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BATCHELOR–CRIGHTON COMMEMORATIVE TALKS

A perspective of Batchelor's research in micro-hydrodynamics

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Batchelor made his name with research in turbulence in the 1940s and 1950s. He became disillusioned with turbulence at the Marseille meeting in 1961. At the end of the 1960s, he started his second wave of research on low-Reynolds-number suspensions of particles. Ten years after he died, I will describe his key results, what was before and what followed. Eight of his 10 most cited papers are in micro-hydrodynamics.

Key words: micro-/nano-fluid dynamics

1. Introduction

As described well by Keith Moffatt in his *Annual Reviews in Fluid Mechanics* (Moffatt 2002), Batchelor started his research in Cambridge in 1945. Understanding, explaining and exploiting two papers by Kolmogorov, and collaborating with fellow-Australian Alan Townsend, he made rapid progress in homogeneous isotropic turbulence, on which he was to write his classic graduate textbook in 1953 (Batchelor 1953). Gradually, the insurmountable mathematical difficulties became worrying. Then at the 1961 IUTAM/IUGG (International Union of Theoretical and Applied Mechanics/International Union of Geodesy and Geophysics) Symposium in Marseille, first Bob Stewart presented observations confirming Kolmogorov with three decades of the $k^{5/3}$ law, to be followed by Kolmogorov himself declaring that Landau had pointed out flaws in his theory, essentially due to intermittency. Batchelor then drifted away from turbulence. In the 1960s he was heavily involved with his *Journal of Fluid Mechanics*, publishing the collected works of G. I. Taylor and writing his own undergraduate textbook. This fascinating story and more is described by Moffatt.

Around 1967, Batchelor launched his research into low-Reynolds-number suspensions of particles, or 'micro-hydrodynamics' as he called it. I started my PhD with him in 1969. I never once heard him speak about turbulence, in public or private. There are few examples of such a complete switch of research topic, from his high to low Reynolds numbers.

In this review, I shall explain Batchelor's important contributions in micro-hydrodynamics, and set them in perspective by describing the research that has followed since. But first I should start with some work which preceded Batchelor.

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2. Previous work

In 1851, over 100 years before Batchelor started, Stokes derived the drag force $6\pi\mu a V$ for an isolated sphere of radius, a, moving at velocity, V, through a fluid of viscosity, μ. This result was used by Einstein in 1905 for his diffusivity of an isolated particle in Brownian motion, $D = kT/6\pi\mu a$, where kT is the Boltzmann temperature. In 1905, Einstein also found the expression $\mu(1 + 2.5c)$ for the viscosity of a dilute suspension of rigid spheres, where μ is the viscosity of the solvent and $c \ll 1$ is the volume fraction of the spheres. Batchelor was to find the first effects of hydrodynamic interactions between the particles on each of these three results.

In 1932, G. I. Taylor generalised Einstein's viscosity result to a dilute suspension of drops of another viscous fluid, with strong surface tension keeping the drops nearly spherical.

Skipping to the 1960s, there were then several groups working on suspensions: H. Brenner and R. G. Cox in New York, S. G. Mason in Montreal, H. Giesekus in Dortmund and in Cambridge F. P. Bretherton and P. G. Saffman.

One can speculate on how Batchelor came to select micro-hydrodynamics as a subject ripe for his attention. Working on his textbook and Taylor's collected works he would have seen opportunities.

3. A start

In 1967, Batchelor launched into his new field with two projects. The best he gave to a research student J. T. Green, and this was to calculate the $O(c^2)$ correction to Einstein's viscosity, which involves difficult hydrodynamic interactions between the particles. For himself, he chose to find an alternative approach to Einstein's for calculating the rheology. Einstein found the dissipative part of the stress characterised by a viscosity, but there are other components. Einstein also manipulated some divergent integrals.

In the study of suspensions of small particles in a viscous liquid, the smallness of the particles gives two simplifications. First, the flow around each particle has a low Reynolds number, and such flows are easier to calculate. Note that the Reynolds number for the bulk macro-scale flow need not be small, because at the macro-scale the velocity differences and length scales are much larger than at the micro-scale around the particles. Second, each particle only sees the local linear variation of the bulk velocity. This means that the rheology will be local, i.e. the bulk stress at one point will only depend on the strain rate (and possibly its Lagrangian history) at the same point, and not an average of the strain rate over some larger neighbourhood.

In Batchelor (1970*a*), he started by writing the stress at every point in the suspension as the expression for the stress in the solvent plus a correction which vanished outside the particles:

$$\sigma_{ij} = -p\delta_{ij} + 2\mu e_{ij} + \sigma_{ij}^{+}.$$

This was then averaged to find the bulk stress:

$$\langle\sigma_{ij}\rangle = -\langle p\rangle\delta_{ij} + 2\mu\langle e_{ij}\rangle + \langle\sigma_{ij}^{+}\rangle.$$

Batchelor used an ensemble average, taken from his research in turbulence, averaging over different experiments which start with the particles in randomly different positions. An ensemble average has the advantage over volume averaging in that there is no complication when taking a spatial derivative in order to evaluate the divergence of the stress in the momentum equation.

In spatially homogeneous conditions, the average of the contribution from within the particles can be switched to a volume integral. Then, using the divergence theorem for particles subject to no internal forces, the volume integral can be converted into an integral over A, the surface of a typical particle:

$$\langle \sigma_{ij}^{+} \rangle = n \int_A \sigma_{ik} n_k x_j - \mu(u_i n_j + u_j n_i)\,\mathrm{d}A,$$

where n is the number of particles per unit volume. This result, and the generalisation for non-zero internal forces, is much cited, even today clocking up around 30 citations per annum.

Batchelor's derivation of an expression for the full bulk stress, rather than just the viscosity, was not new. It can be found in a terse form in Landau & Lifshitz (1959). Batchelor's style was the opposite, to discuss in depth each assumption and each consequence. He was particularly concerned with the possibility of the stress tensor not being symmetric, $\sigma_{ij} \neq \sigma_{ji}$, when there are couples exerted on the particles. If the surface integral over the particle above is moved into the far field, the dominant contributions come from what he called a symmetric 'stresslet' and antisymmetric 'couplet' force dipoles, similar to the 'Stokeslet' when there is a net force exerted by the particle. Batchelor examined the energy budget, which gives the connection with Einstein's calculation of the dissipative part of the bulk stress. Batchelor was concerned with the contribution from surface tension, when the surface integral moves from just inside the particle to just outside.

What followed from this study? Well, to date over 640 citations. These are mostly for evaluation the rheology of many different types of suspensions. In fact, evaluating the bulk stress with Batchelor's expression is the easy part: first one has to solve the difficult micro-scale details, such as how does the microstructure evolve during the flow. Batchelor's ensemble average is essential for the few studies of non-local rheology, such as for the flow of suspensions of fibres when the bulk-flow scale is comparable with the length of the fibres (e.g. Shaqfeh 1988). In the 1980s, there emerged a technique called 'homogenisation' for calculating the macro-response of suspensions of small particles. This I view as a step back from Batchelor's work, because it is restricted to artificially periodic microstructures, although it has to be said that more recent large numerical simulations of suspensions are normally conducted in periodic boxes.

4. A second study

In 1971, Batchelor made his first application of his expression for the bulk stress to a non-dilute suspension of fibres in pure straining motion (Batchelor 1971). Later this regime would become known as 'semi-dilute', a regime in which the typical separation of the fibres is greater than their thickness, b, but less than their length, ℓ, the typical separation being $h = (n\ell)^{-1/2}$ for number density of fibres, n.

A year earlier, he had written a paper on slender-body theory for Stokes flow (Batchelor 1970*b*). He did not adopt the approach of formal matched asymptotic expansions, then being used by Cox and by Tillet, but did understand the two views: an outer view in which the fibre has zero thickness and is represented by a line distribution of force singularities and an inner view in which the fibre is infinitely long and isolated from other fibres. In this paper, Batchelor was concerned with the inner problem. He realised that the details of the shape of the cross-section were unimportant, and for the matching one only needed the 'equivalent radius' available from conformal maps for many shapes. In Batchelor (1971), he was concerned with the outer problem. He realised that in the semi-dilute regime the key length for the outer became the inter-fibre separation. Then he could evaluate his expression for the bulk stress to find the extensional viscosity

$$\mu_{ext} = \mu \frac{4\pi n \ell^3}{9 \log h/b}.$$

Note that in the semi-dilute regime this extension viscosity, μ_{ext}, of the suspension is much larger than the solvent viscosity, μ, which is the shear viscosity of the suspension. Batchelor's papers were theoretical. Batchelor (1971) contains unusually a comparison with a recent experiment, albeit a single data point and a not convincing comparison.

What followed from this study? The large extensional viscosity from a small volume fraction of fibres if $n\ell^3 \gg 1 \gg n\ell b^2$ helped explain how extremely small concentrations of polymers, c. 5 p.p.m., could reduce turbulent drag in pipe flow. Fredrickson & Shaqfeh (1990) justified Batchelor's dimensional analysis for the factor $\log h/b$ using a Brinkman description. The anisotropy in the viscosity, extensional large compared with shear, was found by Mongruel & Cloître (2003) to become reflected in an anisotropy in the flow, forming long narrow upstream vortices when a suspension of fibres flows from a wide pipe into a narrow one. I have speculated that such anisotropy is needed to explain how the turbulent axial velocity fluctuations remain unchanged, while the momentum transport and turbulent drag are reduced.

5. Re-normalisation of hydrodynamic interactions

5.1. *Sedimentation*

Batchelor's most important contributions are certainly his calculations of the first effects of hydrodynamic interactions between particles in dilute suspensions, often called the $O(c^2)$ problems. His first calculation in 1972 was for sedimentation, which is in fact an $O(c)$ correction to the Stokes settling velocity, $V_0 = 2\Delta\rho g a^2/9\mu$, of spheres, where $\Delta\rho$ is the density difference between the spheres and the solvent, g is the acceleration due to gravity and a is the radius of the spheres (see Batchelor 1972). The curious observation is that, while two close spheres settle faster than an isolated one, a suspension settles slower, through what is called 'hindered settling'.

It is useful to consider the effect on a test sphere of a second sphere at a distance, r. The additional fall speed of the test sphere ΔV can be found by the method of reflections

$$\Delta V(r)/V_0 = \left(\frac{a}{r} + \frac{a^3}{r^3}\right) + \left(\frac{a^4}{r^4} + \frac{a^6}{r^6} + \cdots\right) + \left(\frac{a^7}{r^7} + \cdots\right) + \cdots ,$$

where for simplicity the direction dependency of the coefficients has been suppressed. The two spheres react in turn to the disturbance caused by the other. Thus for the first bracket/reflection, the test sphere moves with the velocity field outside the second sphere as if the latter were alone. The leading term in the second bracket/reflection comes from stresslet (force-dipole) exerted by the second sphere in response to the straining motion outside the test sphere as if it were alone.

Attempting to average this expression by a naive pairwise addition of the disturbances from the spheres within a region $2a \leqslant r \leqslant R$, with n spheres per unit volume, one obtains

$$\langle \Delta V \rangle = \int_{r=2a}^{R} V_0 \left(\frac{a}{r} + \cdots\right) n \, \mathrm{d}V = O\left(V_0 c \frac{R^2}{a^2}\right),$$

where $c = (4\pi/3)na^3$ is the volume fraction of spheres. This estimate suggests that the mean settling speed is not an intrinsic property of the suspension but instead depends on the size, R, of the domain, contrary to experimental evidence. Batchelor's resolution of this divergence paradox was to understand the multi-particle effect which cannot be naively summed by pairwise addition.

Batchelor realised that the test sphere would fall at

$$\Delta V = \left(1 + \frac{a^2}{6}\nabla^2\right) u(x)\Big|_{test\ sphere} + \text{higher reflections},$$

where a pairwise sum of the $O(V_0 a^4/r^4)$ higher reflections is now convergent. Global considerations are needed to find the average of the first two terms. Now in the suspension the average velocity vanishes, averaging over falling particles and rising fluid, so long as the bottom of the container is impenetrable, i.e. $\langle u\rangle_{everywhere} = 0$. Each sphere takes with it a volume flux of $6\pi a^3 V_0$ in the excluded volume shell, $a < r < 2a$, where the centre of a second sphere cannot be placed. Hence the average (upward) velocity of the fluid, where the test sphere can be placed is $\langle u\rangle_{test\ sphere} = -5.5\, V_0 c$. This 'back flow' is a multi-particle effect. A slightly more complicated global argument gives the average of the Faxen correction term $\langle a^2\nabla^2 u/6\rangle_{test\ sphere} = \frac{1}{2} V_0 c$. Finally, the higher reflections have an average of $-1.55V_0 c$. Combining, Batchelor found the mean settling speed

$$\langle V\rangle = V_0(1 - 6.55c).$$

This calculation assumes a uniform distribution of the separation of pairs of spheres. As equal spheres sediment without changing their relative separation, the uniform distribution is plausible. As close pairs fall faster, if there were more close pairs than uniform, then the numerical coefficient 6.55 would be lower. On the other hand, if all the particles were far away from one another positioned on a lattice, then the mean settling speed has a different dependence on the volume fraction, $V = V_0(1 - kc^{1/3})$. Finally, there is an interesting difference between sedimentation, where one finds the average velocity with the same force exerted on the particles and a porous medium, where the particles move at the same velocity and one calculates the average force $\langle F\rangle = F_0(1 + 2.12c^{1/2})$.

What followed from this study? Over 750 citations. These include a number of erroneous applications, where the mathematically correct but physically wrong divergent integrand was subtracted to be averaged by global considerations: Batchelor isolated the physical cause of the divergence and tackled that. To settle the ensuing disputes, in 1977 I developed an alternative 'averaged-equation' approach, which recognised the divergences as changes of ρ and μ from their values in the solvent to those in the suspension (Hinch 1977). Batchelor himself used his re-normalisation approach in many further problems. He used his sedimentation result in 1976, along with the virial expansion of the thermodynamic chemical potential, to obtain the correction to Einstein's diffusivity

$$D = \frac{kT}{6\pi\mu a}(1 + 1.45c).$$

5.2. *The $O(c^2)$ correction to Einstein's viscosity*

The 1970 PhD thesis of J. T. Green contained useful data on the hydrodynamic interaction between two spheres in a linear shearing flow, but not the correction to

Einstein's viscosity, i.e. the value of k in

$$\mu* = \mu\left(1 + \tfrac{5}{2}c + kc^2\right).$$

After his breakthrough in the sedimentation problem, Batchelor applied his re-normalisation approach to the viscosity of a suspension of rigid spheres in two papers with Green (Batchelor & Green 1972*a*,*b*). For pure straining motion, they found $k = 7.6$. For simple shear, they could not find the value of k because the probability distribution of the separation of pairs of spheres in a region of closed trajectories could not be determined. Later, Batchelor (1977) added strong Brownian motion which permitted the probability distribution to be found, yielding $k = 6.2$ for all flows. Note that once interactions between the particles are considered, the rheology is *not* described by a constant isotropic viscosity which only depends on the concentration of particles.

5.3. *Polydispersity*

With a senior visitor, C.-S. Wen, Batchelor generalised his re-normalisation calculations for sedimentation (see Batchelor 1982; Batchelor & Wen 1982) and the diffusivity (see Batchelor 1983) to suspensions with a mixture of spherical particles of different densities and radii. With one important exception, I have viewed this development as a minor refinement. The exception is the mean settling speed for a suspension of non-Brownian spheres of the same density and nearly equal radii

$$\langle V\rangle = V_0(1 - 5.6c).$$

In experiments the spheres do not have exactly the same size. The slightly larger ones will fall faster than the smaller, slowing down as they overtake the smaller. This makes more close pairs than randomly uniform. This reduces the coefficient 6.55 for uniformly distributed separations of pairs. The reduced value is more consistent with the dilute limit of the well-established empirical Richardson–Zaki formula $\langle V\rangle = V_0(1 - c)^{5.1}$.

5.4. *What followed Batchelor's $O(c^2)$ re-normalisation*

Perhaps too much effort went into theoretical calculations of the first effects of interactions between particles in dilute suspensions. With the arrival of larger computers, it became possible to make numerical simulations with interactions between scores of particles, and so examine moderate concentrations.

John Brady (Brady & Bossis 1988) pioneered his Stokesian dynamics approach for suspensions of rigid spheres, combining a good treatment of multi-particle effects for well separated particles with a pairwise-additive lubrication treatment of close particles. With about 1000 particles, the rheology and various diffusivities could be found in the following 10 years for concentrations up to 50 %.

With M. Loewenberg in 1996, I applied the by then well-developed boundary integral method for Stokes flows to emulsions of deformable viscous drops suspended in a second viscous fluid (Loewenberg & Hinch 1996). In contrast to rigid spheres, the slippery drops could with small deformations easily slide passed one another, resulting in only a dozen drops being needed to simulate volume fractions up to 30 %.

In contrast, A. J. C. Ladd has made simulations with very many more particles using a lattice-Boltzmann approach. His 1996 paper used over 32 000 particles to examine fluctuations in the velocities of sedimenting spheres (Ladd 1996).

6. More sedimentation

Batchelor's last paper on sedimentation was a mainly experimental paper with his student van Rensburg (Batchelor & van Rensburg 1986). They considered a bidisperse suspension in which one type of particle was heavier than the liquid and so sank, while a second type was lighter and so rose. At total concentrations around 30 %, the behaviour was not at all like that contemplated in his theoretical papers on polydisperse suspensions. The two types of particles separated into streaming vertical columns, which moved much faster than an isolated particle. This resulted in a practically useful faster separation process. A linear analysis of this structural instability was made which was 'generally consistent with the observed features'.

What followed in sedimentation? In 1979, Acrivos started a series of investigations into settling in inclined containers. In what is known as the 'Boycott effect', the separation process is enhanced because the particles need only settle the short vertical distance between the sloping upper and lower walls. The flow and its stability are summarised by Davis & Acrivos (1985).

Too many studies have considered the theoreticians' spherical particles. Koch & Shaqfeh (1989) considered a suspension of sedimenting fibres. A linear stability analysis found it to be structurally unstable to the formation of descending columns of particle-rich regions and ascending columns of particle-sparse regions. This was confirmed later by experiments and numerical simulations. The instability is not dissimilar to that seen by Batchelor and van Rensburg for bidisperse suspensions. Recently, Shaqfeh has suggested that a similar mechanism, in which the shear orients the particles to migrate to the descending denser regions, may also occur in suspensions of deformable drops.

An issue which has concerned me for far too many years is the fluctuations in the velocities of particles in a sedimenting suspension. A naive pairwise addition of the disturbances from spheres within a region $2a \leqslant r \leqslant R$ this time yields an estimate for the root-mean-square (r.m.s.) fluctuations $\langle V'^2 \rangle = O(V_0^2 cR/a)$. This dependence on the size of the container, R, is found in numerical simulations and is supported by a simple theoretical explanation. Experiments, however, found no such dependence. For a review of the current understanding of this paradox, see Guazzelli & Hinch (2011).

7. Suspensions: what happened in parallel

While Batchelor was making major progress in some topics in micro-hydrodynamics, he left alone many topics for his research students, post-docs and colleagues to develop on their own.

Diverted from the PhD project Batchelor had set me on deformable elastic particles by a confused seminar, I worked with the post-doc Gary Leal on the competition of flow-induced orientation and Brownian rotations of small non-spherical particles in a suspension. In a series of 10 papers we explored many limiting cases for the rheology (see e.g. Hinch & Leal 1972).

A post-doc Bill Russel started looking at the effect of electrical double layers which accompany colloidal particles. His first paper on the subject in 1978 (Russel 1978) found that the c^2-coefficient in the correction of Einstein's viscosity could become larger than 1000. He and others later explored other electro-kinetic phenomena, van der Waal attractions for flocculation, phase transitions and other effects in colloidal dispersions (see Russel, Saville & Schowelter 1989).

While G. I. Taylor had examined the small deformation of small viscous drops in linear shearing flows in 1934, Acrivos took up the subject in 1970 (see Frankel & Acrivos 1970) much motivated by an unpublished seminar by H. P. Grace, with several theoretical and numerical studies (see Rallison 1984).

Another post-doc David Leighton made the surprise observation, published later with Smart in 1989 (Smart & Leighton 1989), that glass spheres used in experiments were not smooth as supposed by theoreticians, but were seriously rough. This had important consequences for spheres shearing past one another, and gave one explanation of shear-induced viscous re-suspension.

8. What followed in later years

After the early studies of simple suspensions, many more complex systems have been investigated. Experiments have played a greater role than I have given the impression in this brief review. Modern experimental techniques are more varied, and systems are better characterised to enable international comparisons. Many different rheologies have been explored. There is a class of particles which respond to external imposed electrical or magnetic fields, so creating switchable materials, although I have not seen any serious practical applications.

I have concerned myself with the flow behaviour of fluids with complex rheologies, i.e. the consequences of stress relaxation, elastic tension in the streamlines, elastic stress saturation, thin boundary layers of high stretched fluid and anisotropic features of the flow due to anisotropic viscosities.

There has been an enormous explosion of interest in micro-fluidic devices, sometimes exploiting the techniques developed in micro-hydrodynamics, although more often ignoring them. Of particular concern is the regular production of drops, mixing at low Reynolds numbers and possible slip on hydrophobic walls. There is a new subject of micro-rheology of investigating the motion of small colloidal particles within very small-volume samples.

I have purposely not given references for any of these developments because the subjects are vast and Google would do a much better job.

9. Batchelor's last paper

In the late 1980s and early 1990s, Batchelor moved his attention to fluidised beds, a special type of suspension of particles where the particle Reynolds number is typically not small. As this is not really micro-hydrodynamics, I shall be brief. Batchelor's important contribution was to note that Jackson's 1963 instability of an inertial overshoot of a kinematic wave would produce alternating horizontal layers rich and sparse in particles, and that these layers would suffer a secondary instability of gravitational overturning (Jackson 1963). It was left to Jackson to suggest that Batchelor's instability thereby answered the long standing puzzle of why gas–fluidised beds form gas bubbles devoid of particles, while liquid–fluidised beds have only moderate fluctuations in the concentration of particles.

Batchelor's last paper was in 1997 with his ex-post-doc J. Nitsche on the fall of a cloud of small particles (Nitsche & Batchelor 1997). The cloud behaves like a liquid (a suspension, no less) of density and viscosity higher than that of the solvent. The cloud therefore falls like a dense liquid drop, with no surface tension. The fall speed is $c(R/a)^2$ faster than isolated particles, where R is the radius of the cloud, a is the radius of the particles and c is their volume fraction. The interesting behaviour was a gradual loss of particles from the cloud into the wake due to a random component of the particles' motion. This was the declared object of study. The rate of loss of particles was found in numerical simulations to be proportional to V_b/d, where V_b is the fall speed of the cloud and d is the mean inter-particle separation. This leakage rate was confirmed 10 years later by Metzger, Nicolas & Guazzelli (2007) in experiments and numerical simulations, at least while the cloud fell through the first 10 cloud diameters. After that they observed a $t^{-1/3}$ leakage rate. More interestingly in the late development, the loss of particles from the outer streamlines lead to the formation of a torus which then became unstable, breaking into two smaller clouds. This occurred long after the times observed by Batchelor & Nitsche (1997).

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