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### The Sedimentation of Bimodal Distributions of Unflocculated Microspheres

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## THE SEDIMENTATION OF BIMODAL DISTRIBUTIONS OF UNFLOCCULATED MICROSPHERES

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### ABSTRACT

An improved analysis for sedimentation of binary mixtures (two particle sizes) is proposed and tested with data that include differences in both the particle sizes and densities and sedimentation in both the viscous region and the transitions region, between viscous and turbulent (inertial flow). The new analysis can be used for any ratio of particle sizes even when the particle sizes are relatively close. It has been successfully tested beyond the viscous region and with variations in particle density as well as particle size. Although one previous analysis has been successful in predicting most earlier binary data with particles of different size, the new analysis is believed to be better supported by physical principles and thus more reliably extrapolated to different conditions.

### INTRODUCTION

Sedimentation plays a major role in a number of process industries, and the scientific and engineering literature is filled with numerous studies of sedimentation from various viewpoints. The classic problem of Coe and Clevenger (1) of designing clarifiers and thickeners for a wide variety of feed slurries still does not have a good and general solution.

Mirza and Richardson (2) proposed a theoretical framework based on a material balance between settling particles and upflow of liquid displaced by the settling particles. They were unable to predict bimodal settling rates accurately without the aid of an additional totally empirical parameter.

This work extends the theoretical predictions of settling velocity to bimodal (two particle sizes) suspensions. The model is adapted from the framework suggested by Mirza and Richardson. Because it is based on more sound assumptions, the correlation for predicting bimodal sedimentation is extended to describe multimodal suspensions.

A relatively new experimental technique (3), which measures and follows the settling velocities of particle fractions within a suspension, as well as at its interfaces, using a radioisotope tracer, has been developed. Data are provided for settling velocities versus concentration with bimodal distributions of microspheres of differing density as well as diameter.

## THEORY

Prior to discussing the theory of hindered settling, it is useful to consider the appearance of bimodal suspensions during batch settling. In Fig. 1, there are four distinct zones formed as settling progresses from an initially uniformly mixed suspension. Zone 1 consists of a sediment in which any further compaction is due to solids stresses. Zone 2 consists of large and small particles and is the primary area of interest since its interfacial settling velocity is the most difficult to predict. Zone 3 consists only of the smaller-particle fraction and approaches the behavior of a unimodal suspension. The settling velocity in Zone 3 is easily predicted using reliable correlations for uniform spheres, such as the equation of Richardson and Zaki (4). Zone 4 consists of clear supernatant liquid.

## Prior Research

Unflocculated hindered settling has been studied from various theoretical and empirical viewpoints. The theoretical work usually involves solutions of the Navier-Stokes equation with assumptions of creeping flow, limited particle interactions, monosized particles, and other important simplifications. Prominent investigators in the theoretical research have been Ward (5) and Batchelor (6). Their correlations usually do not predict settling velocities as well as the empirical relationships found by Steinour (7), Richardson and Zaki (4), Garside and Al-Dibouni (8), and Selim et al. (9).

Theoretical and empirical research on multimodal distributions is less developed. Mirza and Richardson (2) proposed a correlation for the settling velocities of bimodal distributions. Selim and coworkers (9) improved the accuracy of their correlation by proposing a density correction factor to the Stokes settling velocity term. They assumed that the effective fluid density around the larger settling particles was the volume weighted mean density of the fluid and the smaller solid particles.

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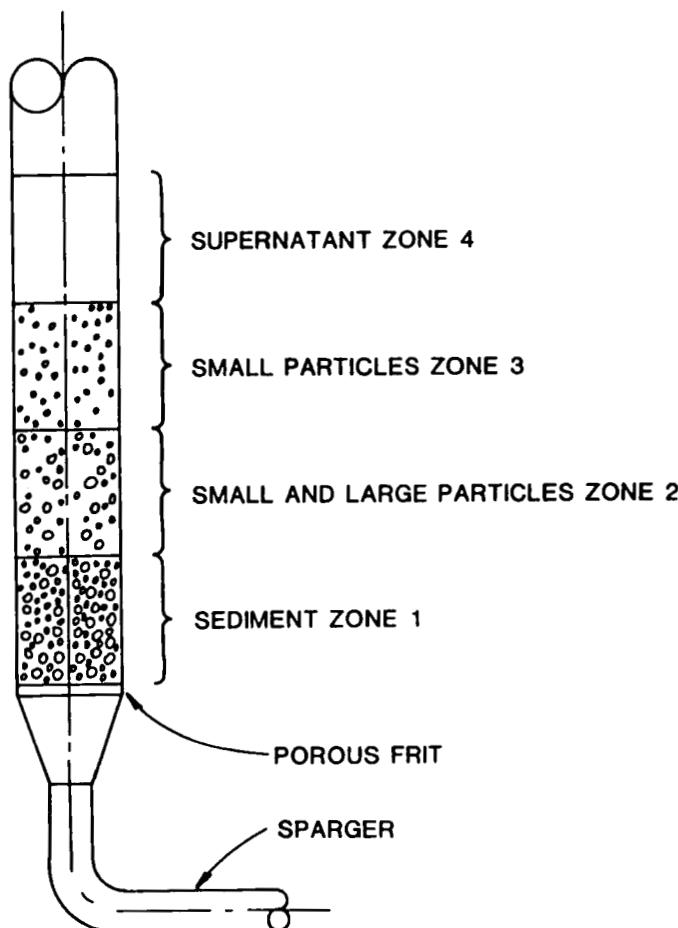


Fig. 1. Developing concentration zones, bimodal particle distribution.

### Proposed New Analysis

The theory of Mirza and Richardson (2) described a material balance between the volume of the settling particles and the upflowing liquid. The "slip velocity,"  $V_{sa}$ , for each size of particle "a" is defined as

$$V_{sa} = V_{ca} + V_f, \quad (1)$$

where  $V_{ca}$  is the settling velocity of particle "a" with respect to the settling container wall, and  $V_f$  is the upward velocity of fluid resulting from displacement of fluid by settling particles.

When this upward motion of displaced fluid is taken into account, the Richardson and Zaki equation can be written in terms of the slip velocity as

$$V_{sa} = V_{oa}(\epsilon^{n-1}), \quad (2)$$

where  $\epsilon$  is the void fraction of the suspension,  $V_{oa}$  is the Stokes settling velocity of particle "a," and  $n$  is an empirical constant.

Mirza and Richardson (2) applied this equation directly to bimodal sedimentation and proposed the following relation for the settling rate of particles "a" in the mixture:

$$V_{ca} = V_{oa}(\epsilon^{n-1})(1 - C_a) - V_{ob}(\epsilon^{n-1})(C_b), \quad (3)$$

where  $C_a$  is the concentration of the large-particle fraction, and  $C_b$  is the concentration of the small-particle fraction.

Selim et al. (9) proposed a modification to the Mirza-Richardson theory that redefines the density of the suspension in the region 2 of Fig. 1:

$$\rho_{eff} = \frac{\rho_b(C_b) + \rho_f(\epsilon)}{(1 - C_a)}, \quad (4)$$

and

$$V_{oa*} = \frac{D^2(\rho_a - \rho_{eff}) \cdot g}{18 \mu_f}; \quad (5)$$

where  $V_{oa*}$  is a modified Stokes settling velocity based on a fluid density as defined by Selim et al. in Eq. (4) that is substituted for  $V_{oa}$  in Eq. (3).

The Selim et al. modification reduced the predicted settling rates for the larger particles that were overestimated by the Mirza-Richardson equation.

### Proposed New Modification

Despite the good agreement between the predictions of Selim et al. and the available experimental data, their analysis fails to account for the effect of large particles on the (static) pressure gradients in the fluid and the resulting differences in pressure forces on the upper and lower surfaces of the particles.

Therefore, an alternative equation is proposed that introduces a modification to the viscosity of the suspension to account for the hindrance to the settling of particles in bimodal suspension (10):

$$V_{oa}^{**} = D^2(\rho_a - \rho_f)g/18 \mu_{eff}, \quad (6)$$

where  $\mu_{eff}$  is the effective viscosity of the suspension as sensed by the larger particles.

To include concentrated as well as dilute suspensions, it is necessary to choose an equation for the viscosity of the slurry. We chose a correlation of Ting and Luebbers (11), which relates the viscosity of a suspension to the concentration of particles and is relatively accurate over a wide range of solid concentrations:

$$\mu_{eff}/\mu_f = \frac{0.464 - 0.78 C_b}{0.464 + 0.21 C_b}. \quad (7)$$

As Ting and Luebbers proposed the correlation,  $C_b$  is the concentration of all particles. However, during settling in a bimodal system, the larger particles settle faster than the smaller particles and the smaller particles are forced to the side (or upward in some cases). Then the smaller particles contribute to an enhanced effective viscosity of the fluid as "seen" by the larger particles.

To use the Ting and Luebbers equation for settling in binary mixtures, it is necessary to know the correct value to use for the solids concentration. Before specifying how the smaller particles contribute to the effective slurry viscosity, it will be instructive to first consider two limiting cases: (1) very small particles that do not settle or hardly settle at all and (2) relatively large particles that are close to the size of the larger particles and thus settle only slightly more slowly than the larger particles. In the first case, one would expect the smaller particles to contribute fully to the effective fluid viscosity. The larger particles, however, settle at a uniform rate and do not interact directly with each other and contribute to the apparent slurry viscosity. However, in the second limiting case, the smaller particles should behave much like the larger particles and therefore contribute little to the effective suspension viscosity. Thus, the contribution of each size fraction is expected to be a function of the relative motion between the larger and the smaller particles. We assumed that the contribution of the smaller particles depends linearly on the relative Stokes settling velocities of the two sizes of particles:

$$C_b' = C_b(1 - V_{ob}/V_{oa}), \quad (8)$$

where  $C_b'$  is the effective solids concentration to use in the Ting and Luebbers equation. The combination of Eqs. (3) and (7) gives the settling rate for the larger particles in the bimodal mixture:

$$V_{ca} = V_{oa}(\mu_f/\mu_{eff})(\epsilon^{n-1})(1 - C_a) - V_{ob}(\epsilon^{n-1})(C_b) . \quad (9)$$

This equation is a simple modification of the Mirza and Richardson equation, with the introduction of the viscosity ratio calculated from the Ting and Luebbers correlation with the effective solids concentration obtained from Eq. (8).

## EXPERIMENTAL

The experimental apparatus is essentially the same as that described in a previous paper, which can be consulted for details (3). Briefly, the apparatus consists of a vertical glass column containing the suspension, which was sparged with air through a porous glass frit in the bottom for mixing the slurry prior to the sedimentation experiment. This sparger was found to produce homogeneous suspensions. The settling rates were determined by two methods: the first involved tagging the settling particles with a radioisotopic tracer, Cobalt-57, and following the gamma ray signal and, hence, the concentration by means of a scintillation crystal enclosed in a lead pig to shield the background radiation.

Since the effect of particle density was the central feature of the Selim correlation, bimodal mixtures that differed in density as well as diameter were studied by using glass beads along with the resin beads used in the previous experiments. The radioisotope tracer could not be used to measure the settling velocities of the glass particles; so the second method was used. A cathetometer was used to observe the settling of the glass particles.

The pertinent physical data on the particles used in these studies are given in Table 1. Additional details of the experimental technique and histograms of the particle size distributions are available (12).

Characterization of the physical properties of the bimodal distributions of microspheres proved difficult. The pycnometer measurements of the density of the ion-exchange resin were difficult to reproduce because of variable swelling and unpredictable amounts of moisture in the resin when it is weighed in air. Direct measurement of particle densities was circumvented by extrapolating the settling velocity of monodisperse suspensions of each particle size to infinite dilution. In the case of the glass particles, the particle Reynolds number was nearly 30 and should not be viewed as a "Stokes velocity."

## RESULTS

The proposed model will first be compared with previous data and with the model of Selim et al. Figure 2 presents data and available physical parameters given by Mirza and Richardson who, like most researchers, used glass beads under conventional conditions of creeping flow. The concentration of large particles is

constant and that of the small particles is variable. It is interesting to note the closeness of the predictions of the present theory to those of the Selim theory, despite the important differences in their developments.

Table 1. Microsphere properties

Property	Glass beads	Resin beads
Density, g/cm <sup>3</sup>	2.6	1.2 <sup>a</sup>
Mean diameter, $\mu\text{m}$	125	44
Particle Reynolds number	30	0.1
Settling velocity at infinite dilution, mm/s	19.3	0.22

<sup>a</sup>Wet.

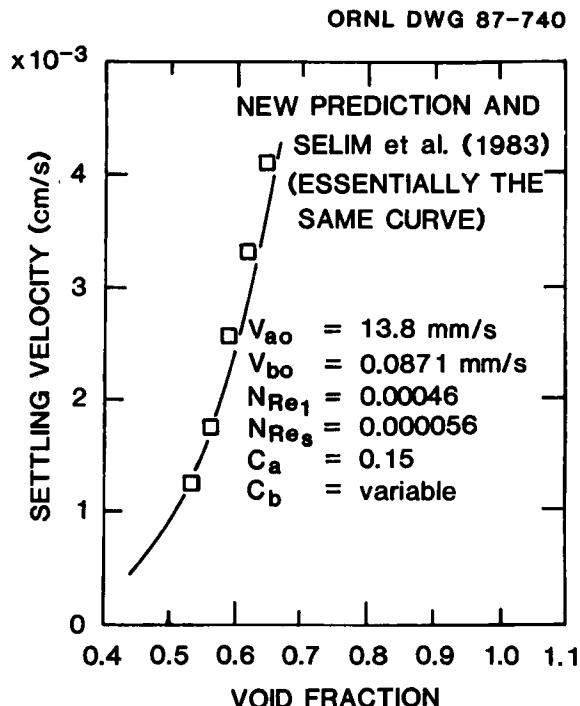


Fig. 2. Settling velocities versus void fraction of bimodal data of Mirza and Richardson.

Figure 3 shows bimodal settling data and available parameters from Smith (13) for a constant ratio of 1:1 for concentrations of the two fractions. The average percent deviation from the predicted value of settling velocity and the data values was 18 for both our new correlation and that of Selim et al.

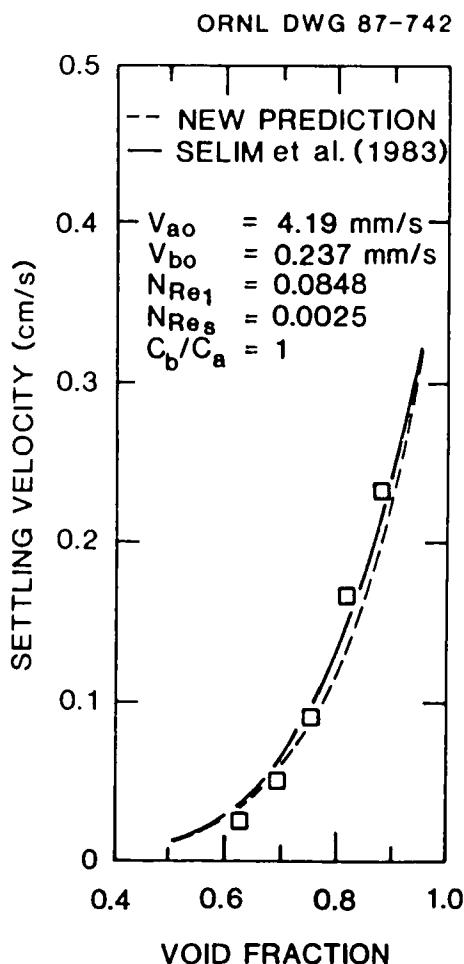


Fig. 3. Settling velocity versus void fraction of bimodal data of Smith.

Figure 4 presents bimodal settling data from Selim et al. (9) and the available parameters and properties for the particles and fluids. The data are presented with a constant concentration of the smaller species of 0.12 and a variable concentration of the larger species. Plotted also are the settling velocities in Zone 3. Both the correlation of Selim et al. and the new correlation from this paper give essentially the same predictions (shown by the solid curves), and agreement with the data is excellent. The new predictions (from both this study and that of Selim et al.) lie below those of Richardson and Zaki and fit the data much better. (The data and predictions for settling of the larger particles are shown in the upper portion of the figure with the square points, and of the smaller particles by the circular points near the bottom of the figure.)

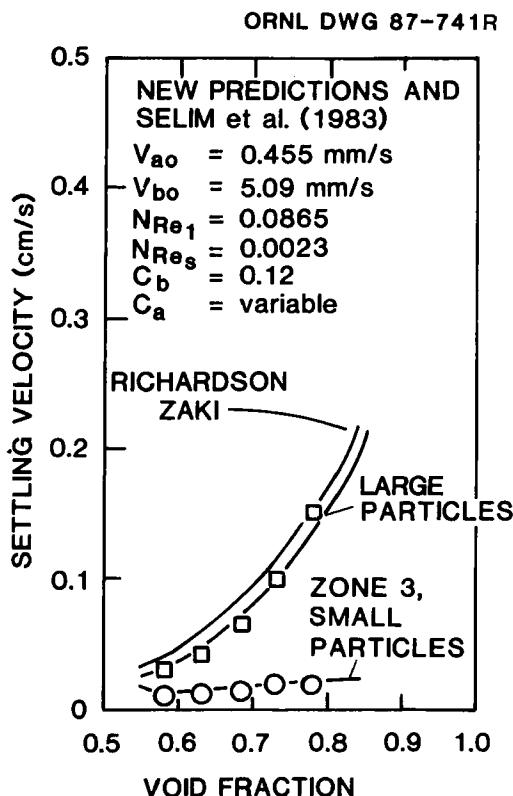


Fig. 4. Settling velocity versus void fraction of bimodal data of Selim.

In our experiments, particles with different densities and different diameters were used. The Reynolds number for the larger particles in free settling was  $\sim 30$  and in the transition region. Therefore, the drag coefficient,  $C_D$ , was used to describe their settling behavior at infinite dilution:

$$C_D = 18.5 N_{Re}^{-0.6}. \quad (10)$$

Solving for the settling velocity, one finds

$$V_{dilute} = \left[ \frac{0.0721 g D^{1.6} (\rho_a - \rho_f)}{\mu^{0.6} \rho_f^{0.4}} \right]^{1/1.4}. \quad (11)$$

Combining Eqs. (3) and (11), one obtains for the transition region

$$V_{ca} = V_{oa}(e^{n-2})(1-C_a) \left( \frac{\mu_f}{\mu_{eff}} \right)^{3/7} \left( \frac{\rho_f}{\rho_{eff}} \right)^{2/7} \left( \frac{\rho_a - \rho'}{\rho_a - \rho_f} \right)^{1/1.4} - V_{ob}(e^{n-2})C_b, \quad (12)$$

with

$$\rho' = \rho_a C_a + \rho_b C_b + \rho_f C_f. \quad (13)$$

In the second term on the right side of Eq. (13), which accounts for the acceleration of fluid around the settling larger particles, we chose to use the density of the suspension as if the larger particles were not present.

The predictions of the proposed model and of that of Selim et al. are compared with the new data for the transition region in Fig. 5. Note that the proposed model is in better agreement with the data than is the model of Selim et al. However, one should remember that the predictions of Selim et al. are based on a model for creeping flow. Similar analysis for the transition region behavior using their assumptions would probably improve the accuracy of their predictions.

### Discussion and Conclusions

The proposed model and the model derived by Selim et al. are compared in Table 2. Both predictive methods give good agreement with the data in the creeping flow region. In the transition region, the proposed model gives significantly better predictions than the model of Selim et al. because the Selim model was not developed for the transition region. For this reason, no comparison is shown for the two predictive methods in this region.

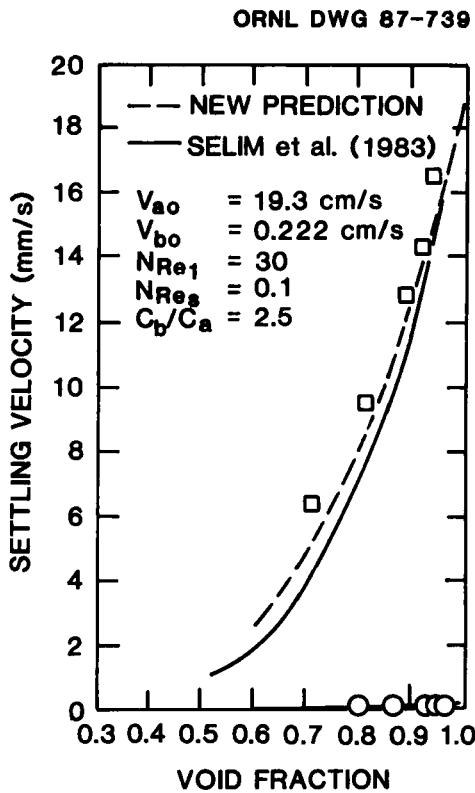


Fig. 5. Settling velocity versus void fraction of bimodal original data.

Table 2. Absolute value of average percent deviation of theoretically predicted values from experimental values in the creeping flow region

Present theory	Selim theory	Experimental data
8.5	7.9	Fig. 2 - Mirza
18	18	Fig. 3 - Smith
10	7.9	Fig. 4 - Selim

The data from the literature chosen for consideration in this study represent different situations and are believed to contain the most reliable and/or best described results available. These are also the data most commonly referred to by others working in this field. Unfortunately, the literature lacks the detailed characteristics of the histograms of particle size, which, according to Ward and Whitmore (5), can influence the relative viscosity. Obviously, the width of the particle size distribution can also affect the values of the settling velocities predicted by the theoretical correlations, since they all pick some average value of the Stokes settling velocity, and the particular choice depends on the researcher.

Both the Selim correlation and the new correlation presented here alter the predictions for settling velocities to values less than those of Mirza-Richardson (2). This result is not unexpected, and it will be recalled that Mirza and Richardson had succeeded in improving their correlations by the addition of an empirical factor, which also reduced the numerical values of the predicted velocities. Both the Selim and the new correlations actually make their best predictions in the areas of the greatest particle concentrations, unlike most earlier work, including that of Richardson and Zaki (4).

## **CONCLUSIONS**

Despite the similarity of the predictions of the new method to those of the earlier correlation presented by Selim et al., the new proposed correlation is believed to be important and, especially, more useful in certain ranges of concentrations and Stokes settling velocities. The Selim correlation, for example, makes no adjustment for the settling velocity if a suspension consists of particles of identical diameter but differing density. Furthermore, the Selim et al. correlation arbitrarily makes the same density adjustment for a suspension consisting of particle fractions differing in diameter by a large factor (such as 10) or a very small factor (such as 1.01). The ability of the new correlation to handle similar size particles as well as large differences is essential for predicting the behavior of continuous particle size distributions.

Although the predictions are similar for the data currently available, the predictions are not necessarily similar over all conditions. Table 3 shows that significant differences are predicted for suspensions with high concentrations of smaller particles. As previously noted, the Selim theory fails completely for situations with two particles of essentially identical diameters and differing densities. The comparison between the Selim model and the proposed model could be made by selecting binary mixtures with almost the same particle sizes.

Table 3. Example conditions for comparing theory predictions

Zone 2 settling velocity	Theory prediction (cm/s)	
	Proposed	Selim et al.
$V_{ca}$	1.97	2.48
$V_{cb}$	0.003	0.038
<hr/>		
Parameters:		
	<u>a (large) particles</u>	<u>b (small) particles</u>
Density, g/cm <sup>3</sup>	2.45	1.15
Volume, fraction conc.	0.0625	0.250

**NOTATION**

$C$	=	concentration in general, expressed as a volume fraction
$C_a$	=	concentration of particle fraction "a," large particles
$C_b$	=	concentration of particle fraction "b," small particles
$C_f$	=	concentration of fluid (equal to $\epsilon$ ), unitless
$D$	=	particle diameter in Stokes settling equation, microns or cm
$\epsilon$	=	void fraction (equal to $C_f$ ), unitless
$g$	=	gravitational acceleration constant, cm/s <sup>2</sup>
$k$	=	theoretical constant in Einstein equation related to particle shape, unitless
$\mu_{eff}$	=	corrected viscosity of suspension, centipoises
$\mu_f$	=	viscosity of liquid component of suspension, centipoises
$n$	=	empirical constant, unitless
$N_{Re_a}$	=	particle Reynolds number of large particles, unitless
$N_{Re_b}$	=	particle Reynolds number of small particles, unitless
$\rho_a$	=	density of particle "a," gm/cm <sup>3</sup>
$\rho_b$	=	density of particle "b," gm/cm <sup>3</sup>
$\rho_{eff}$	=	density of suspension (defined for each theory), gm/cm <sup>3</sup>
$V_{ca}$	=	hindered settling velocity of particle fraction a, cm/s
$V_{cb}$	=	hindered settling velocity of particle fraction b, cm/s
$V_f$	=	velocity of fluid, cm/s
$V_o$	=	Stokes settling velocity, cm/s
$V_s$	=	hindered settling slip velocity, cm/s

### Subscripts

a	relating to particle fraction a
b	relating to particle fraction b
c	relating to settling velocity with respect to wall of settling container
f	relating to fluid in which settling is occurring
o	relating to Stokes settling velocity
s	relating to slip settling velocity

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