Non-continuum anomalies in the apparent viscosity experienced by a test sphere moving through an otherwise quiescent suspension.

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PACS numbers: 83.10, 47.55

Abstract

A comparison is made of the 'apparent viscosity' (as defined by Stokes law) between two different cases of a test sphere moving slowly through an unbounded, otherwise quiescent, globally homogeneous, dilute suspension of identical, neutrally-buoyant spherical particles dispersed in an incompressible Newtonian liquid. In case I the force on the test sphere is maintained constant for all time (and the torque-free sphere allowed to rotate) – corresponding to the so-called "falling ball" case – and its instantaneous velocity allowed to vary with proximity to each suspended sphere encountered during its trajectory; in case II the non-rotating test sphere is towed with a uniform (instantaneous) velocity through the suspension and the force experienced by it allowed to vary with proximity to each suspended sphere. Allowing for two-body hydrodynamic interactions between the ball and a suspended particle, the ensemble-average velocity of the test sphere is calculated in case I and ensemble-average force in case II, and Stokes law used to calculate the apparent viscosity of the suspension from the ensemble-averaged. linear force/velocity ratio obtained. In each case the 'apparent suspension viscosity' coefficient attains, as expected, the limiting, continuum, Einstein value of 2.5 when the test sphere is much larger than the freely suspended particle. However, in the case of disparate relative sizes, the apparent viscosity is found to be significantly larger in case II than in case I. The difference arises from the locally inhomogeneous nature of the suspension and points up a fundamental non-continuum aspect of suspension behavior above and beyond the expected test/suspended-sphere size ratio 'Knudsen' non-continuum effect.

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1 Introduction

The dynamics of a test sphere ('ball') moving through an otherwise quiescent suspension composed of identical, neutrally-buoyant, freely-suspended spherical particles uniformly dispersed (at least globally) throughout a Newtonian fluid has been studied both theoretically and experimentally. Batchelor¹ and Batchelor & Wen² analyzed the problem within the broader context of dilute bidisperse suspensions, whereas Davis & Hill³ calculated, in addition to the settling velocity, the variance and the hydrodynamic diffusivity coefficients. Brenner et al.⁴ obtained the disturbance velocity field far from the ball in the limiting case where the ball size greatly exceeds that of the suspended spheres. In the non-dilute range, Mondy et al.⁵ and Miliken et al.⁶ performed falling-ball rheometry experiments for various particle concentrations, ball sizes, suspended-sphere sizes, etc.

The current work focuses on a completely different aspect of the problem – namely, a comparison between the values obtained for the suspension's 'Stokes-law viscosity' as experienced by a test sphere for two opposite, but seemingly equivalent cases. The first is that observed heretofore in falling-ball experiments and studied theoretically, in which the force on the ball is a fixed, time-independent constant, and the net torque vanishes. In the second case the ball is forced to move with a fixed time-independent velocity while rotary motion is prohibited. [A device which enables the realization of such an experiment has already been utilized to measure the viscosity of semi-dilute polymer solutions (Adam & Delsanti,⁷ Adam et al.⁸).] Were a suspension truly equivalent to a homogeneous Newtonian fluid, no difference would exist between the two cases – even allowing for 'Knudsen-like' non-continuum,

'slip-velocity' size effects [the latter analogous to the classical Cunningham⁹ mean-free path correction factor to Stokes law used in Millikan's¹⁰ classical oil drop experiments for measuring the fundamental electronic charge].

A similar point has recently been made (cf. Almog & Brenner¹¹), but in the context of two opposite rotating test sphere cases (both in unbounded suspensions): (I) a ball rotating with a fixed, time-independent angular velocity; and (II) a rotating ball upon which a prescribed, time-independent couple is exerted. Those results showed that the 'apparent viscosities' (this time calculated through Kirchoff's law for the linear couple/angular-velocity relation) are different, and hence, that the suspension does not behave like a homogeneous medium. But in order to calculate the 'apparent viscosity' in the rotating ball case it was necessary to make an ad hoc assumption of uniformity of the spatial distribution of suspended spheres near the rotating test sphere – equivalent to assuming uniformity of the pair probability density function ('Eisenschitz hypothesis'). This assumption can, however, only be justified for the case of Brownian particles. On the other hand, for the current translating ball problem one can calculate the pair probablity density for non-Brownian particles without invoking any similar a priori assumption. As such, the subsquent analysis demonstrates the manifestly inhomogeneous nature of suspension behavior for a different case, but without requiring any assumption regarding the pair probability density.

In section 2, which follows, we formulate the relevant problems posed for both cases. Section 3 presents results for the probability densities and subsequently for the 'apparent viscosities'. Unlike previous investigations,^{2,3} in which numerical results were presented only for several discrete values of the ball/suspended-sphere size ratio, the dependence on this important parameter is presented here over a domain for which its values vary continously from very small to very large. Appendix A pursues some technical details pertaining to the calculations performed in section 3.

2 Formulation of the problems

Consider a dilute suspension of identical, freely-suspended spherical particles of radii a_2 dispersed in a homogeneous Newtonian fluid of viscosity μ through which a test sphere of radius a_1 translates. If the size of the container is much larger than the sizes of both the suspended and test spheres, it is reasonable to approximate results by those obtained for an unbounded domain.

We shall subsquently compare between two different cases:

- 1. I. The force on the ball is prescribed (and a zero net torque prescribed as well which allows the ball to rotate).
- 2. II. The velocity of the ball is prescribed (and its angular velocity is zero).

In case I we shall be interested in the average reduction in the mean 'sedimentation' velocity of the test sphere due to hydrodynamic interactions, whereas in case II we shall calculate the average extra-force exerted upon it, also due to hydrodynamic interactions. In both situations the base case for calculating the incremental changes is that in which all of the other physical parameters of the problem remain unchanged, except that the suspended particles are now absent.

Suppose that the (center of the) test sphere is located at x_1 . Then any ensemble-averaged quantity may be expressed (Rubinstein & Keller¹²) in the form

$$\langle \boldsymbol{A} \rangle (\boldsymbol{x}_1) = \int \boldsymbol{A}(\boldsymbol{x}_1, \boldsymbol{x}_2, \dots, \boldsymbol{x}_N) f_N(\boldsymbol{x}_2, \dots, \boldsymbol{x}_N/\boldsymbol{x}_1) d\boldsymbol{x}_2 \dots d\boldsymbol{x}_N ,$$
 (2.1)

where f_N is the multiparticle conditional probability density, and $A(x_1, x_2, ..., x_N)$ is any configuration-dependent tensor. Since the suspension is supposed dilute, the multisphere average can be approximated by a two-sphere average (Batchelor,¹ Almog & Brenner¹¹). The average reduction in sedimentation velocity for case I can therefore be approximated by

$$\left\langle \Delta \boldsymbol{U}_{1} \right\rangle (\boldsymbol{x}_{1}) = -n(\boldsymbol{x}_{1}) \int \left[\boldsymbol{U}_{1}(\boldsymbol{x}_{1}), \boldsymbol{x}_{2} \right) - \frac{\boldsymbol{F}}{6\pi\mu a_{1}} \right] p_{12}^{F}(\boldsymbol{x}_{2}/\boldsymbol{x}_{1}) d\boldsymbol{x}_{2}, \qquad (2.2)$$

and the average extra-force for case II by

$$\left\langle \Delta \boldsymbol{F}_{1} \right\rangle (\boldsymbol{x}_{1}) = n(\boldsymbol{x}_{1}) \int \left[\boldsymbol{F}_{1}(\boldsymbol{x}_{1}), \boldsymbol{x}_{2} \right) - 6\pi \mu a_{1} \boldsymbol{U} \right] p_{12}^{U}(\boldsymbol{x}_{2}/\boldsymbol{x}_{1}) dx_{2}.$$
(2.3)

In the above, $n(\boldsymbol{x}_1)$ is the local number density of the freely-suspended particles. The functions p_{12}^F and p_{12}^U respectively denote the pair probability densities for finding a neutrally bouyant sphere at \boldsymbol{x}_2 when the test sphere is located at \boldsymbol{x}_1 for cases I and II. The constant vectors \boldsymbol{F} and \boldsymbol{U} are, respectively, the prescribed force in case I and the prescribed velocity in case II. The two-point vector fields $\boldsymbol{U}_1(\boldsymbol{x}_1, \boldsymbol{x}_2)$ and $\boldsymbol{F}_1(\boldsymbol{x}_1, \boldsymbol{x}_2)$ are, respectively, the velocity (case I) of the ball and the force (case II) exerted upon it when a freely-suspended sphere is present at x_2 .

For case I the configuration-dependent sedimentation velocity can be represented in terms of Jeffrey & Onishi's¹³ mobility functions as

$$\boldsymbol{U}_{1}(\boldsymbol{x}_{1}, \boldsymbol{x}_{2}) = \boldsymbol{U}_{s} \cdot \left[x_{11}^{a} \frac{\boldsymbol{r}\boldsymbol{r}}{r^{2}} + y_{11}^{a} (\boldsymbol{I} - \frac{\boldsymbol{r}\boldsymbol{r}}{r^{2}}) \right] .$$
(2.4)

Similarly, for case II the configuration-dependent force exerted upon the ball is expressible in the form

$$\boldsymbol{F}_{1}(\boldsymbol{x}_{1},\boldsymbol{x}_{2}) = 6\pi\mu a_{1}\boldsymbol{U} \cdot \left[\frac{1}{x_{11}^{a}}\frac{\boldsymbol{r}\boldsymbol{r}}{r^{2}} + \frac{y_{11}^{c}}{y_{11}^{a}y_{11}^{c} - 3(y_{11}^{b})^{2}}(\boldsymbol{I} - \frac{\boldsymbol{r}\boldsymbol{r}}{r^{2}})\right].$$
(2.5)

Here, $U_s = F/6\pi\mu a_1$ is the Stokes-law settling velocity of the ball; $r = x_2 - x_1$ $(r = |\mathbf{r}|)$; and x_{ij}^a , y_{ij}^a , y_{ij}^b and y_{ij}^c are Jeffrey & Onishi's¹³ mobility coefficients, which are expressed as functions of the dimensionless parameters

$$s = \frac{r}{a_1 + a_2} \qquad \qquad ; \qquad \qquad \lambda = \frac{a_2}{a_1}$$

In circumstances where the gap between the ball and suspended sphere is small compared their center-to-center spacing, it proves useful to introduce the normalized gap width

$$\xi = s - 2 \ll 1.$$

as the separation parameter in place of s.

The pair probability densities both satisfy the Fokker-Planck equation (Batchelor,¹ Davis & Hill³)

$$\frac{\partial p_{12}}{\partial t} + \boldsymbol{\nabla} \cdot (p_{12}\boldsymbol{v}_{12}) = 0 \quad , \tag{2.6}$$

accompanied by the boundary condition $\lim_{r\to\infty} p_{12} = 1$. (An appropriate superscript, F or U, should be added depending on the specific case.) The quantity \boldsymbol{v}_{12} stands for the relative velocity of the neutrally buoyant sphere at \boldsymbol{x}_2 with respect to the ball at \boldsymbol{x}_1 . For case I,

$$\boldsymbol{v}_{12}^{F} = -\boldsymbol{U}_{s} \cdot \left[L^{F} \frac{\boldsymbol{rr}}{r^{2}} + M^{F} \left(\boldsymbol{I} - \frac{\boldsymbol{rr}}{r^{2}} \right) \right] , \qquad (2.7)$$

(Davis & Hill³), where the hydrodynamic coefficients L^F and M^F are expressible in the respective forms

$$L^F = x_{11}^a - \frac{2}{1+\lambda} x_{12}^a$$
; $M^F = y_{11}^a - \frac{2}{1+\lambda} y_{12}^a$. (2.8a,b)

Similarly, for case II we have

$$\boldsymbol{v}_{12}^{U} = -\boldsymbol{U} \cdot \left[L^{U} \frac{\boldsymbol{rr}}{r^{2}} + M^{U} \left(\boldsymbol{I} - \frac{\boldsymbol{rr}}{r^{2}} \right) \right] , \qquad (2.9a,b)$$

wherein

$$L^{U} = \frac{L^{F}}{x_{11}^{a}} \qquad ; \qquad M^{U} = \frac{y_{11}^{c}M^{F} - 3y_{11}^{b}B}{y_{11}^{a}y_{11}^{c} - 3(y_{11}^{b})^{2}}$$
(2.10a,b)

in which $B = y_{11}^b - 4/(1+\lambda)^2 y_{12}^b$. As will be subsquently demonstrated (cf. Appendix A), in order to approximate p_{12}^U for $\xi \ll 1$ it is more useful to re-express M^U in terms of Jeffrey & Onishi's¹³ resistance functions:

$$M^{U} = \frac{3Y_{22}^{C} \left[Y_{22}^{A} + \frac{1}{2} \left(1 + \frac{1}{\lambda}\right) Y_{12}^{A}\right] - Y_{22}^{B} \left[Y_{22}^{B} + \frac{1}{4} \left(1 + \frac{1}{\lambda}\right)^{2} Y_{21}^{B}\right]}{(Y_{22}^{B})^{2} - 3Y_{22}^{A} Y_{22}^{C}}.$$
 (2.11)

3 Results and Discussion

3.1 The pair probablity distribution

In both cases the pair probability distribution, depends solely on the radial coordinate, and can be easily obtained in the following form: (Batchelor,¹ Davis & Hill³)

$$p_{12}^{F} = \frac{1}{L^{F}} \exp\left\{\int_{s}^{\infty} \frac{2}{s'} \left(1 - \frac{M^{F}}{L^{F}}\right) ds'\right\},$$
(3.1a)

$$p_{12}^{U} = \frac{1}{L^{U}} \exp\left\{\int_{s}^{\infty} \frac{2}{s'} \left(1 - \frac{M^{U}}{L^{U}}\right) ds'\right\}.$$
 (3.1b)

For s > 2.5, both p_{12}^F and p_{12}^U have been obtained in the form of power series in s up to $O(s^{-300})$, the coefficients of these terms having been derived through recurrence relations (cf. Appendix A) from Jeffrey & Onishi's¹³ series expansions for the mobility functions.

For $\xi \ll 1$ it is necessary to use near-field expansions for the hydrodynamic coefficients, namely

$$L^{F} = L_{1}^{F}\xi + L_{2}^{F}\xi^{2}\ln\xi + L_{3}^{F}\xi^{2} + O(\xi^{3}\ln\xi), \qquad (3.2a)$$

$$M^{F} = M_{0}^{F} + \frac{M_{1}^{F} \ln \xi^{-1} + M_{2}^{F}}{\ln^{2} \xi^{-1} + e_{1} \ln \xi^{-1} + e_{2}} + O(\xi \ln \xi) = M_{0}^{F} + \frac{\alpha_{1}^{F}}{\ln \xi^{-1} - z_{1}^{F}} + \frac{\alpha_{2}^{F}}{\ln \xi^{-1} - z_{2}^{F}},$$
(3.2b)

$$L^{U} = \frac{L_{1}^{F}\xi + L_{2}^{F}\xi\ln\xi + L_{3}^{F}\xi^{2}}{d_{11}^{1} + d_{11}^{2}\xi + d_{11}^{3}\xi^{2}\ln\xi + d_{11}^{4}\xi^{2}} + O(\xi^{3}\ln\xi),$$
(3.2c)

$$M^{U} = \frac{M_{1}^{U} \ln \xi^{-1} + M_{2}^{U}}{\ln^{2} \xi^{-1} + e_{1} = \ln \xi^{-1} + e_{2}} + O(\xi \ln \xi) = \frac{\alpha_{1}^{U}}{\ln \xi^{-1} - z_{1}^{U}} + \frac{\alpha_{2}^{U}}{\ln \xi^{-1} - z_{2}^{U}}.$$
 (3.2d)

The various coefficients required in (3.2) can easily be derived by using Jeffrey & Onishi's¹³ near-field expansions for the mobility and resistance functions in conjunction with (2.8), (2.10) and (2.11).

Upon substituting into (3.1) the expressions for M^F and M^U as well as the leading-order

terms for L^F and L^U , we obtain

$$p_{12}^F \cong \frac{q^F}{L^F} \xi^x (\ln \xi^{-1} - z_1^F)^{\beta_1^F} (\ln \xi^{-1} - z_2^F)^{\beta_2^F}, \qquad (3.3a)$$

$$p_{12}^U \cong \frac{q^U}{L^U} (\ln \xi^{-1} - z_1^U)^{\beta_1^U} (\ln \xi^{-1} - z_2^U)^{\beta_2^U}, \qquad (3.3b)$$

where $x = M_0^F/L_1^F$, $\beta_i^F = -\alpha_i^F/L_1^F$ and $\beta_i^U = -\alpha_i^U/L_1^F d_{11}^1$. The constants q^F and q^U have to be determined by numerical integration of (3.1a) and (3.1b), respectively, at a single point $\bar{\xi} \ll 1$. Batchelor & Wen² obtained an approximation for p_{12}^F by expanding M^F in power series in $\ln \xi^{-1}$ and truncating the expansion after the first 2 terms. Their approximation is therefore accurate up to $O(1/\ln \xi)$, unlike (3.3a) which is accurate up to $O(\xi)$.

Note that p_{12}^U tends to infinity faster than p_{12}^F . This follows from the fact that unlike M^F , $\lim_{\xi \to 0} M^U = 0$; equivalently, this may be regarded as a consequence of the fact that case II boundary conditions (as opposed to the zero net torque condition for case I) do not allow the touching pair of sphere (for $\xi \to 0$) to rotate together.

For s < 2.5 it is still possible to express p_{12}^F and p_{12}^U as power series in s. However, in view of (3.3), the series becomes ineffective for $\xi \ll 1$. Instead, we therefore expand the functions $p_{12}^F L^F \tilde{\xi}^{-x} (\ln \tilde{\xi}^{-1} - z_1^F)^{-\beta_1^F} (\ln \tilde{\xi}^{-1} - z_2^F)^{-\beta_2^F}$ and $p_{12}^U L^U (\ln \tilde{\xi}^{-1} - z_1^U)^{-\beta_1^U} (\ln \tilde{\xi}^{-1} - z_2^U)^{-\beta_2^U}$ (cf. Appendix A). The variable $\tilde{\xi} = \xi/(1 + \gamma \xi)$ is used in place of ξ to make these functions analytic as $s \to \infty$. The resulting series have been found generally to accord well with the numerical integration of (3.1), the explicit value of γ chosen being without significant effect on the results.

Figure 1 displays the dependence on ξ of p_{12}^F (fig. 1a) and p_{12}^U (fig. 1b) on a logarithmic

scale for $\lambda = 0.01$, $\lambda = 1$ and $\lambda = 100$. The curves displayed for both functions derive from the series expansion for s < 2.5. For all values of λ , the asymptotic approximation (3.3) becomes identical (less than 0.1 percent error) with the preceding series expansion for those values of $\xi < 10^{-6}$.

For $\xi \sim O(1)$ it can be seen that $p_{12}^F - 1$ decays much faster for $\lambda = 0.01$ than for the two other cases. The far-field expansion

$$p_{12}^F = 1 + \frac{30\lambda^3}{s^4(1+\lambda)^4} + O(s^{-5})$$
(3.4)

can partially explain this result, since for large s, and λ less than 3, p_{12}^F is an increasing function of λ .

Comparison of the curves for $\lambda = 1$ and $\lambda = 100$ in fig. 1a shows that for $\xi \sim O(1)$, $p_{12}^{F}|_{\lambda=100} < p_{12}^{F}|_{\lambda=1}$, whereas for $\xi \to 0$ the function $p_{12}^{F}|_{\lambda=100}$ tends to infinity much faster. The first difference results from the fact that for $\lambda = 100$, hydrodynamic interactions between the two spheres – which are governed by a_1/r – are still small for $\xi \sim O(1)$. The second difference results from the diminution in the rate of the rotation of touching spheres when λ is either very large or very small; explicitly, this is a consequence of the fact that $\lim_{\lambda \to 0 \text{ or } \infty} M_0^F =$ 0. For $\lambda \ll 1$ the latter statement is physically obvious: a small neutrally buoyant sphere has a minor effect on a much larger test sphere. In the other limit, if both U_s and a_2 are kept constant, and a_1 allowed to tend towards zero, the force on the ball together with M_0^F decay as well. In fact, Davis & Hill³ show $M_0^F \cong 3/4\lambda$.

To explain the steeper behaviour of p_{12}^F near $\xi = 0$, one needs to show that $\lim_{\lambda \to 0 \text{ or } \infty} x = 0$.

For $\lambda \gg 1$ this stronger result can easily be demonstrated. Using the relation (Jeffrey & Onishi¹³)

$$L_1^F(\lambda) + L_1^F\left(\frac{1}{\lambda}\right) = \frac{(1+\lambda)^3}{2\lambda},\tag{3.5}$$

in conjunction with the fact that $\lim_{\lambda \to 0} L_1^F(\lambda) = 0$ [since when $\lambda \to 0$, the velocity field for the two-sphere problem tends uniformly to that for a single sphere, for which $\partial u_R / \partial R|_{R=a_1} = 0$, where u_R is the radial velocity component (Happel & Brenner¹⁴)], we obtain $L_1^F \sim \lambda/2 + 3/2 + O(1/\lambda)$ as $\lambda \to \infty$. Upon combining the results for M_0^F and L_1^F we obtain $\lim_{\lambda \to \infty} x = 0$, where x is defined following (3.3).

For $\lambda \to 0$, similar behavior is observed for x; equivalently, M_0^F decays faster than does L_1^F . Hence, it is expected that for some value of $\xi < 10^{-12}$, the $\lambda = 0.01$ curve will intersect the $\lambda = 1$ curve; in other words, $p_{12}^F|_{\lambda=0.01}$ will become larger than $p_{12}^F|_{\lambda=1}$.

Figure 1b supports the latter statement. Upon observing that the $\lambda = 0.01$ curves in both figures are almost identical, it may be argued that for $\lambda \ll 1$ the problems for cases I and II (for which x = 0 for all λ) become equivalent. Since in this limit the suspension may be regarded as a continuum (cf. Almog & Brenner¹¹), this result is almost certainly expected.

Comparison of the $\lambda = 0.01$ and $\lambda = 100$ curves in fig. 1b for $\xi \sim O(1)$ yields the surprising result that $p_{12}^U|_{\lambda=100} < p_{12}^U|_{\lambda=0.01}$. This has been confirmed analytically by establishing the asymptotic behavior for $\lambda \gg 1$ of the hydrodynamic coefficients d_{11}^1 and d_{11}^2 appearing in (3.2c). First it is easy to show, using the fact that two touching spheres move as a rigid body (Jeffrey & Onishi¹³), that

$$d_{11}^1(\lambda) = \lambda d_{11}^1\left(\frac{1}{\lambda}\right) \quad .$$

Obviously, $d_{11}^1 \to 1$ when $\lambda \to 0$, and thus $d_{11}^1 \sim 1/\lambda$ for $\lambda \gg 1$. In contrast to the decay of d_{11}^1 , it is easy to show that $d_{11}^2 \sim \lambda/2$ in view of the asymptotic behaviour of L_1^F .

If one substitutes the above approximation into (3.2c), expands in terms of $1/\lambda$, and assumes that the higher-order terms in (3.2c) are negligible, it follows for $\xi \sim O(\lambda^{-1})$ that $L^U \sim 1$. The latter result, which – in view of (3.3b) – immediately confirms the large difference between the $\lambda = 0.01$ and $\lambda = 100$ curves, may be intuitively explained. Suppose that the test sphere, which is much smaller than the neutrally buoyant sphere, is located at a distance several times larger than its radius from the larger, neutrally buoyant one, and that its prescribed velocity U is parallel to r. Then, the relative velocity will be almost equal to U (since the larger, neutrally buoyant sphere will be negligibly affected) so long as $\xi \sim O(\lambda^{-1})$. If, however, we instead prescribe the force, the relative velocity will decay much faster, since the effect of the larger sphere on the smaller one will be much more important in that case.

3.2 The suspension's 'apparent viscosity'

Our goal in this subsection is to obtain the average reduction in sedimentation velocity for case I, and the average extra-force for case II. Towards these ends we introduce nondimensional apparent viscosity coefficients, k_m^F and k_m^U , which are defined by the following equations

$$\langle \boldsymbol{U} \rangle = \frac{\boldsymbol{U}_s}{1 + k_m^F c} \qquad , \qquad \langle \boldsymbol{F} \rangle = \frac{6\pi\mu a_1 \boldsymbol{U}}{1 + k_m^U c} , \qquad (3.6a,b)$$

with c the volume fraction of suspended spheres. The quantities $k_m^F c$ and $k_m^U c$ represent the additional apparent viscosities for the respective cases (cf. Miliken et al.⁶). Neither, however, constitutes an intrinsinc property of the suspension since, as will be demonstrated subsquently, each depends upon the ratio λ of the respective size of the suspended particles to that of the test sphere.

Upon substituting (2.4) together with (3.1a) into (2.2) we obtain

$$k_m^F = \frac{(1+\lambda)^3}{8\lambda^3} \int_2^\infty (3 - x_{11}^a - 2y_{11}^a) p_{12}^F s^2 ds \quad . \tag{3.7}$$

Similarly, substitution of (2.5) together with (3.1b) into (2.3) yields

$$k_m^U = -\frac{(1+\lambda)^3}{8\lambda^3} \int_2^\infty \left(3 - \frac{1}{x_{11}^a} - 2\frac{y_{11}^c}{y_{11}^a y_{11}^c - 3(y_{11}^b)^2}\right) p_{12}^U s^2 ds \quad . \tag{3.8}$$

Obviously, the difference between the respective values of k_m results from two different factors:

- (i) Different pair probability densities.
- (*ii*) Different averaged quantities; that is, in order to evaluate k_m^F we average the reduction in sedimentation velocity, whereas k_m^U arises from evaluating the average extra force.

In order to evaluate (3.7) and (3.8) it is first necessary to evaluate the pair probability densities as well as the various hydrodynamic coefficients appearing in the integrands. The hydrodynamic coefficients have been obtained for $\xi > 0.01$ by using their far-field expansions [once again up to $O(s^{-300})$] and for $\xi < 0.01$ by their near-field expansions (cf. Jeffrey & Onishi¹³). The pair probability densities have been obtained for s > 2.5 as power series in s; for $10^{-6} < \xi < 0.5$ the power series expansions of $p_{12}^F L^F \tilde{\xi}^{-x} (\ln \tilde{\xi}^{-1} - z_1^F)^{-\beta_1^F} (\ln \tilde{\xi}^{-1} - z_2^F)^{-\beta_2^F}$ and $p_{12}^U L^U (\ln \tilde{\xi}^{-1} - z_1^U)^{-\beta_1^U} (\ln \tilde{\xi}^{-1} - z_2^U)^{-\beta_2^U}$ have been utilized, whereas for $\xi < 10^{-6}$ Eqs.(3.3a) and (3.3b) have been used.

For $\xi \sim O(1)$ it is possible to obtain the power series representations in s of the integrands appearing in both (3.7) and (3.8), followed by term-by-term integration to effect the quadrature. This process was utilized for s > 2.5. For $10^{-6} < \xi < 0.5$ the integrals were calculated numerically using the Gauss-Concord rule (a Fortran Nag library routine). Finally, for $\xi < 10^{-6}$, the integrands in both (3.7) and (3.8) were respectively expanded into power series in $(\ln \xi^{-1} - z_1^F)$ and $(\ln \xi^{-1} - z_1^U)$; (we assume $|z_1^F| > |z_2^F|$ and $|z_1^U| > |z_2^U|$). Term-by-term integration then yields

$$\int_{0}^{10^{-6}} (3 - x_{11}^{a} - 2y_{11}^{a}) p_{12}^{F} s^{2} ds \approx \frac{e^{-xz_{1}^{F}} 4q^{F}}{x(D - z_{1}^{F})^{y^{F}} L_{1}^{F}} \cdot \sum_{0}^{\infty} \left[E^{F} + \frac{A^{F}}{z_{1}^{F} - z_{2}^{F}} \frac{n}{1 + \beta_{2}^{F} - n} + \frac{B^{F}}{z_{1}^{F} - z_{2}^{F}} \frac{n}{\beta_{2}^{F}} \right] \binom{\beta_{2}^{F}}{n} \cdot [x(D - z_{1}^{F})]^{(y^{F} + n)} \Gamma(1 - y^{F} - n, x(D - z_{1}^{F})) \left[\frac{z_{1}^{F} - z_{2}^{F}}{D - z_{1}^{F}} \right]^{n}$$
(3.9)

and

$$\int_{0}^{10^{-6}} \left(3 - \frac{1}{x_{11}^{a}} - 2\frac{y_{11}^{c}}{y_{11}^{a}y_{11}^{c} - 3(y_{11}^{b})^{2}}\right) p_{12}^{U}s^{2}ds \cong \frac{4q^{U}d_{11}^{1}}{L_{1}^{F}} \cdot \sum_{0}^{\infty} \left[E^{U}\frac{y^{U} + n}{y^{U} + n - 1} + A^{U} + B^{U}\left(1 - \frac{n}{\beta_{2}^{U}}\right)\right] \binom{\beta_{2}^{U}}{n} \frac{(D - z_{1}^{U})^{-y^{U}}}{-(y^{U} + n)} \left[\frac{z_{1}^{U} - z_{2}^{U}}{D - z_{1}^{U}}\right]^{n} (3.10)$$

In the above, $D = \ln 10^6$, $y^F = \beta_1^F + \beta_2^F$, $y^U = \beta_1^U + \beta_2^U$, $\Gamma(\alpha, x)$ is the incomplete gamma function (Abramowitz & Stegun¹⁵), and A^F , B^F , E^F , A^U, B^U and E^U are defined in the following near-field expansions for the hydrodynamic coefficients:

$$3 - x_{11}^{a} - 2y_{11}^{a} \cong E^{F} + \frac{A^{F}}{\ln\xi^{-1} - z_{1}^{F}} + \frac{B^{F}}{\ln\xi^{-1} - z_{2}^{F}} + O(\xi \ln\xi^{-1}), \qquad (3.11a)$$

$$3 - \frac{1}{x_{11}^{a}} - 2\frac{y_{11}^{c}}{y_{11}^{a}y_{11}^{c} - 3(y_{11}^{b})^{2}} \cong E^{U} + \frac{A^{U}}{\ln\xi^{-1} - z_{1}^{U}} + \frac{B^{U}}{\ln\xi^{-1} - z_{2}^{U}} + O(\xi \ln\xi^{-1}). \qquad (3.11b)$$

Figure 2 displays the variation with λ of the parameters k_m^F and k_m^U (the incremental apparent viscosities) on a logarithmic scale. The numerical error is estimated to be less than one percent for $0.1 < \lambda < 10$. As λ tends to either 0 or ∞ , the domain in which the near-field expansions for the hydrodynamic coefficients are valid shrinks (since we must have $\xi \ll 1/\lambda$ as $\lambda \to \infty$ or $\xi \ll \lambda$ as $\lambda \to 0$), whereas the domain in which the far-field expansion is valid remains unchanged. Of course, this effects the accuracy of the numerical calculations of (3.7) and (3.8). However, the contributions to the additional apparent viscosity of both the near field for $\lambda \ll 1$ and far field for $\lambda \gg 1$ have been found to be negligible – both numerically

and (as will be subsquently explained) analytically. Consequently, the calculation appears accurate over the whole range of λ values for which the calculations of k_m^F and k_m^U have been performed.

Three highly intuitive modes of behavior can be observed:

- 1. For $\lambda \to 0$, both k_m^F and k_m^U tend to 2.5, which is exactly Einstein's classical result. Obviously, when the test sphere is much larger than the suspended spheres it becomes a part of the boundary, and the suspension behaves like a true continuum (cf. Almog & Brenner¹¹). It is for this reason that the near-field contribution in that limit is expected to be negligible.
- 2. As $\lambda \to \infty$, both k_m^F and k_m^U tend to infinity as a consequence of the steepening of the probability densities p_{12}^F and p_{12}^U near $\xi = 0$; in other words, the test particle spends most of its time in close vicinity to one of the large, neutrally buoyant spheres. This offers an intuitive explanation of the negligible far-field effect in that limit.
- 3. For all values of λ , the inequality $k_m^U \ge k_m^F$ obtains; in words, the apparent viscosity increment is larger for case II. The main reason for this appears to be the steeper behavior of p_{12}^U near $\xi = 0$; equivalently, a touching pair of spheres cannot rotate like a rigid body.

Less intuitive are the maximum and the minimum respectively attained by k_m^F at $\lambda = 0.1$ $(k_m^F = 2.68)$, and $\lambda = 2$ $(k_m^F = 2.47)$. Unlike the problem of a rotating sphere in an unbounded suspension (cf. Almog & Brenner¹¹), there exist two competing mechanisms which effect the average sedimentation velocity: The first is that the configuration-dependent reduction in sedimentation velocity (averaged over a spherical surface and properly normalized), namely $(1 + 1/\lambda)^3(3 - x_{11}^a - 2y_{11}^a)$, decays much faster with ξ as λ increases. This statement is supported by the asymptotic expansions of Fuentes et al.^{16,17} for large and small values of λ , from which the following approximations can be easily derived:

$$\frac{(1+\lambda)^3}{\lambda^3}(3-x_{11}^a-2y_{11}^a) \cong \frac{15}{4}\left(\frac{2}{s}\right)^4 + O(s^{-6},\lambda s^{-4}) \text{ as } \lambda \to 0,$$
(3.12a)

$$\frac{(1+\lambda)^3}{\lambda^3}(3-x_{11}^a-2y_{11}^a) \cong \frac{15}{4\lambda}\left(\frac{2}{s}\right)^4 + O(\lambda^{-1}s^{-6},\lambda^{-2}s^{-4}) \text{ as } \lambda \to \infty, \quad (3.12b)$$

which reveal that the far-field contribution in the limit $\lambda \gg 1$ is much smaller than in the opposite limit, $\lambda \ll 1$. Intuitively, one can argue that a small sedimenting sphere is affected by a much smaller neighborhood than is a large sphere.

The second mechanism is that of the relatively rapid decay of p_{12}^F with ξ for small values of λ , arising from the fact that the relative velocity of the ball with respect to the suspended sphere is almost the Stokes-law velocity, even for $\lambda \ll \xi \ll 1$. Clearly, the maximum and the minimum attained by k_m^F result from the interaction of these competing mechanisms. For k_m^U , however, the second mechanism appears to be always dominant in view of the monotone behavior manifested.

As already mentioned, the major contribution to the apparent viscosity in the limit $\lambda \to \infty$ is due to near-field effects. However, even a very weak diffusivity would drastically decrease the near-field contribution. This can perhaps explain the monotone decrease in apparent viscosity observed in the experiments of Miliken et al.⁶ It is to be expected,

however, that in a dilute suspension – for which the interparticle diffusivity is expected to be very small – an increase in the apparent viscosity would be observed. (As already noted by Davis & Hill³ such data are not currently available in the literature).

Acknowledgements

Y.A. wishes to thank the Fulbright Scholar Program for their support during the initial period of this research. H.B. was supported by a grant from the office of Basic Energy Sciences of the U.S. Department of Energy. Both authors wish to thank Professor D. J. Jeffrey for allowing them public access to his two-sphere numeric Fortran files.

A Calculation of the pair probability densities

Subsequent calculations will be confined to p_{12}^F since the comparable procedure for obtaining p_{12}^U is virtually identical. We evaluate p_{12}^F using three different methods, each valid in a different range of values of ξ . Explicitly, the far-field expansion is valid for s > 2.5, the intermediate expansion for $10^{-6} < \xi < 0.5$, and the near-field expansion for $\xi < 10^{-6}$.

(i) Far-field expansion range, s > 2.5: For $\xi \sim O(1)$ the hydrodynamic coefficients M^F and L^F can be efficiently calculated by expanding them into power series in 2/s:

$$L^{F} = \sum_{n=0}^{\infty} f_{n}^{x}(\lambda) \left[\frac{2}{(1+\lambda)s}\right]^{n} \qquad ; \qquad M^{F} = \sum_{n=0}^{\infty} f_{n}^{y}(\lambda) \left[\frac{2}{(1+\lambda)s}\right]^{n},$$
(A.1a,b)

wherein the functions $f_n^x(\lambda)$ and $f_n^y(\lambda)$ are given by Jeffrey & Onishi.¹³ From (A.1) it is easy

to deduce a similar series expansion for the integrand in (3.1a), namely

$$\frac{\frac{d}{ds}(p_{12}^F L^F)}{p_{12}^F L^F} = -\frac{2}{s} \left(1 - \frac{M^F}{L^F} \right) = \sum_{n=1}^{\infty} g_n(\lambda) \left[\frac{2}{(1+\lambda)s} \right]^n, \tag{A.2}$$

where the g_n 's are obtainable through the recurrence relation

$$g_n(\lambda) = (1+\lambda)[\bar{g}_{n-1}(\lambda) - \delta_{1n}] \qquad (n \ge 1)$$
(A.3a)

$$\bar{g}_n(\lambda) = f_n^y(\lambda) - \sum_{k=1}^n \bar{g}_{n-k}(\lambda) f_k^x(\lambda) \qquad (n \ge 0).$$
(A.3b)

By assigning to $p_{12}^F {\cal L}^F$ the expansion

$$p_{12}^F L^F = \sum_{n=1}^{\infty} h_n(\lambda) \left[\frac{2}{(1+\lambda)s} \right]^n \quad , \tag{A.4}$$

we obtain the following recurrence relation upon substitution of the latter into (A.2):

$$h_n(\lambda) = -\frac{2}{1+\lambda} \frac{1}{n} \sum_{k=1}^n g_{k+1}(\lambda) h_{n-k}(\lambda),$$
 (A.5)

together with the initial condition $h_n(0) = 1$, resulting from the boundary condition $\lim_{s\to\infty} p_{12}^F L^F = 1$. The series expansion for p_{12}^F can now be easily obtained from (A.1) and (A.4).

(ii) Intermediate expansion range, $10^{-6} < \xi < 0.5$: In view of (3.3a) we expand in this

intermediate domain the function

$$f = p_{12}^F L^F \tilde{\xi}^{-x} (\ln \tilde{\xi}^{-1} - z_1^F)^{-\beta_1^F} (\ln \tilde{\xi}^{-1} - z_2^F)^{-\beta_2^F} ,$$

where $\tilde{\xi} = \xi/(1 + \gamma \xi)$. To this end we firs notice that

$$\frac{\frac{df}{ds}}{f} = -\frac{2}{s} \left(1 - \frac{M^F}{L^F} \right) - \left[\frac{1}{\xi} - \frac{\gamma}{1 + \gamma\xi} \right] \left[x - \frac{\beta_1^F}{\ln\xi^{-1} - z_1^F} - \frac{\beta_2^F}{\ln\xi^{-1} - z_2^F} \right]$$
(A.6)

Since all the terms in (A.6) are expandable as power series of 2/s (so long as $\ln \gamma > z_i^F$) it is easy to derive from it the series expansion for $\frac{df}{ds}/f$ from which the expansion for f can be readily obtained.

(iii) Near-field expansion range, $\xi < 10^{-6}$: The near-field approximation (3.3a) is obtained in a rather straightforwad manner from (3.1a) and (3.2).

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Figure captions

- Figure 1 Variation of the probability densities p_{12}^F (a) and p_{12}^U (b) with the normalized gap width ξ between the spheres for different values of the suspended/test-sphere radius ratio λ .
- Figure 2 Dependence of the apparent viscosity coefficients k_m^F and k_m^U , normalized by a 5/2 factor, on the suspended/test-sphere size ratio λ . Note that in the limit $\lambda \to 0$ of a large ball they both attain the limiting Einstein value of 2.5.