Hindered Settling of Semidilute Monodisperse and Polydisperse Suspensions

A light-extinction principle was used to measure the fall speeds of the interfaces that develop during gravity sedimentation of monodisperse, bidisperse, and tridisperse suspensions of noncolloidal particles with small particle Reynolds numbers in the semidilute total particle volume fraction range $0.0003 \leq c_0 \leq 0.15$. For monodisperse suspensions, the hindered settling velocity of the interface at the top of the suspension was found to be well represented by the correlation of Richardson and Zaki, provided that the isolated particle fall speed was chosen by linear extrapolation of the data to $c_0 = 0$, with 90% confidence intervals on the exponent of $n = 5.0 \pm 0.1$. For dilute monodisperse, bidisperse, and tridisperse suspensions, the hindered settling velocities of the interfaces showed agreement within experimental uncertainty with the theory of Batchelor, which predicts that the settling velocity decreases linearly with increasing particle concentration and which is based upon pairwise particle interactions.

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Introduction

The sedimentation of small particle forms the basis for common means of separating particles from fluid, separating particles with different settling velocities from one another, and sizing particles. Much of the fundamental research in this area has focused on determining the hindered settling function for monodisperse suspensions, $f(c)$, defined such that the statistical average particle setting velocity relative to the bulk suspension velocity, which is zero for batch settling, is given by $v = uf(c)$, where u is the settling velocity of an isolated particle and c is the local volume fraction of particles in the suspension. There is considerable uncertainty on the quantitative functional form of $f(c)$, especially in the dilute limit, where most correlations and theories yield one of the following two forms for $f(c)$:

$$f(c) \sim 1 - \alpha c \tag{1}$$

$$f(c) \sim 1 - \beta c^{1/3} \tag{2}$$

Which of these two functional forms is expected to hold depends upon the microstructure of the sedimenting suspension, as described by Davis and Acrivos (1985).

A common method of determining $f(c)$ experimentally is to measure the fall velocity of the interface at the top of the suspension by visual means as it descends, at several different concentrations. One difficulty in using this method for dilute suspensions is that the fall velocity of the interface at the top of the suspension is ambiguous because the interface does not remain sharp but rather becomes increasingly diffuse as the sedimentation proceeds. A second difficulty in determining $f(c)$ experimentally is that its departure from unity for dilute suspensions is small. Therefore, not only an accurate measurement of the mean settling velocity of particles in the suspension is needed, but an accurate assessment of the velocity of an isolated particle is required as well. One possibility is to predict this velocity using Stokes' law, which is valid for an isolated spherical particle settling due to gravity through a quiescent Newtonian fluid in a large container under conditions of small particle Reynolds number,

$$u = \frac{2}{9}\frac{(\rho_s - \rho)}{\mu} a^2 g \tag{3}$$

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Unfortunately, due to particle polydispersity and the inaccuracies in measuring the particle and fluid properties, the uncertainty in the predicted Stokes velocity is generally on the order of 5–10%. As reported by Oliver (1961), similar uncertainties result if Stokes velocities are determined by direct observation of the fall speed of isolated particles.

The research described in this paper uses an improved experimental technique for determining the hindered settling function for nearly monodisperse suspensions of spheres at low Reynolds numbers and at semidilute concentrations ($c \leq 0.15$). This technique involves passing a thin horizontal slit of light through the suspension in order to measure the particle volume fraction at any location within the diffuse interface at the top of the sedimenting suspension; it therefore is able to measure the settling velocity to a higher degree of accuracy than earlier methods based on visual observation. A second thrust is the adaption of the experimental technique to bidisperse and tridisperse suspensions.

Experimental Materials and Methods

Experiments were carried out by introducing weighed amounts of fluid and particles into a rectangular vertical glass vessel that was immersed in a glass-walled water bath controlled at a temperature of 25.0°C. The initial or bulk volume fractions of the particles in the experiments were varied throughout the range $0.0003 \leq c_0 \leq 0.15$. After allowing the system to reach thermal equilibrium, a plunger was used to mix the suspension. The suspension sedimented past two narrow horizontal slits of light (1.0 cm wide by 0.06 cm high) each produced by passing a 2 mW He-Ne laser beam through a set of cylindrical lenses. The ligth transmitted through the vessel was focused onto a photodiode, and the voltage signal proportional to its intensity was recorded as a function of time on a strip chart. For each suspension and volume fraction, the experiments were repeated so that typically four to six measurements of the interface were made for each value of h (where h is the vertical distance of the light slit below the liquid/air interface), and three to six different values of h were examined.

As the interface at the top of the suspension fell past each light slit, there was a continuous change in the light absorbance from its maximum value, which corresponds to the light absorbed by the bulk suspension with particle volume fraction $c = c_0$, to a value of zero, which corresponds to the light passing through pure fluid. According to the Lambert-Beer law, the light absorbance is proportional to the particle volume fraction for dilute suspensions:

$$A \equiv \log_{10} (I_\infty/I) = kc \tag{4}$$

where $A(t)$ is the light absorbance at time t, $I(t)$ is the intensity of light transmitted through the suspension of particle volume fraction $c(t)$, I_∞ is the intensity of light passing through pure fluid, and k is an extinction coefficient that is proportional to the path length of light through the suspension and is inversely proportional to the average particle diameter for opaque particles (Herdan, 1960). For a polydisperse suspension containing N discrete particle species, the Lambert-Beer law is modified to become

$$A = \sum_{i=1}^{N} k_i c_i \tag{5}$$

where c_i is the volume fraction of particles of species i in the light beam. Equations 4 and 5 are valid only if the particle volume fraction is sufficiently low that multiple scattering effects are negligible. Calibrations performed with the suspensions described in this paper showed that the Lambert-Beer law became invalid when the particle concentration exceeded about 2 vol. % unless the refractive indices of the particles and fluid were matched. Thus, for higher concentrations, fluid mixtures were chosen that provided the best possible refractive index match with the particles. A perfect match was not possible owing to the presence of small air bubbles in the particles, thereby limiting the validity of the Lambert-Beer law for the matched refractive index suspensions to particle concentrations of 10–15 vol. % or less.

Several sizes of rigid spherical glass and plastic particles were used in the experiments; their properties are given in Table 1. The glass beads were class V microbeads (Ferro Corporation), and the acrylic beads were Lucite 47G (DuPont). The Newtonian fluids used in the experiments were hydrocarbons composed of UCON 50HB-280X (Union Carbide) and Monsanto HB40; the properties of the pure fluids and mixtures are given in Table 2.

For the glass spheres of types I and III in pure UCON 50HB-280X, Stokes velocity determinations were made by introducing a single sphere with an eyedropper into the vessel and then timing it as it fell the distance between the two slits of laser light. The particle flashed as it fell through each light slit, thus facilitating the timing. The experiment was repeated with 21 different particles of type I and 55 different particles of type III.

Results and Discussion

A typical example of the raw transmitted light intensity v. time data is shown in Figure 1. The noise in the raw data resulted from statistical fluctuations in the number of particles in the beam. A computer-aided smoothing procedure was applied to the raw data; the result is also shown in Figure 1. As is evident from the shape of the curve, considerable spreading of the interface occurred. We define the median velocity, $v_{1/2}$, of

Table 1. Properties of Particles Used in Experiments

Particle Type	Material	U.S. Std. Sieve Range	Median Dia. 10^{-6} m	Std. Dev 10^{-6} m	Density kg/m^3	Refract. Index
I	Glass	−100 + 120	136	4.5	2.49×10^3	1.51
II	Glass	−70 + 80	186	9.6	2.49×10^3	1.51
III	Glass	−50 + 60	261	7.0	2.49×10^3	1.51
IV	Acrylic	−100 + 120	135	14	1.185×10^3	1.49
V	Acrylic	−80 + 100	161	12	1.185×10^3	1.49

Table 2. Properties of Fluids Used in Experiments

Fluid Mixture	Composition wt. %	Temp.	Viscosity kg/m · s	Density kg/m^3	Refract. Index
—	100% UCON 50HB-280X	25°C	0.113	1.034×10^3	1.455
—	100% Monsanto HB40	25°C	0.072	1.006×10^3	1.570
I	49% UCON 50HB-280X 51% Monsanto HB40	25°C	0.086	1.015×10^3	1.517
II	70% UCON 50HB-280X 30% Monsanto HB40	25°C	0.098	1.027×10^3	1.493

the interface as the average velocity over the distance h of the isoconcentration plane where $c/c_0 = 1/2$. For a slightly polydisperse suspension, this median velocity is predicted to be virtually identical to the hindered settling velocity at the same bulk volume fraction c_0 of a monodisperse suspension with all particles having a size equal to the median particle size of the polydisperse suspension, regardless of the value of c_0 (Davis and Hassen, 1987). When $c = c_0/2$, the corresponding transmitted light intensity, $I_{1/2}$, is given by the geometric mean between the light intensity transmitted through the bulk suspension, I_0, and that for the clear fluid, I_∞:

$$I_{1/2} = (I_0 I_\infty)^{1/2} \tag{6}$$

Equation 6 follows from Eq. 4 with a constant value for the light extinction coefficient, k. Since the extinction coefficient is actually inversely proportional to the average particle diameter in the light beam, its value changes slightly with time for a sedimenting suspension with a distribution of particle sizes. In order to correct for this effect, we applied the analysis described by Davis and Hassen, which yields a correction of less than 2% in the measured interface fall speeds for the nearly monodisperse suspensions described in this paper.

In Figure 2, the hindered settling velocity curve, $v_{1/2}$ v. c_0, is given for monodisperse suspensions of glass beads of types I and III in fluid mixture I. The data points in this and subsequent graphs represent averages over all measurements made at a given initial volume fraction, and the error bars are plus and minus one standard deviation. The solid lines are least-squares

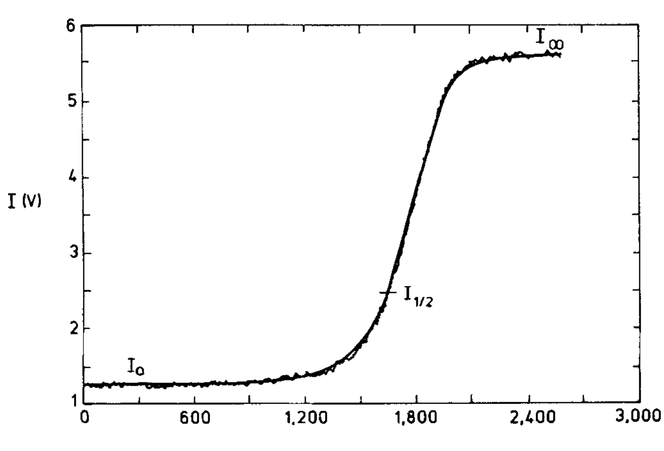

Figure 1. Transmitted light intensity v. time raw and smoothed data for a nearly monodisperse suspension.

best-fit regressions of the Richardson-Zaki (1954) correlation form:

$$v_{1/2} = u(1 - c_0)^n \tag{7}$$

The 90% confidence intervals on the best-fit values are $u = (1.63 \pm 0.02) \times 10^{-4}$ m/s and $n = 4.9 \pm 0.1$ for particles of type I, and $u = (6.10 \pm 0.05) \times 10^{-4}$ m/s and $n = 5.1 \pm 0.1$ for particles of type III. Clearly, this correlation provides an excellent fit of the data. For dilute suspensions ($c_0 \leq 0.03$), a linear correlation of the form given by Eq. 1 may be tested. The resulting best-fit values and 90% confidence intervals are $u = (1.66 \pm 0.02) \times 10^{-4}$ m/s and $\alpha = 5.4 \pm 0.5$ for particles of type I, and $u = (6.2 \pm 0.1) \times 10^{-4}$ m/s and $\alpha = 5.8 \pm 0.2$ for particles of type III. Hindered settling experiments were also carried out with acrylic beads of type V in fluid mixture II over the concentration range of $0.01 \leq c_0 \leq 0.10$. These data are well fitted by the Richardson-Zaki correlation given by Eq. 7, with 90% confidence intervals of $u = (2.26 \pm 0.04) \times 10^{-5}$ m/s and $n = 4.7 \pm 0.4$. Combining the three sets of data yields a Richardson-Zaki exponent of $n = 5.0 \pm 0.1$ at the 90% confidence level, which is not statistically different from the result for any single data set. This exponent is also in good agreement with values reported previously in the literature, such as $n = 5.1$ at $Re \rightarrow 0$ given by Garside and Al-Dibouni (1977), and which are based on considerable data at higher concentrations. In the dilute limit, this correlation and exponent yields $f(c_0) \sim 1\text{–}5.0\ c_0$, which is of the form represented by Eq. 1, with $\alpha = 5.0$. This linear form provides a good fit of our data over the range $0.005 \leq c_0 \leq 0.03$, although the slopes of the best-fit lines are slightly higher and in agreement within experimental uncertainty with the theoretical value of $\alpha = 5.6$ reported by Batchelor and Wen (1982). This result also agrees well with the findings of Chen and Schachman (1955), Kops-Werkhoven and Fijnaut (1981), Buscall et al. (1982), and Tackie et al. (1983), who used much smaller micron and submicron uniform colloidal spheres. Their observed values of α were in the range of 4 to 6, and the linear form of the hindered settling function was accurate provided c_0 was less than about 0.08. This range of accuracy is greater than what we observed, as may be seen from Figure 2. Note that the original theory of Batchelor (1972) gave $\alpha = 6.55$ for a dilute monodisperse suspension of randomly distributed hard spheres. However, as discussed by Batchelor and Wen (1982), when spheres are different in size by a small fraction such that the Peclet number based upon their relative Stokes velocity is large, then $\alpha = 5.6$ is the predicted result. The appropriate Peclet numbers for the nearly monodisperse suspensions described in this paper are on the order of 10^7–10^9.

In order to more carefully examine the hindered settling

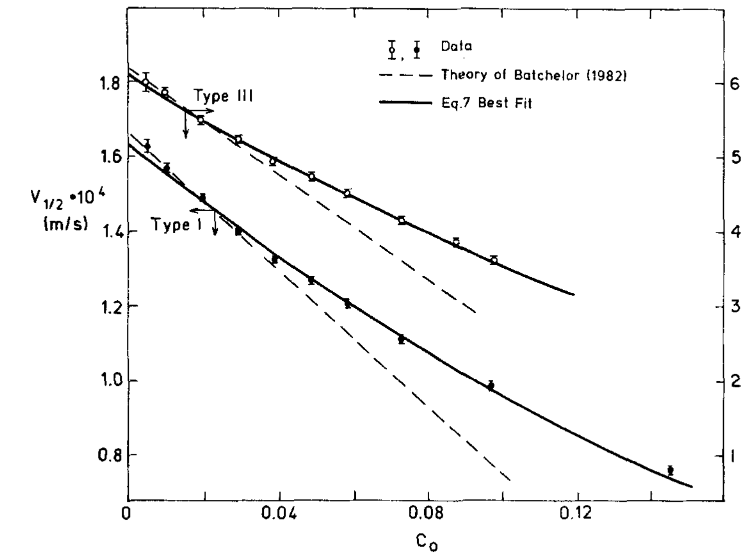

Figure 2. Hindered settling data for nearly monodisperse particles of types I and III in fluid mixture I.

behavior for very dilute suspensions, experiments were performed with pure UCON 50HB-280X. In Figure 3, the hindered settling data are shown for particles of types I and III. For these very dilute suspensions, a linear hindered settling function given by Eq. 1 again provided a good fit of the data, although a larger value of $\alpha = 7.7$ gave the best fit for the two sets of data combined. Correlations of the form represented by Eq. 2 provided nearly as good a fit of these data, although with values of $\beta = 0.80$ (type I) and $\beta = 0.45$ (type III) that are significantly different from each other and that are lower than other values reported in the literature by Barnea and Mizrahi (1973) and Lynch and Herbolzheimer (1983). Also, the best-fit intercept values for correlations of this form are higher than the calculated Stokes velocities and the corresponding intercept values for the correlations that are linear in c_0, as shown in Table 3.

The major difficulty in experimentally establishing the form of the hindered settling function at very dilute concentrations is that independent determinations of the Stokes settling velocity, such as from using Stokes' law or performing tedious single-sphere velocity measurements, are often not accurate to better than about 5–10% uncertainty. On the other hand, careful measurements of the fall speed of the interface at the top of a sedimenting suspension, such as those described in this paper, are accurate to within about 1% uncertainty. Extrapolating these measurements to zero particle concentration then provides a more accurate determination of the Stokes settling velocity. However, as shown in Figure 3, the result may be dependent on the form chosen for extrapolation of the settling velocity v. concentration data. In the future, more accurate Stokes velocity determinations may be possible by using ultramonodisperse suspensions of spheres with well-defined radii. Such particles are currently available in very small sizes (a few microns, or less) and in very large sizes (a few millimeters, or greater), whereas particles of intermediate size are usually made with broad size distributions and then sieved to give more narrow distributions, such as those used in this work. We also note that the microscale structure of the relative particle locations that would form in an ultramonodisperse suspension may be different from that of the slightly polydisperse suspensions used in this work, thereby leading to a change in the hindered settling function.

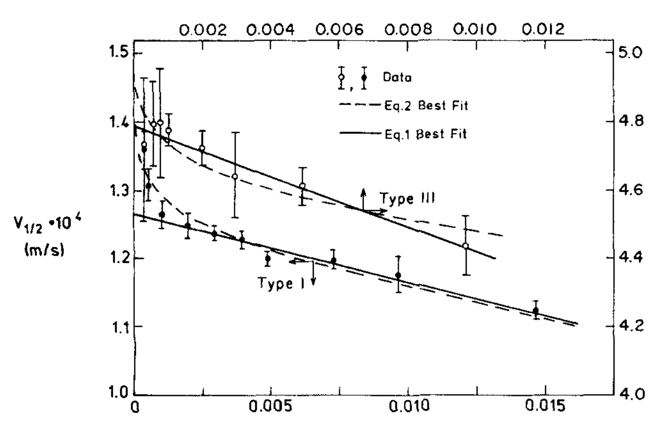

Figure 3. Hindered settling data for nearly monodisperse particles of types I and III in UCON 50HB-280X fluid at dilute concentrations.

A sedimenting bidisperse suspension divides into two regions, with the lower region containing both particle species and the upper region containing only the slower-settling species. The light extinction method may be used to analyze bidisperse suspensions in a similar manner to that for monodisperse suspensions, except that the transmitted light intensity increases in two steps corresponding to the interior and upper interfaces falling past the light beam. In Figure 4 the median velocities of the interior and upper interfaces are plotted v. initial total particle volume fraction, c_0, for a bidisperse mixture of particle types I and III at equal initial volume fractions, suspended in a new batch of fluid mixture I, which has a viscosity 0.083 kg/m · s. The solid and dashed lines represent the theory of Batchelor (1982) for the median velocity of the lower interface, $v^{(1)}_{1/2}$, and the median velocity of the upper interface, $v^{(2)}_{1/2}$, respectively. Batchelor's theory is based upon pairwise particle interactions, thus being limited to dilute suspensions, and states that the hindered settling velocity of a particle of species i in a suspension of

Table 3. Velocity Determinations for Four Different Methods

Suspension	$u(1)$ m/s	$u^{(2)}$ m/s	$u^{(3)}$ m/s	$u^{(4)}$ m/s
Particle type I in UCON 50HB-280X	1.29×10^{-4}	1.27×10^{-4}(−2%)	1.40×10^{-4}(+8%)	1.23×10^{-4}(−5%)
Particle type III in UCON 50HB-280X	4.79×10^{-4}	4.78×10^{-4}(−0.2%)	4.96×10^{-4}(+4%)	5.03×10^{-4}(+5%)

(1) Calculated from Stokes' law using Eq. 3 and properties in Tables 1 and 2
(2) Best-fit extrapolation of sedimentation data to $c_0 = 0$ using Eq. 1
(3) Best-fit extrapolation of sedimentation data to $c_0 = 0$ using Eq. 2
(4) Median of single-sphere velocity measurements

N discrete particle species is given by

$$v_i = u_i\left\{1 + \sum_{j=1}^{N} S_{ij}c_j\right\} \tag{8}$$

where u_i is the Stokes velocity of species i, c_j is the local volume fraction of species j, and the S_{ij} values are sedimentation coefficients that depend upon the radius ratio, $\lambda \equiv a_j/a_i$, and the relative density ratio, $\gamma \equiv (\rho_j - \rho)/(\rho_i - \rho)$, plus any Brownian or interparticle forces that might be significant. Values of the sedimentation coefficients have been computed by Batchelor and Wen (1982), and they are negative under most conditions. Using the appropriate values of these coefficients for the suspension reported in Figure 4, Eq. 8 becomes $v_{1/2}^{(1)} = u_1(1 - 5.6 \cdot c_1^{(1)} - 4.3\, c_2^{(1)})$ for the lower interface, and $v_{1/2}^{(2)} = u_2(1 - 5.6 \cdot c_2^{(2)})$ for the upper interface. Superscript (1) refers to the lower region in the suspension and superscript (2) refers to the upper region in the suspension; similarly, subscripts 1 and 2 refer to the larger and smaller particles, respectively. The volume fractions of both particle species in the lower region of the suspension are equal to their initial values, i.e. $c_1^{(1)} = c_2^{(1)} = c_0/2$. However, the volume fraction of the slower-settling species in the upper region of the suspension, $c_2^{(2)}$, is not equal to $c_2^{(1)}$ except in the dilute limit. This is a consequence of the particle flux continuity requirement for species 2 crossing the interior interface. As described by Smith (1966), this requirement is equivalent to a jump condition:

$$c_2^{(2)}(v_1^{(1)} - v_2^{(2)}) = c_2^{(1)}(v_1^{(1)} - v_2^{(1)}) \tag{9}$$

Because $v_2^{(1)} < v_2^{(2)}$ as a consequence of hindered settling then, $(v_1^{(1)} - v_2^{(1)}) > (v_1^{(1)} - v_2^{(2)})$, which leads to the result that $c_2^{(2)} > c_2^{(1)}$. In Figure 5, $c_2^{(2)}$ is plotted as a function of $c_2^{(1)}$ for the same suspension as depicted in Figure 4. The fact that the predicted fall velocity of the upper interface becomes nonlinear as the initial volume fraction is increased (see dashed line, Figure 4) is a direct consequence of $c_2^{(2)} > c_2^{(1)}$.

Bidisperse sedimentation experiments were also carried out for equal initial concentrations of glass particles of types I and III in pure UCON 50HB-280X at very low concentrations ($c_0 \leq 0.01$). The data for the velocities of the upper and lower inter-

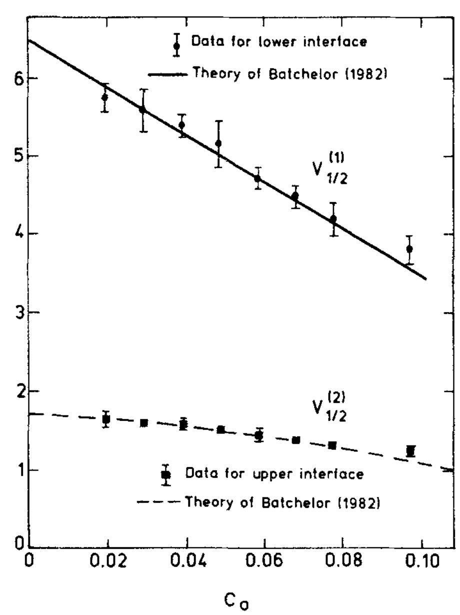

Figure 4. Bidisperse hindered settling data for particles of types I and III at equal concentrations in fluid mixture I.

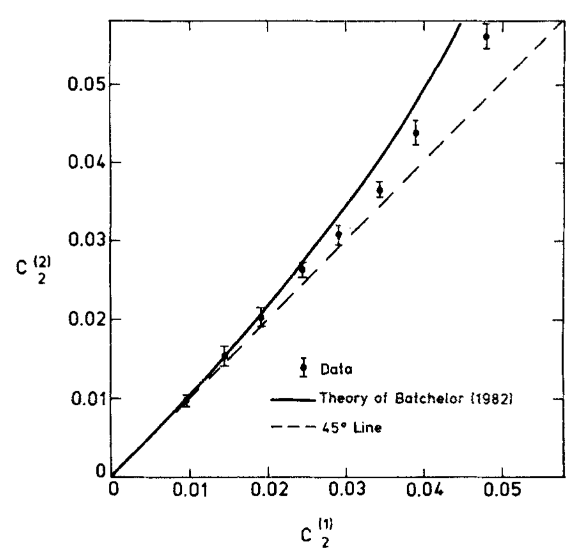

Figure 5. Particle concentrations.

Concentration of slower-settling particles in upper region v. the concentration of the same particles in lower region during bidisperse sedimentation of particles of types I and III at equal concentrations in fluid mixture I

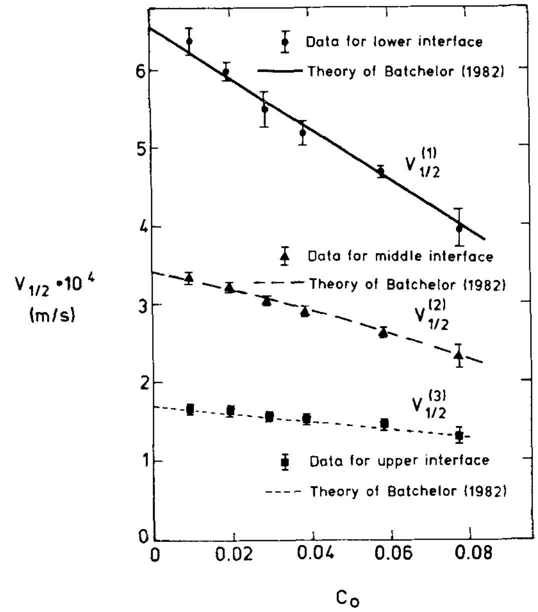

Figure 6. Tridisperse hindered settling data for particles of types I, II, and III at equal concentrations in fluid mixture I.

faces were found to agree well with the theory of Batchelor (1982). The best-fit intercepts of $u_1 = 4.87 \times 10^{-4}$ m/s and $u_2 = 1.28 \times 10^{-4}$ m/s are not significantly different from those for monodisperse experiments of the individual particle species in the same fluid.

Tridisperse experiments were undertaken for equal initial concentrations of glass particles of types I, II, and III in fluid mixture I over the total initial particle volume fraction range $0.01 \leq c_0 \leq 0.08$. The hindered settling velocities $v^{(1)}_{1/2}$, $v^{(2)}_{1/2}$, and $v^{(3)}_{1/2}$ of the lower, middle, and upper interfaces, respectively, are plotted v. c_0 in Figure 6. Using the appropriate sedimentation coefficients found by Batchelor and Wen (1982), the theoretical predictions for the interface fall speeds are $v^{(1)}_{1/2} = u_1(1 - 5.6 \cdot c^{(1)}_1 - 4.7\, c^{(1)}_2 - 4.3\, c^{(1)}_3)$; $v^{(2)}_{1/2} = u_2(1 - 5.6\, c^{(2)}_2 - 4.7\, c^{(2)}_3)$; and $v^{(3)}_{1/2} = u_3(1 - 5.6\, c^{(3)}_3)$. The concentrations in the lower region of the suspension were equal to the initial concentrations, $c^{(1)}_1 = c^{(1)}_2 = c^{(1)}_3 = c_0/3$, whereas the concentrations in the middle and upper regions of the suspension were found from particle flux continuity requirements equivalent to Eq. 9, as described by Birdsell (1985). As shown in Figures 4 and 6, our experimental results for bidisperse and tridisperse suspensions show measured fall speeds of interfaces both at the top of the suspension and between two adjacent regions internal to the suspension that are in excellent agreement with the theory of Batchelor (1982) as given by Eq. 8 and using the predicted sedimentation coefficients of Batchelor and Wen (1982). The agreement is within experimental uncertainty up to $c_0 = 0.08$, which is greater than the range of validity of Batchelor's theory of $c_0 \leq 0.03$ observed for monodisperse suspensions.

Finally, experiments designed to directly measure sedimentation coefficients were performed using bidisperse suspensions in which the concentration of faster-settling particles (species 1) was held fixed at $c^{(1)}_1 = 0.01$ and the concentration of slower-settling particles (species 2) was varied. From Eq. 8, the velocity of the interior interface predicted to be $v^{(1)}_{1/2} = u_1(1 + S_{11}c^{(1)}_1 + S_{12}c^{(1)}_2)$. Noting that $c^{(1)}_1 = 0.01$ and that $S_{11} = -5.6$ from the theory and experiments for nearly monodisperse suspensions, then the slope and intercept values for the linear regression of the data yield a measured value of S_{12}. Including the results of nearly monodisperse experiments, measurements of sedimentation coefficients were made for three cases: $\gamma = 1.0$ with $\lambda \rightarrow 1.0$ (nearly monodisperse); $\gamma = 1.0$ with $\lambda = 0.5$; and $\gamma = 0.11$ with $\lambda = 1.0$, all for noncolloidal particles with Peclet numbers much greater than unity. The resulting 90% confidence intervals on the sedimentation coefficients agreed in all cases with the values predicted by Batchelor and Wen (1982) of $S_{ij} = -5.6$, -4.3, and -2.5, respectively. (Recall, however, that the magnitude of S_{ij} was slightly greater for nearly monodisperse suspensions at very low concentrations, as seen in Figure 3.) Although our three measurements cover only a small subset of all possible particle size and relative density ratios, their verification of the predictions provides confidence in using the theory for other mixtures.

Acknowledgment

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Notation

- a = sphere radius
- A = light absorbance
- c = particle volume fraction
- c_0 = total initial particle volume fraction
- $f(c)$ = hindered settling function
- g = gravitational acceleration constant
- h = distance of light beam below top of fluid
- I = intensity of transmitted light
- I_0 = intensity of light transmitted through bulk suspension
- I_∞ = intensity of light transmitted through pure fluid
- k = light extinction coefficient
- n = exponent in Richardson-Zaki correlation
- Re = particle Reynolds number, $\rho v a/\mu$
- S_{ij} = sedimentation coefficient
- t = time from start of sedimentation
- u = Stokes settling velocity of an isolated particle
- v = hindered settling velocity

Greek letters

- α = hindered settling constant, Eq. 1
- β = hindered settling constant, Eq. 2
- γ = relative density ratio $(\rho_j - \rho)/(\rho_i - \rho)$
- λ = radius ratio, a_j/a_i
- μ = fluid viscosity
- ρ = fluid density
- ρ_s = solid density

Subscripts and superscripts

- i, j = particle species, numbered successively starting with the fastest settling species
- (i) = region or interface in interior of the suspension, number successively starting with lowest region or interface in the suspension
- $1/2$ = median location in interface at which concentration is half-way between that below the interface and that above the interface

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