrates the time evolution of a blob as a sequence of 'snapshots' in which the particle positions in three-dimensional space are projected onto a vertical plane, arbitrarily taken to be the (X,Z)-plane. It may be observed from these calculations, and other calculations not exhibited, that: (i) a sedimenting blob slowly loses particles by shedding them sporadically along a vertical line emanating from the rear; (ii) particles remaining in the blob are distributed randomly and uniformly throughout its interior at all times; and (iii) the blob remains roughly spherical, with radius roughly constant at its initial value of unity. It is a conspicuous feature of the blob that it remains cohesive, maintaining a rather sharp and clearly defined boundary between its particle-filled interior and the clear fluid outside. This boundary is not imposed in the calculation, but rather represents an observed outcome of the dynamical simulation. The volume fraction  $\varphi = N_b \epsilon^3$  of particles in the interior gradually decreases owing primarily to a decrease in the number of particles  $(N_b)$ and not to any significant increase in its size. In late stages of the settling process, when few particles remain in the blob, the blob boundary becomes ragged, exhibiting significant departures from the spherical shape.

Insofar as the discrete particles comprising the blob approximate a continuous spherical distribution of excess mass, the fall speed  $V_b$  of the blob can be approximated using the H-R expression for a spherical drop of fluid sedimenting through lighter fluid (see Batchelor 1967, §4.9), namely

$$\frac{(\overline{\rho}-\rho)gR^2}{3\mu}\frac{\mu+\overline{\mu}}{\mu+\frac{3}{2}\overline{\mu}},$$

in which  $\rho$  and  $\mu$  respectively denote the density and viscosity of the clear fluid outside the blob, and  $\overline{\rho}$  and  $\overline{\mu}$  denote the corresponding properties of the dilute suspension inside. The excess density  $\overline{\rho} - \rho$  is just  $\varphi(\rho_p - \rho)$ , where  $\varphi$  is the particle volume fraction  $N_b \epsilon^3$ . The effect of the particles on the interior viscosity is small ( $\overline{\mu} \approx \mu$ ) because  $\varphi$  is assumed to be small. The blob fall speed actually exceeds the H-R value



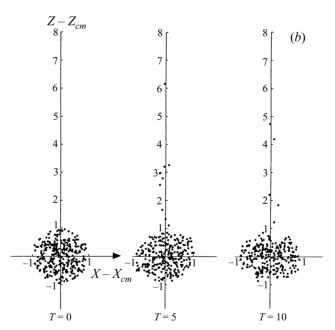


FIGURE 1. Sequences of snapshots showing the computed time evolution of falling blobs. Particle positions are projected onto the (X,Z)-plane, and are referred to axes moving with the computed centre-of-mass position of the blob  $(X_{cm})$ . The dots representing the particles are drawn to scale. The initial particle volume fraction,  $\varphi$  at T=0, is 0.02 in both cases. (a) Blob initially contains 160 particles of size  $\epsilon=a/R=0.05$ . (b) Blob initially contains 320 particles of size  $\epsilon=a/R=0.0397$ .

Transfer coeff., $K$	0.077 0.095 0.091 0.077	0.065 0.086 0.090	0.105 0.083 0.076
Initial rate of leakage, $-dN_b/dT$	$1.7 \pm 0.1$ $2.1 \pm 0.5$ $3.9 \pm 0.6$ $6.4$	$1.2 \pm 0.3$ $3.0 \pm 0.6$ 6.1	$1.6 \pm 0.4$ $2.4 \pm 0.4$ $4.2$
Prediction of $V_b/u_0$ from (3.1)	4.02 8.62 8.62 13.1 20.2	3.40 7.05 10.6 16.2	2.91 5.80 8.62 13.1
Initial fall speed, $V_b/u_0$ at $T=0$	3.64 ± 0.12 8.26 ± 0.07 8.27 ± 0.07 12.8 ± 0.3 20.1	$3.14 \pm 0.11$ $6.79 \pm 0.11$ $10.5 \pm 0.1$ 16.3	$2.70 \pm 0.09$ $5.64 \pm 0.10$ $8.55 \pm 0.10$ $13.2$
No. of simula-tions	ν ν ν κ <del>-</del> -	v v v u	ν ν π <del>-</del> -
Particle size, $\epsilon = a/R$	0.126 0.0794 0.0794 0.063 0.05	0.1 0.063 0.05 0.0397	0.0794 0.05 0.0397 0.0315
Mean particle spacing, d/R	0.594 0.374 0.374 0.297 0.236	0.594 0.374 0.297 0.236	0.594 0.374 0.297 0.236
No. of particles, $N$	20 80 80 160 320	20 80 160 320	20 80 160 320
Init. particle vol. fraction, $\varphi$ at $T=0$	0.04 *	0.02	0.01

leakage from blobs  $(-dN_b/dT)$  estimated from the simulations in the manner described in the text. The values given for  $V_b/u_0$  and  $-dN_b/dT$  represent the mean and standard deviation of simulation results for several realizations of the random initial distribution of particles. Also listed are predictions of  $V_b/u_0$  from equation (3.1), and values of the transfer coefficient K determined such that equation (4.1) reproduces the observed rates of particle leakage. The parameter  $C_2$  (which determines the range of the repulsive force preventing particle overlaps) is set to 0.1 in all cases except that distinguished by the asterisk (\*), for which  $C_2 = 0.05$ . TABLE 1. Initial fall speeds of blobs  $(V_b/u_0 \text{ at } T=0)$  observed in the numerical simulations, and initial rates of particle

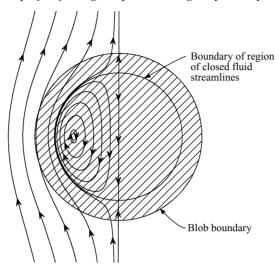


FIGURE 2. Representation of a sedimenting blob as a continuous uniform spherical distribution of excess mass that slips relative to the ambient fluid owing to gravity. The figure shows fluid streamlines referred to axes moving downward with the blob at speed  $V_b$ , and corresponds to the parameter values  $N_b = 160$  and  $\epsilon = 0.05$ . The region of closed fluid streamlines is bounded by a spherical surface lying inside the blob boundary.

because the excess density (in the form of finite-size particles) slips relative to the ambient fluid, with speed roughly equal to the isolated-particle settling velocity  $u_0$  ignoring the small  $(O(\varphi))$  hindered settling effect (cf. Batchelor 1972). We therefore obtain the approximate result

$$\frac{V_b}{u_0} \approx \frac{6}{5} N_b \epsilon + 1,\tag{3.1}$$

in which the second (slip) term has only a minor effect if  $N_b$  is at all large. Table 1 compares values of  $V_b/u_0$  predicted by this equation with corresponding values of the initial blob fall speed determined from the simulations by finite-difference approximation of the derivative  $\mathrm{d}X_{cm}/\mathrm{d}T$ . The agreement is good, even for relatively small N (i.e. even when there is a relatively coarse discretization of the excess mass). Gravitational slip of the excess mass of the blob alters the streamline pattern of the fluid from that for a spherical fluid drop, shifting the boundary of the region of closed fluid streamlines to a spherical surface lying a finite distance inside the boundary of the blob, as shown schematically in figure 2. This compound structure is similar to that arising in a consideration of low-Reynolds-number bubbles in fluidized beds (Batchelor 1974).

Figure 3 shows representative curves giving the number  $N-N_b$  of particles that have leaked away from the blob as a function of time T, at intervals of 0.5 in T from T=0 to 10. There is a considerable variation among simulations starting from different realizations of the initial particle distribution, which decreases somewhat as the number N of particles increases. For each simulation the initial rate of leakage may be estimated as the slope of a line fitted to the corresponding curve by the method of least squares. The time interval  $0 \le T \le 10$  is short enough for the fractional change in  $N_b$  (or  $\varphi$ ) to be relatively small. Values of  $-\mathrm{d}N_b/\mathrm{d}T$ , determined in this way and averaged over several simulations, are included in table 1 for various combinations of the initial particle volume fraction and number of particles. A

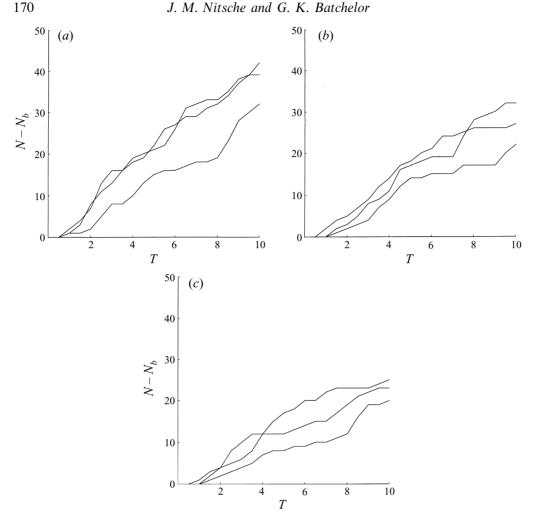


FIGURE 3. The number of particles that have leaked from the blob,  $N-N_b$ , as a function of dimensionless time T. Curves represent numerical simulations starting from three different realizations of the initial particle distribution. N=160 in all cases. (a)  $\epsilon=0.063$  ( $\varphi=0.04$  at T = 0). (b)  $\epsilon = 0.05$  ( $\varphi = 0.02$  at T = 0). (c)  $\epsilon = 0.0397$  ( $\varphi = 0.01$  at T = 0).

comparison of the second and third lines of the table suggests that the results are rather insensitive to the numerical value of the parameter  $C_2$  (which determines the range of the artificial repulsive force preventing particle overlaps) provided it is small.

Figure 4, which shows the trajectories of two particular particles in a representative simulation, illuminates the mechanism by which the particles leak away from the blob. Circulatory motion, expected in view of the well-known toroidal flow inside a sedimenting spherical drop of liquid, is clearly evident. Superposed on this motion is a sequence of random displacements arising from hydrodynamic interactions. Sooner or later these random displacements lead to an instantaneous position just outside the blob, where the streamlines sweeping it round the blob surface are no longer closed (as they are inside), but rather extend upwards to infinity. Once the particle falls a little behind the faster moving blob, it never catches up again, and so is lost.

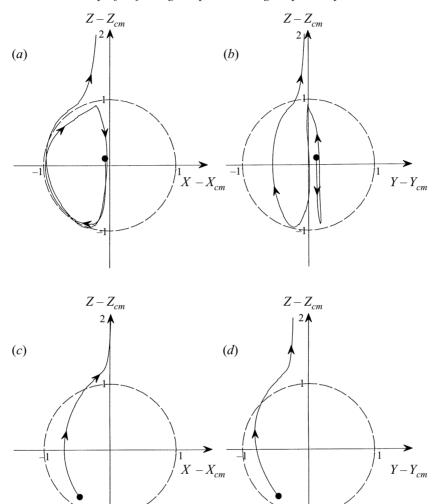


FIGURE 4. Trajectories of two particular particles from a representative simulation with N=160 and  $\epsilon=0.05$  (so that  $\phi=0.02$  at T=0) illustrating transfer across the blob boundary: (a) and (b) represent two orthogonal views of the trajectory of one particle, and (c) and (d) those of another particle.

# 4. A correlation for the rate of particle leakage from the blob

Having now a picture of the mechanism of the break-up of a blob, we consider whether the data provided by the numerical simulations can be represented analytically. We seek in particular an analytical relation for the rate at which particles are lost from a blob. It is evident, as a beginning, that the velocity of fall of a blob  $(V_b)$  is a general rate-determining factor, in which case

$$\frac{1}{V_b} \frac{\mathrm{d} N_b}{\mathrm{d} t}$$

is a quantity with the dimensions of reciprocal length ( $N_b$  being dimensionless) and only a weak dependence on time. And if, as we may expect, the unsteadiness of the velocity of particles near the interface is local in character, a relevant unit of length is the mean particle spacing  $d = (4\pi/3N_b)^{1/3}R$ . We are led therefore to postulate that

$$-\frac{\mathrm{d}N_b}{\mathrm{d}t} = K\frac{V_b}{d} \tag{4.1}$$

where K represents a numerical coefficient to be determined. The last column in table 1 lists the values of K needed to reproduce the rates of particle leakage observed in the simulations. It appears that this coefficient is roughly constant over the significant ranges of particle volume fraction  $\varphi$  and number of particles N covered by the table, suggesting that our assertion is sound. We conclude that the break-up process can be quantified to reasonable accuracy by equation (4.1) with  $K \approx 0.08$ , although this value is subject to some uncertainty.

Since  $V_b$  is dominated by the first (H-R) term in equation (3.1), the right-hand side of equation (4.1) is well approximated by substituting  $\frac{6}{5}N_b\epsilon u_0$  for  $V_b$ . The rate of leakage of particles may then be rewritten in an alternative way, namely

$$-dN_b/dt \approx \text{const.} \, u_0 \, R^2 \, a/d^4, \tag{4.2}$$

which is physically revealing. We have supposed that the leakage of particles is local in character. If this is indeed so, then the rate of transfer across a portion of the blob surface should be proportional to the area of that portion, making the total rate of leakage proportional to the blob surface area  $4\pi R^2$ . Moreover, because it derives from random velocity fluctuations, the rate of leakage of particles should scale with the magnitude of these fluctuations, which is  $O(u_0)$ . Both of these expectations are reflected in equation (4.2).

Our rate expression has the feature that if  $N_b$  increases and  $\epsilon$  decreases in such a way as to keep the particle volume fraction  $\varphi$  constant, then  $\mathrm{d}\varphi/\mathrm{d}t$  (as opposed to  $\mathrm{d}N_b/\mathrm{d}t$ ) tends to zero (as  $N_b^{-2/3}$ ). In other words, the rate of change of excess mass vanishes as the excess mass is discretized more and more finely.

We have already noted the correspondence between a sedimenting cloud of heavy particles and a sedimenting drop of heavy fluid (with density  $\overline{\rho} = \varphi \rho_p + (1 - \varphi) \rho$ , viscosity  $\overline{\mu} \approx \mu$ , and vanishing interfacial tension). A spherical fluid drop has no tendency to deform and falls with a constant speed given by the H-R formula. The fact that the cloud of particles has small random deviations from radial symmetry suggests a connection with fluid drops that start out with slightly non-spherical shapes. Such drops (with vanishing interfacial tension) often develop tails (see e.g. Stone 1994, figure 10). It appears to be an inherent feature of the flow engendered by sedimentation of excess mass that the drop may become elongated at the rear. In comparing the particulate cloud with a fluid drop it is important to take note of essential differences in the mechanisms by which excess mass is left behind in the form of a tail. A liquid drop has constant density, so the loss of mass is necessarily accompanied by a decrease in its size; this loss arises from a non-random flow process. In contrast, we observe that a cloud of particles maintains roughly constant size, so the loss of mass is accompanied by a decrease in its density; this loss arises from random outgoing transport across the interface. Both mechanisms might be operative to a significant extent in the evolution of a cloud comprising very many small particles, because the rate of change of excess mass arising from the latter random process tends to zero as the excess mass is discretized more and more finely.

Kao & Mason (1975) and Powell & Mason (1982) have studied experimentally the

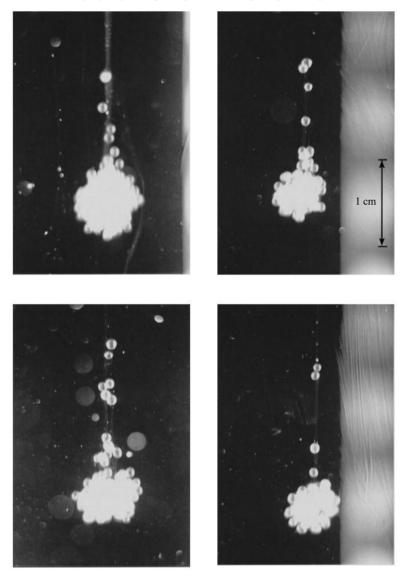


FIGURE 5. Representative photographs of blobs containing glass particles sedimenting through glycerine.

break-up of highly concentrated initially spherical aggregates of particles in some twodimensional linear ambient flows, including the cases of pure elongation and simple shear. The aggregates are observed to remain roughly spherical for a time, with some accumulation of particles at opposite sides of the sphere, and later to become elongated by mechanisms that differ between the two flows. While the aggregate remains spherical, the rate at which particles leak away is correlated roughly by an expression which can be written in our notation in the form

$$-dN_b/dt = \text{const.} (\gamma a) R^2 a^{-3}, \tag{4.3}$$

where  $\gamma$  denotes the shear rate, R the aggregate radius and a the particle radius. Given the high concentrations of particles employed, it is not surprising that the

average particle spacing d = O(a) is not distinguishable from a as a parameter upon which the rate of leakage might depend. The factors  $a^{-3}$  and  $a/d^4$  respectively appearing in equations (4.2) and (4.3) may therefore be considered to be identical. It is then noteworthy that these two expressions have the same form, both indicating that  $-\mathrm{d}N_b/\mathrm{d}t$  is proportional to the blob surface area and to velocities ( $u_0$  and  $\gamma a$ , respectively) having the order of magnitude of particle velocity fluctuations. Mason and coworkers regarded it as a basic fact that the rate at which particles leak away from a blob is proportional to its surface area. It is worth noting that these authors observe their aggregates to become smaller as they lose particles, in contrast to our sedimenting blob, for which the size remains roughly constant and the particle volume fraction decreases.

## 5. Some photographs of a falling blob

A description of the intriguing shedding of particles from a falling blob has emerged from the numerical experiments. We have sought to confirm this description by appealing to some real (physical) experiments. To this end, we employed a small Plexiglas-walled tank (interior cross-section 10 cm by 10 cm, height 21 cm) filled to a depth of 16 cm with nearly pure glycerine (viscosity  $\mu \approx 7-8 \,\mathrm{g\,cm^{-1}}$  s, density  $\rho \approx 1.2 \,\mathrm{g\,cm^{-3}}$ ) to observe the settling of blobs of spherical glass particles. The particles had diameters around 0.9 mm, with some polydispersity (0.05 mm standard deviation), and were observed to settle individually at speeds of roughly 0.06 to 0.1 cm s<sup>-1</sup>, which corresponds to a Reynolds number of order 10<sup>-3</sup> and a Stokes number of order 10<sup>-4</sup>. Blobs were formed by pre-wetting the particles with glycerine and then carefully pushing a small drop of the resulting dispersion onto the liquid surface from a spoon clamped 2-3 cm above the surface. Smallish blobs generated in this way had diameters around 0.7 cm and typically settled with speeds of about 0.6 to 2 cm s<sup>-1</sup>, which correspond to blob Reynolds numbers less than about 0.25. The particle and blob Reynolds numbers thus were sufficiently small to satisfy the condition for Stokes flow needed for applicability of the numerical analysis. The actual blobs were concentrated, not dilute, and their initial shapes (being uncontrolled) were not as nearly spherical as those simulated numerically.

As in the numerical calculations, falling blobs were observed to remain cohesive, to maintain roughly constant size, and to shed particles sporadically in a line trailing from the rear. Figure 5 shows representative photographs of a number of these blobs, which have a general appearance very similar to those shown in figure 1. The streaks of fluid faintly visible behind the blobs (distinct from the particle tails) may represent surface contaminants (e.g. water adsorbed from the atmosphere) dragged into the liquid when the blob splashed through the surface.

The authors are pleased to thank Dr E. J. Hinch for useful and enlightening discussions regarding the numerical simulations, and Mark Hallworth for his help and advice on the experimental set-up. Support from the US National Science Foundation is gratefully acknowledged.

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https://doi.org/10.1017/50022112097005223 Published online by Cambridge University Press

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