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On the bulk viscosity of suspensions

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The bulk viscosity of a suspension relates the deviation of the trace of the macroscopic or averaged stress from its equilibrium value to the average rate of expansion. For a suspension the equilibrium macroscopic stress is the sum of the fluid pressure and the osmotic pressure of the suspended particles. An average rate of expansion drives the suspension microstructure out of equilibrium and is resisted by the thermal motion of the particles. Expressions are given to compute the bulk viscosity for all concentrations and all expansion rates and shown to be completely analogous to the well-known formulae for the deviatoric macroscopic stress, which are used, for example, to compute the shear viscosity. The effect of rigid spherical particles on the bulk viscosity is determined to second order in volume fraction and to leading order in the Péclet number, which is defined as the expansion rate made dimensionless with the Brownian time scale. A repulsive hard-sphere-like interparticle force reduces the hydrodynamic interactions between particles and decreases the bulk viscosity.

1. Introduction

Particulate suspensions occur widely in nature and industry. Since the original work of Einstein one hundred years ago on the effective viscosity of a dilute suspension of rigid spheres (Einstein 1906) many studies have been devoted to determining theoretically or by experiment the ‘effective’ properties, e.g. viscosity, sedimentation rate, diffusion coefficients, etc., of suspensions under a wide range of conditions. G. K. Batchelor, the founder of the Journal we celebrate with this issue, was instrumental in laying out a programme of how to relate the macroscopic properties of two-phase materials to the underlying microscale physics when fluid mechanical interactions are important (Batchelor 1974). In a series of papers Batchelor and co-workers established the averaging procedure to pass from the microscale to the macroscale (Batchelor 1970), determined the necessary particle interactions at the microscale, e.g. for the shear viscosity (Batchelor & Green 1972*a*), and showed how the microstructure of the suspension – the spatial and temporal distribution of particles – must be determined as part of the problem in order to properly compute the averaged properties (Batchelor & Green 1972*b*; Batchelor 1977). Although the method laid out by Batchelor applies to all concentrations, his analytical work was limited to dilute suspensions where only single- or two-particle interactions are taken into account. In the last two decades computational methods have allowed the extension of this procedure to higher particle concentrations, even all the way up to close packing (Brady & Bossis 1988; Phillips, Brady & Bossis 1988; Ladd 1990; Sierou & Brady 2001). It has now become almost routine to compute effective properties of two-phase materials for a wide range of concentrations and conditions and these effective properties have been utilized in constitutive equations to model the macroscopic flow of multiphase materials (e.g. Jackson 2000).

In this paper we follow the procedure laid out by Batchelor to determine the ‘effective bulk viscosity’ (also known as the second or expansion viscosity) of a dispersion of spherical particles. The only study of the bulk viscosity of two-phase materials appears to be that of G. I. Taylor (1954*a*), who used dissipation arguments to determine the bulk viscosity of a dilute suspension of compressible bubbles expanding in an incompressible fluid. We shall show that our approach of computing the bulk viscosity directly from the average macroscopic stress leads to the same result as Taylor for bubbles. Our focus in this paper, however, is on the complementary problem of incompressible particles in a compressible fluid.

Apart from Taylor’s work, we are not aware of any studies of the bulk viscosity of viscous suspensions, probably for the very good reason that in most applications both the fluid and particles are incompressible and therefore the bulk viscosity would not seem to matter. However, although locally on the microscale the particles and fluid may not be compressible, when viewed macroscopically as a phase both the fluid and particle phases are compressible. Thus, bulk viscosity effects may be important in constructing two-phase flow equations to model suspension behaviour. Indeed, kinetic-theory-based constitutive equations for the rapid flow of granular media contain a contribution from the bulk viscosity (Lun *et al.* 1984; Gidaspow 1994), even though the particles and air making up the granular media are individually (and in sum) incompressible. Why should the situation be any different in a viscous suspension? Furthermore, in the colloidal regime the microscale expressions for the shear viscosity contain terms that are almost identical to those in molecular fluids, and well-established expressions and results exist for the bulk viscosity of molecular fluids (Sigurgeirsson & Heyes 2003). Again, why should the situation be different in a suspension?

In pure fluids bulk viscosity effects are usually only important in regions of rapid compression (or expansion), such as may accompany shocks, and thus are not common in most everyday applications. In many two-phase flows, however, the particles and fluid often separate from each other leading to regions of very rapid variation in particle concentration – shocks in particle volume fraction – and thus bulk viscosity effects might be quite important in such systems. For example, in viscous suspensions the relative drag between the particle and fluid phases is balanced by the divergence of the particle-phase stress which, when combined with the particle-phase continuity equation, gives rise to a diffusion equation for the particle concentration field (Nott & Brady 1999; Fang *et al.* 2002). The bulk viscosity will add a non-diffusive term to this particle balance (partial differential) equation, changing its mathematical type, which may have important implications for modelling, especially in time-dependent flows.

On a different note, as we shall see below, the computation of the bulk viscosity ‘completes’ the set of possible rheological problems: the shear viscosity has a quadrupolar forcing proportional to the rate-of-strain tensor, the self-diffusion coefficient, which is equivalent to the ‘microviscosity’, has a dipolar forcing, and the bulk viscosity has a monopolar forcing. Also, we shall see that a new hydrodynamic interaction function is needed to compute the bulk viscosity, and this has been determined, which finally completes the set of hydrodynamic resistance functions for spherical particles.

The paper is organized as follows. In § 2 we recall the definition of the bulk viscosity for a pure fluid as relating the difference between the thermodynamic and mechanical pressures to the rate of volume expansion and ask how suspended particles affect this relationship. In § 3 we define the effective bulk viscosity for a suspension and show that for low-Reynolds-number flows the fluid motion can be decomposed into a uniform rate of expansion and an *incompressible* Stokes flow. In § 4 we consider a single isolated spherical 'particle' and compute the correction to the bulk viscosity to leading order in volume fraction, recovering G. I. Taylor's result for a bubble in the limit where the suspending fluid is incompressible. In § 5 we compute the $O(\phi^2)$ correction to the bulk viscosity for rigid particles in the linear response regime (small Péclet number), where the expansion flow's distortion of the equilibrium microstructure is small. This calculation is carried out for Brownian particles that interact through an excluded-volume interparticle force whose range is varied, thereby allowing one to investigate the influence of hydrodynamic interactions in a simple, systematic manner. Concluding remarks are given in § 6.

2. The bulk viscosity of a pure fluid

The constitutive equation for the stress tensor $\boldsymbol{\sigma}$ in a compressible Newtonian fluid is

$$\boldsymbol{\sigma} = -p_{th}\boldsymbol{I} + 2\eta\, \boldsymbol{\mathsf{e}} + \left(\kappa - \tfrac{2}{3}\eta\right)(\nabla\cdot\boldsymbol{u})\boldsymbol{I},$$

where p_{th} is the thermodynamic pressure, $\boldsymbol{\mathsf{e}} = \frac{1}{2}[\nabla\boldsymbol{u} + (\nabla\boldsymbol{u})^\dagger]$ is the rate-of-strain tensor with $\boldsymbol{u}$ the fluid velocity, η is the shear viscosity, and $\boldsymbol{I}$ is the isotropic tensor. The bulk viscosity κ gives the difference between the 'mechanical' pressure, defined as the mean mechanical stress, and the thermodynamic pressure due to the rate of volume expansion $\nabla\cdot\boldsymbol{u}$, namely,

$$-\tfrac{1}{3}\boldsymbol{I}:\boldsymbol{\sigma} \equiv p_{mech} = p_{th} - \kappa\nabla\cdot\boldsymbol{u}, \tag{2.1}$$

and measures the dissipation of energy in the fluid upon expansion. If the fluid is incompressible, $\nabla\cdot\boldsymbol{u} = 0$, then the mechanical and thermodynamic pressures are equivalent and the former is known to within an arbitrary constant; only the dynamical pressure, ∇p, enters the momentum balance. For a compressible fluid the thermodynamic pressure, temperature and density are related through an equation of state; the density satisfies the continuity equation and the temperature is determined by an energy balance.

To measure experimentally the bulk viscosity of a fluid, one could, for example, heat (or cool) the fluid uniformly so that it expands (or contracts) at a rate e $(=\nabla\cdot\boldsymbol{u})$ and measure the mechanical pressure, say by containing the fluid in a flexible membrane. The difference between the thermodynamic and mechanical pressures then gives the bulk viscosity times the rate of expansion.

The question we address in this work is how does the bulk viscosity change when particles are dispersed in the fluid? We can imagine repeating the heating process for a suspension, measuring the confining mechanical pressure and determining the bulk viscosity via the analogue of equation (2.1) for the suspension. Since the particles do not expand at the same rate as the fluid, they will create a disturbance flow in the fluid and thus dissipate more energy than the fluid would alone. This excess dissipation is manifested as a change in the bulk viscosity of the material.

3. Definition of the bulk viscosity of a suspension

To determine the bulk viscosity of a suspension we proceed as for determining the shear viscosity by computing the average stress in the material. The ensemble or volume average of the Cauchy stress $\boldsymbol{\sigma}$ in the material is given by

$$\langle\boldsymbol{\sigma}\rangle = -\langle p_{th}\rangle_f \boldsymbol{I} + 2\eta\langle\boldsymbol{e}\rangle + \left(\kappa - \tfrac{2}{3}\eta\right)\langle\nabla\cdot\boldsymbol{u}\rangle\boldsymbol{I} + n\langle\boldsymbol{S}^H\rangle,$$

where $\boldsymbol{e}$ is the rate of strain in the fluid, $\langle\ldots\rangle$ denotes an average over the entire suspension (particles plus fluid), $\langle\ldots\rangle_f$ denotes an average over the fluid phase only, n is the particle number density and the average 'hydrodynamic' stresslet is defined as a number average by $\langle\boldsymbol{S}^H\rangle = (1/N)\,\Sigma_{\alpha=1}^{N}\boldsymbol{S}_\alpha^H$, where the stresslet of particle α is given by

$$\boldsymbol{S}_\alpha^H = \tfrac{1}{2}\int_{S_\alpha}\left[(\boldsymbol{r}\boldsymbol{\sigma}\cdot\boldsymbol{n} + \boldsymbol{\sigma}\cdot\boldsymbol{n}\boldsymbol{r}) - 2\left(\kappa - \tfrac{2}{3}\eta\right)(\boldsymbol{n}\cdot\boldsymbol{u})\boldsymbol{I} - 2\eta(\boldsymbol{u}\boldsymbol{n} + \boldsymbol{n}\boldsymbol{u})\right]\mathrm{d}S; \qquad (3.1)$$

the integral is over the surface of particle α with normal $\boldsymbol{n}$ pointing into the fluid and $\boldsymbol{r}$ is a vector from the centre of the particle to a point on its surface. In writing (3.1) we have assumed that there is no net force on a particle, which can easily be relaxed, as we do below.

In Stokes flow it is permissible to impose a uniform rate of expansion everywhere in the fluid, with the disturbance flow created by the particles (and their fluid-mediated interactions) satisfying the usual incompressible equations of motion. That is, the fluid velocity can be decomposed into a uniform expansion and a disturbance (Stokes) velocity

$$\boldsymbol{u} = \tfrac{1}{3}e\boldsymbol{x} + \boldsymbol{u}^s,$$

such that

$$\nabla\cdot\boldsymbol{u} = e \quad \text{and} \quad \nabla\cdot\boldsymbol{u}^s = 0.$$

Corresponding to this velocity decomposition is the fluid stress associated with the expansion flow

$$\boldsymbol{\sigma}^e = -(p_{th} - \kappa\, e)\boldsymbol{I},$$

while the disturbance stress $\boldsymbol{\sigma}^s$ satisfies the Stokes equations

$$\nabla\cdot\boldsymbol{\sigma}^s = 0 \quad \text{with} \quad \boldsymbol{\sigma}^s = -p^s\boldsymbol{I} + 2\eta\boldsymbol{e}^s,$$

where p^s is the dynamical pressure distribution associated with the incompressible Stokes flow.

To the average Cauchy stress we must add any interparticle-force contribution $-n\langle\boldsymbol{x}\boldsymbol{F}^P\rangle$ and the stress due to Brownian motion. (We will not include the inertial Reynolds stresses as our interest is in low-Reynolds-number flows; these could be added if desired.) The final form for the bulk stress can be written as (Brady 1993)

$$\langle\boldsymbol{\Sigma}\rangle = -\langle p_{th}\rangle_f\boldsymbol{I} + 2\eta\langle\boldsymbol{e}\rangle + \left(\kappa - \tfrac{2}{3}\eta\right)\langle\nabla\cdot\boldsymbol{u}\rangle\boldsymbol{I} - nkT\boldsymbol{I} + n[\langle\boldsymbol{S}^B\rangle + \langle\boldsymbol{S}^P\rangle + \langle\boldsymbol{S}^E\rangle], \qquad (3.2)$$

where kT is the thermal energy and we have assumed that there are no external couples on the particles. The particle stresslets in (3.2) can be expressed in terms of N-particle hydrodynamic resistance functions as

$$\begin{aligned}
\langle\boldsymbol{S}^B\rangle &= -kT\left\langle\nabla\cdot\boldsymbol{R}_{SU}\cdot\boldsymbol{R}_{FU}^{-1}\right\rangle,\\
\langle\boldsymbol{S}^P\rangle &= -\left\langle\left(\boldsymbol{R}_{SU}\cdot\boldsymbol{R}_{FU}^{-1} + \boldsymbol{x}\boldsymbol{I}\right)\cdot\boldsymbol{F}^P\right\rangle,\\
\langle\boldsymbol{S}^E\rangle &= -\left\langle\boldsymbol{R}_{SU}\cdot\boldsymbol{R}_{FU}^{-1}\cdot\boldsymbol{R}_{FE} - \boldsymbol{R}_{SE}\right\rangle:\langle\boldsymbol{e}\rangle,
\end{aligned}$$

where the derivative is with respect to the last index of the inverse of the resistance matrix $\boldsymbol{R}_{FU}^{-1}$ and $\boldsymbol{F}^P$ is the colloidal interparticle force. The notation for the hydrodynamic resistance functions follows the convention introduced by Brady (1993);

their forms necessary to compute the bulk viscosity will be given in the next section. Note that the hydrodynamic functions are for an incompressible Stokes flow.

In the absence of any deforming motion, $\langle \boldsymbol{e} \rangle \equiv 0$, the suspension is in equilibrium, which we take to be an isotropic dispersion, and the bulk stress is the sum of the average equilibrium fluid pressure, $\langle p_{th} \rangle_f^{eq}$, and the osmotic pressure of the suspended particles:

$$\langle \boldsymbol{\Sigma} \rangle^{eq} = -\left(\langle p_{th} \rangle_f^{eq} + \Pi\right)\boldsymbol{I},$$

where the osmotic pressure Π is given by

$$\Pi = nkT - \tfrac{1}{3}n[\langle S^B \rangle^{eq} + \langle S^P \rangle^{eq}],$$

and S denotes the trace of the corresponding stresslet, e.g. $\langle \boldsymbol{S}^B \rangle^{eq} = \frac{1}{3}\langle S^B \rangle^{eq}\boldsymbol{I}$, and the superscript eq denotes an average over the equilibrium distribution of the suspension microstructure.

To determine the bulk viscosity, κ_{eff}, we subject the suspension to a uniform average rate of expansion $\langle e \rangle \equiv \langle \nabla \cdot \boldsymbol{u} \rangle$ and define the bulk viscosity as the scalar coefficient giving the difference between the mean suspension stress and the equilibrium stress. From (3.2) for the bulk stress we have

$$\kappa_{eff} \equiv \kappa + \left(-\langle p_{th} \rangle_f + \langle p_{th} \rangle_f^{eq}\right)/\langle e \rangle + \tfrac{1}{3}n[(\langle S^B \rangle - \langle S^B \rangle^{eq}) + (\langle S^P \rangle - \langle S^P \rangle^{eq}) + \langle S^E \rangle]/\langle e \rangle. \tag{3.3}$$

The Brownian and interparticle force contributions to the bulk stress require the interaction between at least two particles and therefore contribute $O(\phi^2)$ to the bulk viscosity in the dilute limit. The stresslet due to the rate of strain, $\langle \boldsymbol{S}^E \rangle$, is non-zero for a single particle, however, and therefore contributes $O(\phi)$ to the bulk viscosity.

4. The bulk viscosity to $O(\phi)$

The $O(\phi)$ contribution to the bulk viscosity arises from the disturbance flow induced by the presence of a single particle (which is needed to satisfy the no-slip condition on the particle's surface) immersed in the ambient uniform expansion flow. This disturbance flow increases the rate of dissipation in the fluid thus enhancing the bulk viscosity of the dispersion. Here, we explicitly calculate the $O(\phi)$ contribution for two limiting cases: (i) a rigid spherical particle in a uniform expansion flow of the surrounding compressible fluid; and (ii) G. I. Taylor's problem of a compressible spherical bubble expanding uniformly in an incompressible fluid.

4.1. *Rigid spherical particle*

Consider a rigid spherical particle of radius a immersed in a compressible fluid expanding uniformly at a rate e. As the particle is rigid it cannot deform (i.e. the expansion rate in the particle, e_p, is zero); hence, the average expansion rate in the dispersion $\langle e \rangle = e(1-\phi) + e_p\phi = e(1-\phi)$. The velocity field in the fluid is

$$\boldsymbol{u} = \tfrac{1}{3}e[1 - a^3/r^3]\boldsymbol{r},$$

which is a combination of the uniform expansion $\frac{1}{3}e\boldsymbol{r}$ and the disturbance Stokes flow $\boldsymbol{u}^s$. (Here, we have taken the origin at the centre of the particle, and $r = |\boldsymbol{r}|$.) Furthermore, the particle does not cause the pressure in the fluid to change (i.e. $p^s = 0$). There is no net force or torque on the particle, but there is a stresslet from (3.1) given by

$$S^E = 4\pi a^3\left(-p_{th} + \kappa e + \tfrac{4}{3}e\eta\right). \tag{4.1}$$

Thus, from (3.3) the bulk viscosity is found to be

$$\begin{aligned}\kappa_{eff} &= \left(\kappa + \tfrac{4}{3}\eta\,\phi\right)\frac{e}{\langle e\rangle},\\ &= \left(\kappa + \tfrac{4}{3}\eta\,\phi\right)\tfrac{1}{1-\phi}\\ &\sim \kappa + \tfrac{4}{3}\eta\,\phi \quad \text{as} \quad \phi \to 0, \end{aligned} \tag{4.2}$$

which corresponds to the 'Einstein' correction to the bulk viscosity for rigid particles. Note that the first- (and subsequent) order correction(s) to κ_{eff} is(are) proportional to the shear viscosity η and thus might not be small even for dilute suspensions, depending on the magnitude of the fluid's bulk viscosity κ.

4.2. *Spherical bubble*

Consider a compressible spherical bubble of (instantaneous) radius a, bulk viscosity κ_p, and zero shear viscosity. The bubble expands uniformly at a rate e_p in an otherwise quiescent incompressible fluid. Hence, the average expansion rate in the dispersion is $\langle e\rangle = e_p\phi$. The velocity field inside the bubble is a uniform expansion $\boldsymbol{v} = \frac{1}{3}e_p\boldsymbol{r}$, and the pressure inside the bubble is constant (say, p_p). The (Stokes disturbance) velocity field in the surrounding fluid caused by the expanding bubble is

$$\boldsymbol{u} = \tfrac{1}{3}e_p(a^3/r^3)\boldsymbol{r}.$$

Note that the interior and exterior flows are the same even if the bubble were a drop with a finite shear viscosity; hence, the results below apply equally well to spherical drops of arbitrary shear viscosity.

In contrast to the rigid particle problem, the expanding bubble causes the pressure in the surrounding fluid to be altered from its equilibrium value. The pressure in the fluid is found through a normal stress balance on the surface of the bubble, $(\boldsymbol{\sigma}_p - \boldsymbol{\sigma})\cdot\boldsymbol{n} = 0$ (here, we neglect surface tension effects; these can be added easily if desired and do not alter the final result), and is given by

$$p_{th} = p_p - \kappa_p e_p - \tfrac{4}{3}\eta e_p.$$

Thus, at equilibrium ($e_p = 0$) we have $p_{th} = p_p$ (the pressure inside the bubble is equal to that of the surrounding fluid; surface tension would simply add the capillary pressure). The stresslet on the bubble is then

$$S^E = 4\pi a^3(-p_p + \kappa_p e_p),$$

and from (3.3) the bulk viscosity is found as

$$\begin{aligned}\kappa_{eff} &= \left[\kappa_p + \tfrac{4}{3}\eta(1-\phi)\right]\frac{e_p}{\langle e\rangle},\\ &= \left[\kappa_p + \tfrac{4}{3}\eta(1-\phi)\right]\tfrac{1}{\phi},\\ &\sim \left[\kappa_p + \tfrac{4}{3}\eta\right]/\phi \quad \text{as} \quad \phi \to 0. \end{aligned} \tag{4.3}$$

Setting $\kappa_p = 0$ in the above we have $\kappa_{eff} = \frac{4}{3}\eta/\phi$, a result first derived by Taylor (1954*a*). At first glance, the inverse $(1/\phi)$ volume fraction scaling of κ_{eff} may seem somewhat surprising. Indeed, it concerned Taylor, who, in a subsequent paper (Taylor 1954*b*), showed that it may be removed by taking into account the small compressibility of the surrounding fluid. Nevertheless, for the bubble problem the $1/\phi$ scaling may be understood as follows. The small $O(\phi)$ concentration of bubbles produces a small (again, $O(\phi)$) average expansion rate ($\langle e\rangle = e_p\phi$) throughout the dispersion. However, the expansion of a bubble perturbs the pressure field over the entire surrounding fluid space, which occupies a large $(1-\phi)$ fraction of the dispersion. (This is in contrast to the stresslet exerted by a rigid particle, which is localized to its surface – in total a small ϕ fraction of the dispersion.) Thus, the dominant contribution to the bulk viscosity comes from the $\langle p_{th}\rangle_f/\langle e\rangle$ term in (3.3), which is of $O((1-\phi)/\phi) \sim O(1/\phi)$ as $\phi \rightarrow 0$. This should be contrasted with the $O(\phi)$ single rigid particle correction (4.2), for which the dominant contribution comes from the stresslet term $n\langle S^E\rangle/\langle e\rangle \sim O(\phi/(1-\phi)) \sim O(\phi)$ as $\phi \rightarrow 0$. That the coefficients are the same in the two problems, namely $\frac{4}{3}\eta$, stems from the fact that the disturbance flow in the surrounding fluid is the same both cases.

We have obtained results for the two limiting cases of a rigid particle in a compressible fluid and a compressible bubble (or drop) in an incompressible fluid. The general problem of a compressible drop in a compressible fluid is not simply a superposition of the above two problems (although the velocity fields are superimposable), as it entails the partitioning of the expansion between the two phases and the associated changes in the pressures in the two phases. When only one phase is compressible, its pressure does not change upon expansion and all the pressure change goes into the incompressible phase, which is able to support any pressure level, and the problem can be resolved purely mechanically without knowledge of the thermodynamics of the phases. In general, this is not the case and thermodynamic considerations must also enter.

5. The bulk viscosity to $O(\phi^2)$

The $O(\phi^2)$ correction to the bulk viscosity for rigid particles requires the interaction between two particles and now the Brownian, interparticle-force and rate-of-strain stresslets may all contribute. Furthermore, it is necessary to determine the distribution of particles – the suspension microstructure – and how this distribution is influenced by the expansion flow. The particle distribution satisfies the N-particle Smoluchowski equation, which, upon integration over $N-2$ particles and neglecting all three-particle effects, becomes the pair-evolution equation for identical particles

$$\frac{\partial P_{1/1}}{\partial t} + \nabla_r \cdot \left[\boldsymbol{U}_r^E + \boldsymbol{M}_r \cdot \left(\boldsymbol{F}_r^P - kT\nabla_r \ln P_{1/1}\right)\right] P_{1/1} = 0, \tag{5.1}$$

where $P_{1/1}(\boldsymbol{r}, t)$ is the conditional probability density of finding a second particle at $\boldsymbol{r}$ relative to one at the origin, $\boldsymbol{U}_r^E = \boldsymbol{U}_2^E - \boldsymbol{U}_1^E$ is the relative velocity of the two particles owing to the expansion flow, $\boldsymbol{M}_r = 2(\boldsymbol{M}_{22} - \boldsymbol{M}_{21})$ is the relative mobility, $\boldsymbol{F}_r^P$ is the relative interparticle force and $\boldsymbol{r} = \boldsymbol{x}_2 - \boldsymbol{x}_1$ is the relative separation vector. Equation (5.1) is subject to the boundary conditions of no relative flux at contact

$$\hat{\boldsymbol{r}} \cdot \boldsymbol{D}_r \cdot \nabla_r P_{1/1} = \hat{\boldsymbol{r}} \cdot \left[\boldsymbol{U}_r^E + \boldsymbol{M}_r \cdot \boldsymbol{F}_r^P\right] P_{1/1} \quad \text{at} \quad r = 2a, \tag{5.2}$$

where $\hat{\boldsymbol{r}}$ denotes a unit vector and $\boldsymbol{D}_r = kT\boldsymbol{M}_r$ is the relative diffusivity of the pair, and at large separations the probability density of finding a second particle is the number density

$$P_{1/1} \sim n(t) \quad \text{as} \quad r \rightarrow \infty. \tag{5.3}$$

While constant in space, as indicated in (5.3), the number density of particles is a function of time owing to the net rate of volume expansion. Specifically, the conservation of number density for a uniform bulk expansion with no macroscopic gradients in number density is given by

$$\frac{\partial n}{\partial t}=-\nabla\cdot\langle\boldsymbol{u}\rangle n=-n\nabla\cdot\langle\boldsymbol{u}\rangle=-n\langle e\rangle.$$

Writing the conditional probability density as $P_{1/1}=n(t)g(\boldsymbol{r},t)$ the equation for the pair-distribution function $g(\boldsymbol{r},t)$ becomes

$$\frac{\partial g}{\partial t}+\tfrac{1}{3}\langle e\rangle\boldsymbol{r}\cdot\nabla g+\nabla\cdot(\hat{\boldsymbol{U}}^E+\boldsymbol{M}\cdot\boldsymbol{F}^P)g=\nabla\cdot\boldsymbol{D}\cdot\nabla g,$$

where $\hat{\boldsymbol{U}}^E=\boldsymbol{U}^E-\frac{1}{3}\langle e\rangle\boldsymbol{r}$ is the disturbance velocity relative to the imposed expansion flow and we have dropped the subscript r for clarity. The boundary condition (5.2) remains the same for g, while the far-field condition (5.3) now becomes the time-independent statement $g\sim 1$ as $r\to\infty$.

The evolution of the pair-distribution function reflects the competition between the expansion flow, which drives the microstructure away from its equilibrium state, and Brownian diffusion, which acts to restore equilibrium. These competing effects are influenced by the level of hydrodynamic interactions between particles and it is desirable to tune the strength of the hydrodynamic interactions in a simple manner. As has become common in similar studies of shear rheology, this tuning can be accomplished through the 'excluded annulus' model where the two-body interparticle potential $V(r)$ is given by

$$V(r)=\begin{cases}\infty & \text{if } r\leqslant 2b\\ 0 & \text{if } r>2b.\end{cases}$$

The length $b(\geqslant a)$ s the excluded-volume, or thermodynamic, radius of a particle, and the centre-to-centre separation between particles can be no less than $2b$. Interactions of this nature may arise from e.g. surface roughness, grafted polymer chains, or electrostatic repulsion. Altering the parameter $\hat{b}\equiv b/a$ allows one to examine the role of hydrodynamic interactions in determining the microstructure and the bulk viscosity. In the limit $\hat{b}\to\infty$ the particles do not experience hydrodynamic interactions, while when $\hat{b}\equiv 1$ the particles experience full hydrodynamic interactions with one another. With this hard-sphere excluded-annulus model, the interparticle force does not appear explicitly in the pair-evolution equation; rather, the no-flux boundary condition (5.2) is imposed at $r=2b$. The diluteness assumption now requires the volume fraction based on the excluded-volume radius b to be small, i.e. $\phi_b=4\pi nb^3/3\ll 1$.

The spherical symmetry of the expansion flow allows us to write the relative particle velocity as

$$\boldsymbol{U}^E=\tfrac{1}{3}\langle e\rangle b\,v(r)\hat{\boldsymbol{r}}=\tfrac{1}{3}\langle e\rangle(r+b\,v'(r))\hat{\boldsymbol{r}},$$

where $v(r)$ is a non-dimensional function of the scalar separation r and $v'(r)$ is the non-dimensional disturbance velocity relative to the imposed expansion flow. Similarly, the pair-distribution function will be spherically symmetric. Scaling all lengths with the excluded-volume radius b, the relative diffusivity with its value at large separations $2D$, where $D=kT/6\pi\eta a$ is the diffusivity of an isolated particle, and time with the diffusive time $b^2/2D$, the non-dimensional pair-evolution equation and boundary conditions become

$$\frac{\partial g}{\partial t}+Pe_b\,r\frac{\partial g}{\partial r}+Pe_b\frac{1}{r^2}\frac{\partial}{\partial r}r^2v'(r\hat{b})g=\frac{1}{r^2}\frac{\partial}{\partial r}r^2G(r\hat{b})\frac{\partial g}{\partial r}, \tag{5.4}$$

$$G(r\hat{b})\frac{\partial g}{\partial r}=Pe_b[2+v'(2\hat{b})]g \quad \text{at} \quad r=2,$$

and

$$g \sim 1 \quad \text{as} \quad r \to \infty.$$

where $G(r\hat{b})$ is the usual scalar function for the radial component of the relative mobility of two particles, i.e. $\boldsymbol{D} = 2D[G(r\hat{b})\hat{\boldsymbol{r}}\hat{\boldsymbol{r}} + H(r\hat{b})(\boldsymbol{I} - \hat{\boldsymbol{r}}\hat{\boldsymbol{r}})]$ (Batchelor 1976). The $r\hat{b}$ argument of G and v' serves as a reminder that the separation distance has been scaled with the excluded-volume radius b, while the tabulated values of the hydrodynamic functions are scaled with the actual particle radius a. The Péclet number is also based on the excluded-volume radius

$$Pe_b = \frac{\frac{1}{3}\langle e \rangle b^2}{2D}.$$

Note that since we can have either positive or negative expansion rates, the Péclet number can likewise be positive or negative.

Equation (5.4) applies for all Péclet numbers and can be used in conjunction with the stresslet expressions below to determine the bulk viscosity far from equilibrium. In this first study, however, we restrict ourselves to the linear response regime for small departures from equilibrium. To leading order in Pe_b we may write

$$g = 1 + Pe_b\, f(r,t),$$

where f satisfies

$$\frac{\partial f}{\partial t} - \frac{1}{r^2}\frac{\partial}{\partial r} r^2 G(r\hat{b}) \frac{\partial f}{\partial r} = -\frac{1}{r^2}\frac{\partial}{\partial r} r^2 v'(r\hat{b}),$$

$$G(r\hat{b})\frac{\partial f}{\partial r} = 2 + v'(2\hat{b}) \quad \text{at} \quad r = 2,$$

and

$$f \sim 0 \quad \text{as} \quad r \to \infty.$$

Clearly, as the expansion flow proceeds the number density of particles will continually decrease (or increase for compression) and the corresponding interparticle interactions will decrease (increase). Nevertheless, it is possible to obtain a steady relative distribution of particle pairs $f(r)$ and therefore steady (apart from number density dependence) contributions to the bulk viscosity.† The steady solution for f can be written simply as the quadrature

$$f(r) = -\int_r^\infty \frac{1}{G(r\hat{b})}\left[\frac{8}{r^2} + v'(r\hat{b})\right] \mathrm{d}r. \tag{5.5}$$

In the absence of hydrodynamic interactions $\hat{b} \to \infty$, $v' = 0$ and the solution for f is

$$f_{NH} = -\frac{8}{r}.$$

This 'monopole' disturbance also applies with hydrodynamic interactions at large enough r since $v' \sim \frac{4}{3}\hat{b}r^{-2}$ and $G \sim 1 + O(1/r)$.

As is typical of advection–diffusion problems in linear flows, the problem for g is singular at distances $r \sim O(Pe_b^{-1/2})$ where the effects of the advection are just as important as those of diffusion. The solution (5.5) for f represents the second term in the 'inner' region, which, fortunately, can be obtained without matching to the

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† In the linear response regime a steady distribution for all expansion rates (positive or negative) is possible. Beyond the linear regime, however, a steady distribution can only be found for compression.

outer region, whose first non-trivial term is $O(Pe_b^{3/2})$. The integrals below for the bulk viscosity are all absolutely convergent and explicit consideration of the outer region is not necessary.

It is interesting to note that for this expansion flow problem the source (or monopole $f \sim 1/r$) forcing leads to an expansion of the form: $g \sim 1 + Pe_b f + O(Pe_b^{3/2})$, while the dipolar ($f \sim 1/r^2$) forcing of the diffusion or microrheology problem (where the outer region is now at $r \sim O(Pe_b^{-1})$, Khair & Brady 2006) proceeds as $g \sim 1 + Pe_b f + Pe_b^2 f_2 + O(Pe_b^3)$, and the quadrupolar ($f \sim 1/r^3$) forcing for the shear viscosity (again the outer region is at $r \sim O(Pe_b^{-1/2})$) behaves as $g \sim 1 + Pe_b f + Pe_b^2 f_2 + O(Pe_b^{5/2})$; in the shear viscosity problem the first three terms in the inner region can be obtained without regard to the outer problem (Brady & Vicic 1995).

The relative velocity $v'(r\hat{b})$ arises from the hydrodynamic interactions between the two particles in the expansion flow. To determine this, we recall that the grand resistance matrix relates the hydrodynamic force (the torque is included in the definition of the force for simplicity of presentation) and stresslet to the velocity (which includes the angular velocity) and rate of strain according to

$$\begin{pmatrix} \boldsymbol{F}^H \\ \boldsymbol{S}^H \end{pmatrix} = -\begin{pmatrix} \boldsymbol{R}_{FU} & \boldsymbol{R}_{FE} \\ \boldsymbol{R}_{SU} & \boldsymbol{R}_{SE} \end{pmatrix} \cdot \begin{pmatrix} \boldsymbol{U} - \boldsymbol{U}^\infty \\ -\boldsymbol{E}^\infty \end{pmatrix}, \tag{5.6}$$

where $\boldsymbol{E}^\infty$ is the rate of strain tensor and the superscript ∞ denotes the imposed flow. Traditionally (Kim & Karrila 1991), only traceless stresslet and rate-of-strain tensors were considered and the elements of the resistance matrices were constructed in such a way as to guarantee this. But this restriction is unnecessary. The grand resistance matrix applies equally well when the stresslet and rate of strain possess a trace; only the elements need to be modified to incorporate these interactions.

Jeffrey, Morris & Brady (1993) computed the trace of the stresslets for two spherical particles in an incompressible Stokes flow, which serves to completely determine $\boldsymbol{R}_{SU}$, and by symmetry $\boldsymbol{R}_{FE}$. Specifically, the trace of the particle stresslets is related to the velocities by

$$\begin{pmatrix} S_1^H \\ S_2^H \end{pmatrix} = -\eta \begin{pmatrix} \boldsymbol{P}_{11} & \boldsymbol{P}_{12} \\ \boldsymbol{P}_{21} & \boldsymbol{P}_{22} \end{pmatrix} \cdot \begin{pmatrix} \boldsymbol{U}_1 - \boldsymbol{U}_1^\infty \\ \boldsymbol{U}_2 - \boldsymbol{U}_2^\infty \end{pmatrix}, \tag{5.7}$$

where

$$\boldsymbol{P}_{\alpha\beta} = 4\pi a^2 X_{\alpha\beta}^P \boldsymbol{d},$$

for identical particles and $\boldsymbol{d} = (\boldsymbol{x}_2 - \boldsymbol{x}_1)/|\boldsymbol{x}_2 - \boldsymbol{x}_1|$.

From (5.6), (5.7) and $\boldsymbol{R}_{FE} = \boldsymbol{R}_{SU}^\dagger$ we have for the relative velocity due to the expansion flow for the force- and torque-free particles

$$v'(r\hat{b}) = \frac{4}{3}\frac{1}{\hat{b}} G(r\hat{b})\left(X_{22}^P - X_{21}^P\right),$$

and the $X_{\alpha\beta}^P$ are evaluated at $r\hat{b}$.

In addition to the trace of the stresslets due to the particle velocities, Jeffrey *et al.* (1993) also computed the trace of the stresslets for an incompressible linear shear flow, which determines the elements of $\boldsymbol{R}_{SE}$. The analogue of (5.7) is $S_\alpha^H = \eta\, \boldsymbol{Q}_{\alpha\beta} \cdot \boldsymbol{E}_\beta^\infty$, with $\boldsymbol{Q}_{\alpha\beta} = 8\pi a^3 X_{\alpha\beta}^Q (\boldsymbol{dd} - \frac{1}{3}\boldsymbol{I})$ for identical particles. For our application, however, we have a compressible flow with $\boldsymbol{E}^\infty = \frac{1}{3}\langle e\rangle \boldsymbol{I}$ and there is an additional term in $\boldsymbol{R}_{SE}$ that was not computed by Jeffrey *et al.* (1993). We have designated this term $T_{\alpha\beta}^Q$, so that the complete Q-function becomes for identical particles

$$\hat{\mathbf{Q}}_{\alpha\beta} = 8\pi a^3 \left[X^Q_{\alpha\beta} \left(\boldsymbol{dd} - \tfrac{1}{3}\boldsymbol{I} \right) + T^Q_{\alpha\beta} \tfrac{1}{3}\boldsymbol{I} \right].$$

The scalar functions $T^Q_{\alpha\beta}$ have been determined for all separations and particles of different radii in Khair, Swaroop & Brady (2006).

Armed with the hydrodynamic functions we can return to the expression for the effective bulk viscosity (3.3) and write to $O(\phi_b^2)$

$$\kappa_{eff} = \left(\kappa + \tfrac{4}{3}\eta\,\phi \right) \frac{1}{1-\phi} + (\kappa^B + \kappa^P + \kappa^E)\phi_b^2,$$

where the three $O(\phi_b^2)$ coefficients correspond to the Brownian, interparticle-force and rate-of-strain contributions to the bulk viscosity. Note that the volume fraction for the two-particle contributions is based on the excluded-volume radius b. The explicit forms for these contributions can be written as

$$\begin{aligned}
\kappa^B &= -\eta \frac{1}{\hat{b}^2} \frac{1}{2} \int_2^\infty \left[\left. \frac{\mathrm{d}}{\mathrm{d}r} \left[X^P_{22} x^a_{22} + X^P_{21} x^a_{21} \right] + \frac{2}{r} \left[X^P_{22} x^a_{22} + X^P_{21} x^a_{21} \right] \right] \right|_{r\hat{b}} f(r) r^2 \,\mathrm{d}r, \\
\kappa^P &= -\eta \frac{1}{\hat{b}} \frac{3}{4} v(2\hat{b}) f(2), \\
\kappa^E &= \eta \frac{1}{\hat{b}^3} 2 \int_2^\infty \left[\left(T^Q_{12} + T^Q_{22} \right)' - \tfrac{1}{4}\hat{b} \left(X^P_{22} - X^P_{21} \right) v'(r\hat{b}) \right] r^2 \,\mathrm{d}r,
\end{aligned}$$

where $x^a_{\alpha\beta}$ are the non-dimensional scalar mobility functions relating velocity to force (Kim & Karrila 1991), all functions are evaluated at $r\hat{b}$ and the prime on $(T^Q_{12} + T^Q_{22})$ is a reminder that the isolated-particle value, (4.1), has been removed.

The two-particle contributions are all positive and are proportional to the shear viscosity η as they arise from the incompressible disturbance flow caused by the bulk expansion. The two ‘thermodynamic’ contributions, κ^B and κ^P, depend on the perturbation to the microstructure caused by the expansion flow, $f(r)$, while the rate-of-strain contribution is by definition linear in $\langle e \rangle$ and therefore to leading order in Pe_b only depends on the equilibrium distribution $g(r) = 1$.

Figure 1 shows the perturbation to the microstructure for the two limiting cases of no hydrodynamic interactions: $\hat{b} \to \infty$, and the case of nearly pure hydrodynamic interactions: $\hat{b} = 1.00001$. The two cases are very similar for all r, and, in particular, both grow as $1/r$ for large r. For intermediate values of $\hat{b}$ the perturbation function lies between these two curves. Evidently hydrodynamic interactions in expansion flows are weak at small Pe_b and have little effect on the microstructure. The situation may be different at high Pe_b where, in analogy to the shear viscosity problem, a boundary layer may form and hydrodynamics be important.

Figure 2 shows the three two-particle contributions to the bulk viscosity and their sum as a function of the ratio of the excluded-volume to actual particle radius $\hat{b} = b/a$. As $\hat{b} \to 1$ the Brownian and rate-of-strain contributions are finite, while the interparticle-force contribution vanishes (as $(\hat{b} - 1)$) because the relative particle velocity is zero on contact at twice the hydrodynamic radius. The limiting values of the $O(\phi_b^2)$ coefficients for pure hydrodynamics, $\hat{b} \equiv 1$, are $\kappa^B = 5.348\eta$ and $\kappa^E = 1.57\eta$. As $\hat{b}$ increases hydrodynamic interactions are reduced and the Brownian and rate-of-strain contributions decrease, while the interparticle-force contribution grows. Once $\hat{b}$ exceeds approximately 1.244, the particles are kept so far apart by the excluded-volume interaction that all two-particle contributions to the bulk viscosity decrease to zero, scaling as: $\kappa^B \sim \hat{b}^{-7}$, $\kappa^P \sim \hat{b}^{-1}$, and $\kappa^E \sim \hat{b}^{-9}$ as $\hat{b} \to \infty$.

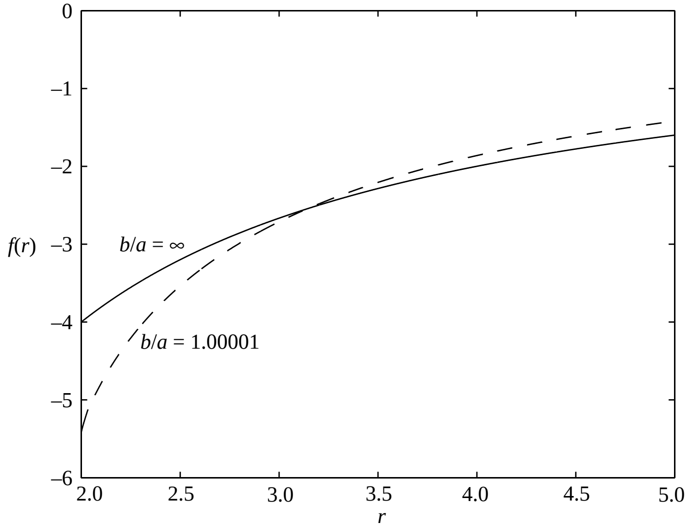

FIGURE 1. Microstructural perturbation function $f(r)$ for $b/a = 1.00001$ (dashed line) and $b/a = \infty$ (solid line). Both curves behave as $1/r$ for large r. For other values of b/a the perturbation f is intermediate between these two curves.

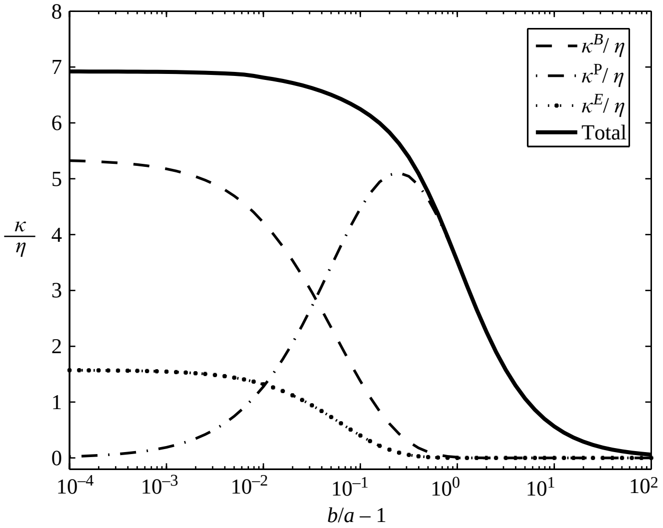

FIGURE 2. The $O(\phi_b^2)$ two-particle contributions to the bulk viscosity: Brownian κ^B/η (dashed line), interparticle force κ^P/η (dot-dashed line), rate of strain κ^E/η (dotted line) and total $(\kappa^B + \kappa^P + \kappa^E)/\eta$ (solid line).

As defined, all two-particle contributions to the bulk viscosity vanish as $\hat{b} \rightarrow \infty$ because the volume fraction has been based on the excluded-volume radius b. When $\hat{b} \rightarrow \infty$ the actual volume fraction ϕ is so small that the particles are isolated and only the single-particle Einstein contribution remains. This same dependence on $\hat{b}$ occurs in the shear viscosity problem (Bergenholtz, Brady & Vicic 2002). Note that the interparticle-force contribution κ^P decays most slowly as $1/\hat{b}$, and reflects the fact that the Péclet number depends on the actual particle diffusivity ($D = kT/6\pi\eta a$), which depends on the hydrodynamic, not thermodynamic, size.

6. Conclusions

In this work we have shown that an effective bulk viscosity can be defined for a suspension of particles undergoing uniform volume expansion. The bulk viscosity relates the deviation of the trace of the macroscopic stress from its equilibrium value to the average rate of expansion in a manner completely analogous to the well-known expressions for the deviatoric stress. Explicit formulae have been given to compute the bulk viscosity for all volume fractions of suspended particles and for all expansion rates. Although we considered steady expansion rates in this work, this restriction is not necessary and an arbitrary time-dependent expansion flow is permissible. The general formulae should also apply to bubbles and drops as well as rigid particles, and we have explicitly recovered G. I. Taylor's result for expanding bubbles.

The fact that the particles cannot expand with the fluid creates a disturbance motion which dissipates energy and increases the bulk viscosity of the suspension. A repulsive interparticle force decreases the bulk viscosity as particles are prevented from coming close to each other where hydrodynamic lubrication interactions are strong. Because we scaled the volume fraction with the excluded-volume radius b the interparticle-force contribution to the bulk viscosity κ^P decreases as (a/b). What is important, however, is the product $\kappa^P\phi_b^2 \sim (b/a)^5\eta\phi^2$, which is seen to grow as the range of the repulsive force is increased when the actual volume fraction is used.

In this study we have only considered the linear-response regime of small Péclet number where the departure of the microstructure from equilibrium is proportional to Pe_b. In this limit all three contributions to the bulk stress are finite and contribute comparable amounts (depending, of course, on the range of the repulsive force $\hat{b}$) to the bulk viscosity. Beyond the linear-response regime it is not known how the bulk viscosity varies with Pe_b. Preliminary results indicate that in compression in the absence of hydrodynamic interactions the bulk viscosity (κ^P) decreases with decreasing Pe_b (recall that the Péclet number is signed and negative in compression) until a plateau is reached as $Pe_b \rightarrow -\infty$, analogous to the shear thinning of the shear viscosity (Bergenholtz *et al.* 2002). Whether the inclusion of hydrodynamic interactions ($\hat{b} \rightarrow 1$) leads to 'thickening' of the bulk viscosity as it does for the shear viscosity is not known. Also, preliminary results indicate that in expansion (positive Pe_b) beyond the linear response regime there is no steady solution for $g(r)$. The implications of this for the bulk viscosity are not known.

We were able to obtain analytical results for dilute suspensions where pair interactions are sufficient. Higher volume fractions can be readily studied by adapting the Stokesian Dynamics paradigm (Brady & Bossis 1988; Sierou & Brady 2001) to allow for a uniform rate of expansion. In molecular fluids the bulk viscosity is a rapidly increasing function of volume fraction analogous to the shear viscosity (Sigurgeirsson & Heyes 2003) and we should anticipate similar behaviour here. It should be appreciated that during the course of expansion the volume fraction of particles continually decreases (or increases to close packing in compression) and thus some care is needed in defining the bulk viscosity for a given volume fraction. (This care is not needed in the linear-response regime considered here; work on molecular fluids has been limited to linear response.) If the time scale for the evolution of the microstructure is the same as that for the number density, which will be the case at large Péclet number, then a steady bulk viscosity will not exist. Also, in application a suspension will probably be undergoing shear in addition to expansion and it may be more fruitful to consider the effects of a weak expansion rate (relative to the shear rate) on the suspension microstructure and the resulting bulk viscosity. It is most likely that this property is what is needed in two-phase flow modelling, although this is as yet an unexplored area.

As a final remark, in this paper we have considered low-Reynolds-number flows only. While the general expression for the average stress can easily be modified to include inertial effects (of both particles and fluid), the decomposition of the fluid velocity into a uniform expansion and an incompressible disturbance flow is not as straightforward and further care is needed to pose a well-defined spatially homogeneous microscale problem from which the bulk viscosity can be determined when inertia is important. And, of course, there is the question of how to measure the bulk viscosity experimentally. The fact that the particles give a contribution proportional to the shear viscosity may make this feasible.

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