Disorder and Mixing

Convection, Diffusion and Reaction in Random Materials and Processes

edited by

Etienne Guyon
University of Paris XI, and
Ecole Supérieure de Physique et de Chimie Industrielles de la Ville de Paris, Paris, France

Jean-Pierre Nadal

and

Yves Pomeau
Département de Physique de L'Ecole Normale Supérieure, Paris, France

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Chapter IX

SEDIMENTATION OF SMALL PARTICLES

E.J. Hinch
D.A.M.T.P., Cambridge University
England

Introduction

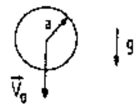
This lecture is too theoretical, so let's start with some practical examples. The sedimentation of particles is one of the basic techniques available in chemical engineering to unmix things - precisely the opposite of the subject of this volume! Sometimes the purpose of the sedimentation is the recovery of the liquid as in the purification of water in sewage treatment plants, while sometimes it is the collection of the particles as in the recovery of cream (fat globuls) from milk. Different types of particles can be separated from one another as in the enrichment of mineral ores. In the atmosphere rain drops, dust and pollution in ash fall out under the action of gravity. Where a fresh water river meets the saline sea water, the repulsive electrical forces between clay platelets become screened by the counter-ions from the salt, so that the platelets can aggregate under van der Waals forces, and the aggregates sediment thus silting up the estuary with mud. In all these applications, one is interested in the time for the sedimentation, and also the structure of the sediment.

I will be concerned with the sedimentation of small particles for which the Reynolds number is small. In water this means typically that the particles should be smaller than 0.1mm, while in air the restriction is more severe at 10μm. The main difficulty will be found to be the very long range hydrodynamical interactions between the particles.

One sphere

Balancing the Stokes drag $6\pi\mu aU$ with the weight of the particle (suitably compensated for buoyancy) mg, we have the fall speed of an isolated sphere:

$$V_0 = mg/(6\pi\mu a)$$



It will be recalled that the drag on a non-spherical particle is less than that on the smallest enclose sphere, but typically not less than one third of that value. Hence the above fall speed can be applied as a good estimate to arbitrary shaped particles, including tenuous fractal ones.

Because the weight of a solid sphere increases with its volume, the sedimentation rate increases with the square of the radius of the sphere. Hence larger particles overtake smaller particles, and this provides an efficient aggregation mechanism for cohesive (sticky) particles.

Now we have calculated the flow outside a falling sphere

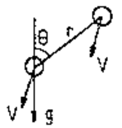
$$u = V_0\left[3a/4r + a^3/4r^3\right] + V_0.xx\left[3a/4r^3 - 3a^3/4r^5\right]$$

I would like to note three features of this flow field for use later. First, the flow disturbance decays very slowly, like O(1/r). This will cause severe problems for nearly every theoretical calculation of interactions between particles. Second, at a fixed large distance, the speed varies between $3V_0a/4r$ at locations on the same horizontal plane as the particle to $3V_0a/2r$ precisely above or below the particle. This factor of 2 is due to the pressure field maintaining the flow as divergent-free. Finally it should be noted that the (instantaneous) streamlines are converging above the particle and diverging below.

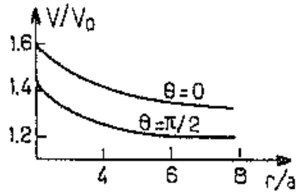
Two equal spheres

The Stokes flow for two moving spheres can be obtained by a number of techniques. It is possible to make a harmonic expansion in bi-spherical polar co-ordinates, resulting in a recurrence relation between the amplitudes. There are various numerical methods such as the boundary integral method with a lubrication theory correction if the spheres are close. Finally there is a method of reflections which will be explained in the next section.

By reversibility and the symmetry, two equal spheres will fall at the same velocity, and so they will not change their separation and orientation. Except when $\theta = 0$ or $\pi/2$, they will have a horizontal component of their fall velocity. This sideways motion leads to a horizontal mixing as a suspension sediments. There is also a vertical mixing due to the variation of the vertical velocity with the separation of pairs.



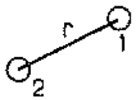
The variation of the vertical fall speed is sketched in the figure. A pair will fall faster than an isolated sphere, roughly 50% faster for close pairs. Note that for $r \geq 4a$, the extra velocity for $\theta = 0$ is roughly twice that for $\theta = \pi/2$. This reflects the 2:1 variation noted in the flow outside an isolated sphere.



Method of reflection (a rather technical section)

This is an asymptotic method for calculating the interaction between two greatly separated spheres, $r >> a$.

Reflection 0. In the lowest approximation each sphere is effectively isolated. Thus sphere 1 generates a flow $\mathbf{u}_1(\mathbf{x})$ as if sphere 2 is not present.



Reflection 1. In the first reflection, sphere 2 sees the flow $\mathbf{u}_1(\mathbf{x})$. In the neighbourhood of sphere 2, we may expand this in a Taylor series

$$\mathbf{u}_1(\mathbf{x}) = \mathbf{u}_1(\mathbf{x}_2) + (\mathbf{x} - \mathbf{x}_2).\nabla\mathbf{u}_1 \Big|_{\mathbf{x}_2} + \frac{1}{2}(\mathbf{x} - \mathbf{x}_2)^2.\nabla\nabla\mathbf{u}_1 \Big|_{\mathbf{x}_2} + \ldots$$

The first, dominant term is a uniform flow in the neighbourhood of sphere 2, and sphere 2 will respond to it just by moving faster than V_0 by precisely this extra velocity. These corrections to the speed of sphere 2 are $O(V_0 a/r) + O(V_0 a^3/r^3)$.

The second, smaller term in the Taylor series is a linear velocity $O(V_0 a^2/r^2)$. This linear velocity variation will not change the speed at which sphere 2 falls. The vorticity will, however, cause sphere 2 to rotate. As the rigid sphere cannot deform with the straining component of $\nabla \mathbf{u}_1 |_{x2}$, the sphere will respond with a force-dipole $O(V_0 a^2/r^2)$.

The third, even smaller term in the Taylor series is a quadratic flow $O(V_0 a^3/r^3)$. We have seen that Faxen's formula implies that sphere 2 will fall at an extra velocity $O(V_0 a^3/r^3)$. As the sphere is rigid, it will also respond with a force-quadrupole $O(V_0 a^3/r^3)$.

It should be noted that while sphere 2 takes on various force-multipoles in response to sphere 1, the force-monopole is the weight of the particle and so is fixed.

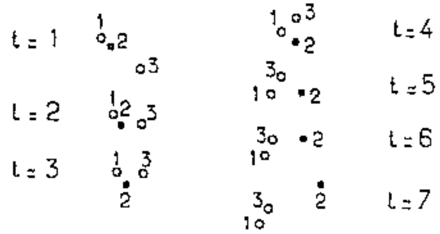
Reflection 2. Sphere 1 now sees the response of sphere 2 to $\mathbf{u}_1(\mathbf{x})$. There will be the flow induced by the $O(V_0 a^2/r^2)$ dipole, the flow induced by the $O(V_0 a^3/r^3)$ quadrupole, and many smaller effects.

Sphere 1 will respond by moving at an extra velocity $O(V_0 a^2/r^2.a^2/r^2) + O(V_0 a^3/r^3.a^3/r^3)$, i.e. $O(V_0 a^4/r^4) + O(V_0 a^6/r^6)$, as well as some corrections to the strengths of the force-multipoles.

The algebraic details in proceeding to higher order terms (and even this far!) are tedious. Fortunately the details are very predictable, and so it is possible to train a computer to manipulate the algebra, proceeding easily out to 90 reflections. Techniques to accelerate the convergence can then be applied.

Three equal spheres

As an amusing interlude, I present the results of a numerical simulation by van Rensburg in Cambridge of the three spheres interacting. The pair 1-2 catch up with sphere 3. During the collision, the central sphere 2 is pushed down out of the way thereby allowing spheres 1 and 3 to come together. The new pair 1-3 then moves off faster, leaving sphere 2 behind.



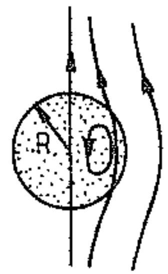
Many, N, particles in a cloud surrounded by clear fluid

A finite cloud of particles will behave like a suspended fluid drop which has a different density and viscosity, but with no surface tension. If the cloud starts spherical and the motion has a low Reynolds number, then it happens that the cloud will remain spherical and it will move at a velocity

$$U = Nmg/4\pi\mu R$$

The factor 4 is for a cloud with a low volume fraction of particles $\phi = Na^3 << 1$, which will have an effective viscosity equal to that of the clear fluid. The factor must be increased to 6 as ϕ increases and the viscosity of the cloud becomes large.

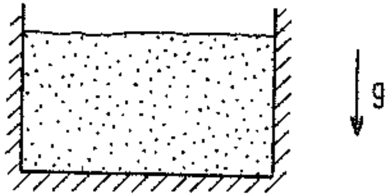
It should be noted that the speed of the cloud is $3V_0R^2/(2a^2)$, i.e. much faster than the fall speed V_0 of the individual particles. This large co-operation effect is a result of the long range $O(1/r)$ hydrodynamic interactions. In practice the Reynolds number for the motion of the cloud will not be small, and so a modification will be needed to the above description.



Streamlines of fluid relative to moving cloud

Many particles dispersed uniformly throughout a vessel with vertical side walls (and a bottom).

The horizontal homogeneity rules out the strong convection of the previous section. We will see in a later section about the Boycott effect that the walls must be vertical in order to preserve this horizontal homogeneity. The existence of a fixed bottom of the vessel requires that the average velocity in the suspension vanishes, $\langle u \rangle = 0$. Here the fluid must move upwards in order to compensate for the descending particles. This back flow in fluid hinders the fall of the particles and leads to a sedimentation speed which is *less* than the fall speed of an isolated particle.



We will calculate the sedimentation speed for a dilute suspension with a small volume fraction, $\phi << 1$. In a dilute suspension, one might hope just to sum the effects between pairs of particles. If the velocity of a pair of particles at a separation r is written as $V_0 + \Delta V(r)$, then naively averaging over all possible separations which occur with probability p(r) would lead to the expression for the average fall speed

$$V_0 + \int \Delta V(r)\, p(r)\, d^3r$$

Unfortunately the long range hydrodynamic interactions make this integral diverge, as $\Delta V(r) = O(1/r)$ and $p \rightarrow$ constant as $r \rightarrow \infty$. Although only the fine details of the interaction between pairs of particles is required, there is however, a multi-particle effect in that the important back-flow in the fluid is non-zero only when all the particles are considered.

Batchelor's renormalization

Batchelor noted that the fall speed of a pair of particles had the form at large separations (see the earlier section on the method of reflections)

$$\Delta V(r) = \left(1 + \frac{1}{2} a^2 \nabla^2\right) u_1 \Big|_{x=x_2} + \text{higher reflections}$$

The contributions from the first two terms are $O(1/r)$ and $O(1/r^3)$ which lead to divergent integrals. Batchelor was, however, able to calculate their averages by some global considerations. The contribution from the higher reflections (which contain the fine details of the interactions between pairs) is $O(1/r^4)$ and this leads to a convergent integral with a value $-1.55\phi V_0$.

Batchelor identified the first term as representing the velocity of the fluid at a position where a test sphere could be located. Now the average velocity of the fluid must be asymptotically $-\phi V_0$ in order to compensate for the volume fraction ϕ of particles which sediment approximately at V_0 when $\phi << 1$. The centre of the test sphere cannot be located, however, within an excluded volume shell $a < r < 2a$ of any other sphere (or else the two spheres would overlap). Within this excluded shell, an isolated sphere carries with it a flux of fluid $6\pi a^3 V_0$, and so the average velocity within the fluid of the suspension at locations where the test sphere can be placed is $-5.5\phi V_0$.

Batchelor constructed a similar argument for the second term, relating the average curvature in the flow of the fluid (the average velocity is upward, but downward near each particle) to the average pressure gradient, and so finding an average $0.5\phi V_0$.

Hence the average fall speed in the suspension was found to be

$$V_0 \left(1 - 6.55\phi + O(\phi^2)\right)$$

Alternative renormalization

I have developed an alternative approach which starts from the equations governing the fluid outside a test particle.

$$-\nabla p + \mu \nabla^2 \mathbf{u} + \rho g = \sum_p \int \sigma.n\, dA$$

where the sum is taken over all the other particles in the suspension.

Now a second particle acts like :- (i) a distributed weight which induces a change in the fall speed $O(1/r) + O(1/r^3)$, (ii) a stress (force-dipole) which enhances the viscosity and induced a change in the fall speed $O(1/r^4)$ and (iii) higher reflections which induces a change in the fall speed $O(1/r^6)$.

If the first two effects are taken onto the left-hand side of the equation, and we then average over all possible second particles, we obtain an equation for the suspension

$$-\nabla\langle p\rangle_1 + \mu\left(1 + \frac{5}{2}\phi\right)\nabla^2\langle u\rangle_1 + (\rho + \Delta\rho\phi)g$$

$$= \int\int \left[\sigma.n dA - \text{weight} - \text{viscosity} = O(1/r^6)\right] p(r) d^3r$$

Actually the density has a more complicated form in the region $a < r < 3a$, and the buoyant part reduces the fall speed of the test sphere by $-5\phi V_0$. The modification in the viscosity, which also has a more complicated form in the region $a < r < 3a$, reduces the fall speed by $205\phi V_0/128$. Adding in the convergent contribution from the higher reflections recovers Batchelor's result.

This alternative renormalization yields the self-consistent field model (in the excluded shell version) if one ignores all the higher reflections. This second renormalization is required when tackling more severe hydrodynamic interactions such as those occur in the 'porous medium' problem.

Measurements of the hindered settling of a suspension

Batchelor's theoretical result has been tested against careful experimental data. The asymptotic result fits the data well for $\phi < 0.05$, and begins to deviate significantly at $\phi = 0.1$ where the observations find $0.5V_0$ compared with the theoretical result $0.345V_0$. As ϕ increases to the maximum packing value, the average fall speed drops to a small, but non-zero, value about $0.015V_0$. This situation corresponds to the weight of the particles driving the fluid through a packed bed.

The phenomenon of hindered settling has some significant implications for the diffusion of particles in a suspension. The diffusive flux of particles down a concentration gradient does feel the hindering effect, whereas the random walk of one marked test sphere in a spatially homogeneous suspension does not. The latter random walk is however made complicated by hydrodynamic interactions, there being a short time diffusivity (where the other particles effectively slow the test sphere by enhancing the viscosity) and a long time diffusivity (in which the test sphere is retarded by an accumulation of other spheres blocking its way ahead). It should also be noted that when particle interactions are taken into account dynamic light scattering does not measure any of the above diffusivities.

Batchelor's theory for sedimentation of a suspension of spheres has been extended to polydisperse suspensions (in which one must calculate the probability distribution of pairs now that they do move relative to one another) and to suspensions with short range interactive forces (attractive forces leading to more close pairs and hence a higher settling speed).

Fluctuations and dispersion

So far we have concentrated on the average fall speed of the particles in the suspension. Some spheres, however, will be travelling faster than the average and some slower. These fluctuations around the average will lead to a dispersion or mixing process. The diffusivity is given by

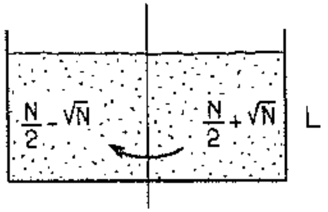
$$\kappa = \int_0^\infty \langle v'(t) v'(t+\tau)\rangle\, d\tau$$

An attempt to evaluate this expression for a dilute suspension leads to a divergent integral : for a pair of particles separated by distance r, $v' = O(V_0 a/r)$ and the time for Brownian motion to change the separation is r^2/D, which gives an integrand $O(V_0^2 a^2/D)$ independent of r. The slightly easier question of the r.m.s. of the velocity fluctuations also leads to a divergent integral. It is not known how to renormalize these two problems.

One can begin to understand the problem with the velocity fluctuations by considering a box of size L containing N uniformly distributed particles. Now following an idea by Russ Caflish if we divide the box into two equal volumes by a vertical plane, one half of the box will contain $N/2 - \sqrt{N}$ particles, while the other half will contain $N/2 + \sqrt{N}$.

The extra weight in the one side will drive a convection current

$$v' \sim \sqrt{mg}/6\pi\mu L = V_0 \phi^{1/2} (L/a)^{1/2}$$



if the convection is limited by friction forces. Thus the fluctuations become larger with increasing the box size, and indeed experiments are observed to be dominated by strong convection in the initial moments. One can speculate that after some time these convection currents will remove horizontal fluctuations in the number density down to the irreducible scale of interparticle separations, $a\phi^{-1/3}$. This would lead to a prediction of fluctuations $O(V_0\phi^{1/3})$.

In many experiments the initial large convection currents are limited by inertia rather than viscous forces with

$$v' \sim (ga)^{1/2}(L/a)^{-1/4}$$

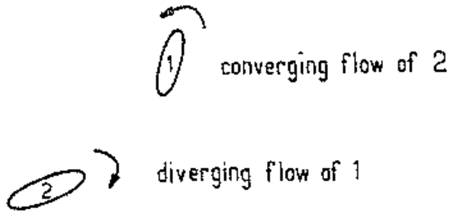
As this result for high Reynolds number decreases with the size of the box, whereas the fluctuations increase for low Reynolds numbers, one might expect that the observed fluctuation be those with a length scale which gives a Reynolds number of order unity, i.e.

$$v' \sim V_0\phi^{1/3}(V_0a/\nu)^{-1/3}$$

It is even less clear in this case how the state of the suspension evolves in time. Please note that the above discussion is very speculative.

Hydrodynamic screening by reorientation

The ideas in this section are new, speculative and controversial - they are the result of a collaboration with Eric Shaqfeh and Don Koch who come to a different conclusion!



Consider the two rod-like particles in the figure sedimenting under gravity. Because particle 1 points downwards more than particle 2, it will fall faster and hence overtake particle 2. Now particle 1 is in the converging flow behind particle 2, and this makes particle 1 align more with the direction of gravity. On the other hand particle 2 is in the diverging flow in front of particle 1, and this makes particle 2 align more with the horizontal. These $O(1/r^2)$ rotations of the particle will speed up particle 1 and slow down particle 2. This change in speed accumulates over time $O(r/V_0)$ and so particle 1 overtakes particle 2 at a faster speed $O(1/r)$ due to the reorientation.

Now in a steady state there will be a constant flux of particles like 1 overtaking particles like 2. With a constant flux, there will be reduction in the probability distribution where the particles are travelling faster, i.e. $\Delta_{prob} = -O(\phi/r)$. This slowly decaying change in the pair probability distribution leads to a diverging integral $\int V_0\Delta_{prob}(r)r^2\,dr$ in the calculation of the effect of particle interactions on the average sedimentation rate.

The resolution of this divergence may be as follows. Particle 2 will not see the average velocity outside particle 1 as in the pure fluid, u_1, but will see the average velocity outside particle 1 as in the suspension, $\langle u\rangle_1$, and the deficit of particles near particle 1 will make $\langle u\rangle_1$ decay to zero much faster than does u_1, because the far-field reflects the total weight (force- pole solution). Now the angular velocity of particle 2 induced by particle 1 will be $O(\nabla\langle u\rangle_1)$. Integrating this along a trajectory will lead to a reorientation $O(\langle u\rangle_1)$, and so a net change in the probability density $\Delta prob = -k\phi\langle u\rangle_1$. The constant k has to be determined by averaging over all the initial orientations of particles 1 and 2, although I argued in the previous paragraph that it must be

positive. The equation governing $\langle u\rangle_1$ will be the Stokes equation with a buoyancy term varying to reflect $\Delta prob(r)$, i.e.

$$0 = -\nabla\langle p\rangle_1 + \mu\nabla^2\langle u\rangle_1 + (\rho g\Delta prob = -k\phi\langle u\rangle_1)$$

Hence $\langle u\rangle_1$ has exponentially screened solutions like $\exp(-\phi^{1/2}r)$, and so the divergence difficulties disappear.

I should point out that Don Koch and Eric Shaqfeh believe that the constant k is negative, and so there is an excess rather than a deficit of other particles in the neighbourhood of the test particle. This excess would lead to an instability of the suspension by a spontaneous aggregation.

Theoretical studies of the sedimentation of concentrated suspension

It is worth noting two soundly based theoretical approaches. One is flow through periodic media. There are numerical results for the drag (and hence the sedimentation rate) for spheres at the maximum packing of a number of different lattices. The other, more general, promising approach is the Stokesian dynamics pioneered by Brady and Bossis.

Mixture of particles

As mentioned earlier, the theoretical studies of sedimentation may be extended to mixtures of different types of particles. There are some minor technical complications in the calculation of the probability distribution for the separation between different types of particles in the cases where pairs of particles stay nearby one another in a periodic relative motion instead of one particle steadily overtaking the other. These studies, however, give no indication of the fascinating phenomenon which occurs at moderate concentrations $\phi > 20\%$.

With a mixture of two types of particles, one heavier than the suspending fluid and one lighter, one would expect the downward flux of the heavy particles to hinder the upward motion of the light particles, thus leading to a reduction in the sedimentation speeds. At moderate concentrations, however, the two types of particles separate from one another into vertical columns several particles thick, and these columns move at greatly enhanced speeds, typically $10V_0$, just as the macroscopic finite cloud moves faster than an isolated particle. The formation of these streaming columns has been studied in Cambridge theoretically by Batchelor as an instability and experimentally by van Rensburg observing a geometrical blockage (to light ones rising by heavy ones descending) which is convectively unstable. In practice there is a complication that the Reynolds numbers for the streaming columns is not small.

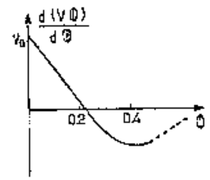
Formation of sedimentation shocks in a horizontally homogeneous suspension

We now turn to some macroscopic phenomena. First we consider a suspension sedimenting in a vessel with vertical side walls, which will preserve the assumed horizontal homogeneity. Variations of the concentration in the vertical, $\phi(z,t)$ will be considered. We assume that the sedimentation speed depends only on the local concentration, i.e. $v(\phi)$. With no bulk convection currents, the conservation of the particles is given by

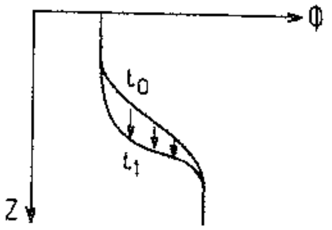
$$\frac{\partial c}{\partial t} + \frac{\partial}{\partial z}(v\phi) = 0$$

This may be re-written in the hyperbolic form

$$\frac{\partial c}{\partial t} + \frac{d(v\phi)}{d\phi}\cdot\frac{\partial c}{\partial z} = 0$$



The hindered settling phenomenon means that, while $d(v\phi)/d\phi$ starts from V_0 at $\phi = 1$, it decreases rapidly to negative values before increasing at a small negative value at maximum packing. Thus lower values of ϕ propagate faster than higher values. This leads to the formation of sharp shocks if the initial concentration has lower values located above higher values (the stable configuration!). The speed of these shocks can be calculated from conserving the flux of particles across the shock, resulting in $(\phi_+ V_+ - \phi_- V_-)/(\phi_+ - \phi_-)$.

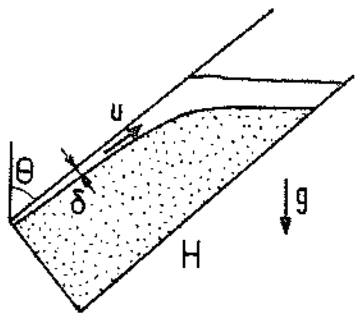


In practice the shocks are observed to be very sharp. The structure of the shocks has not been studied theoretically because of the complications of the local inhomogeneity and the selective loss of close groups of particles which fall faster than the front.

Settling in an inclined vessel - the Boycott effect

This interesting phenomenon has recently received much attention by Acrivos and a series of research students.

When separating particles from the fluid in a test tube, it is an established laboratory practice to tilt the test tube in order to reduce the settling time by a factor of about 3. This reduction simply reflects the fact that the particles need only fall the shorter distance vertically across the tube, instead of the whole length of the tube, in order to clear the fluid.



There are some interesting details in the motion. As the particles fall they leave a layer of clear fluid near the upper side wall. This layer of clear fluid is very buoyant and so rises at a much greater speed than V_0. We can estimate the thickness of this thin layer δ and its speed U by (i) balancing the viscous and buoyancy forces in the layer $\mu U/\delta^2 = \Delta\rho g \cos\theta$ and (ii) conserving the flux of clear fluid generated $V_0 \sin\theta H = U\delta$. This yields $\delta = (\mu H \tan\theta/\Delta\rho g)^{1/3}$ and $U = V_0 \sin\theta H/\delta$. This fast layer of buoyant clear fluid suffers a shear flow instability which is partially stabilized by gravity when the tube is either nearly vertical (low flux in layer) or nearly horizontal (strong stratification). This instability limits the practical application to industrial separators because it re-mixes the clear fluid with the suspension.

Student exercise

Where a fresh water river meets the saline sea water, the repulsive electrical forces between clay platelets become screened by the counter-ions from the salt, so that the platelets can aggregate under van der Waals forces, and the aggregates sediment thus silting up the estuary with mud. There are many similar situations in industry where the small suspended particles suddenly become attractive and so are precipitated. The exercise is to predict the time for the particles to fall out and to predict the structure of the precipitate.

Initially the particles and their clusters are small and so they come into contact by Brownian motion. Thus the aggregates grow according to D.L.A. in its cluster-cluster form. At a critical size, however, their sedimentation speed becomes significant and thereafter the aggregates grow by the larger (faster) aggregates sweeping up the smaller ones. This growth will perhaps be limited by the strength of the bonds in the aggregates compared with the fluid stresses due to the falling. In some applications there will also be some fluid stresses due to convective motions or imposed stirring. For compact bodies rather than the tenuous fractal aggregates, I believe the critical size for sedimentation to be important is about 10μm.

When the growth of the aggregates is understood, it is possible to predict the time it takes for the particles to fall out. We would also like to know the structure of the precipitate. The density of the precipitate would give the height it occupies - in elementary chemistry experiments I can recall the precipitate at the bottom of a test tube occupied more than ten times the volume of the filtered and dried material. One could also find the strength of the precipitate and consider whether it would compact under its own weight - there are some embarrassing industrial problems where some waste material will not compact and so it occupies a large volume. For drying the precipitate one would like to know its permeability, assuming that the fluid was not actively bonded to the solids.

I gave a third review lecture on the motion of an interface between two liquids. Much of the material on the deformation and breakup of small drops in shearing flows has been described clearly in an article by J.M. Rallison in the Annual Reviews of Fluid Mechanics in 1984.

References

General references :

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see also references given in chapter III

More specialis-zed references on hydrofynamic interactions :

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(3) E.J. Hinch, J.Fluid Mech. **83**, 695 (1977)