Distillation," AIChE J., 29, No. 1, 49 (1983).

Reid, R. C., J. M. Prausnitz, and T. K. Sherwood, Properties of Gases and Liquids, 3rd Ed., McGraw-Hill (1977).
Rosenbrock, H. H., "Some General Implicit Processes for the Numerical

Solution of Differential Equations," Comp. J., 5, 329 (1963).

Seider, W. D., C. W. White, III, and G. J. Prokopakis, "Stiff Ordinary Differential Equations in Chemical Process Analysis," Proceedings of the AIChE-CIESC Meeting in Beijing, China (1982).

Sourisseau, K. D., and M. F. Doherty, "Dynamic Simulation of Stiff Distillation Systems," J.A.C.C., San Francisco (1980).

Tyreus, B. D., W. L. Luyben, and W. E. Schiesser, "Stiffness in Distillation Models and the Use of an Implicit Integration Method to Reduce Computation Times," I&EC Proc. Des. Dev., 14, 427 (1975)

Waggoner, R. C., and C. D. Holland, "Solution of Problems Involving Conventional and Complex Distillation Columns at Unsteady-State Operation," AIChE J., 11, 112 (1965).

Manuscript received March 3, 1982; revision received December 15, and accepted

# Sedimentation of Multisized Particles in **Concentrated Suspensions**

A model is developed for predicting the sedimentation velocity in suspensions of multisized nonflocculating solids, in which the retarding effect of the smaller particles on the setting velocities of the larger ones is taken into account. Tests of the model, and comparisons with other models, demonstrate that it provides improved prediction of data on suspensions comprising both discrete particle size mixtures and continuous size distributions, and that it is applicable to continuous countercurrent solid-liquid operations.

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# **SCOPE**

Sedimentation in concentrated suspensions of particles is a broad subject because of the wide range of particle sizes, the variety of particle shapes, and the complex nature of the hydrodynamic and physicochemical phenomena which govern particle-fluid and particle-particle behavior. The present work is limited to investigation of settling in concentrated noncolloidal suspensions of spherical particles of mixed sizes. Therefore, the important class of flocculant suspensions, usually comprised of particles in the submicron-size range, is excluded. Even with this circumscription the problem is substantial.

Sedimentation in noncolloidal suspensions comprised of spheres of uniform size and density has been investigated extensively. Theoretical results relating the settling velocity to solids volume fraction for such monodisperse suspensions of spheres have been treated in the dilute limit (Smoluchowski, 1912; Burgers, 1941, 1942; Uchida, 1949; McNown and Lin, 1952; Happel, 1958; Kawaguti, 1958; Hasimoto, 1959; Pyun and Fixman, 1964; and Batchelor, 1972). The intermediate and the high-concentration limits are more important in practice, but less amenable to rigorous theoretical treatment; they have been investigated rather extensively experimentally. Reliable empirical methods for determining the settling velocity-volume fraction (or voidage) relationship for concentrated suspensions of rigid spheres of uniform size and density are available (Garside and Al-Dibouni, 1977).

The subject of sedimentation involving suspensions containing mixed particle sizes is not as well developed, and reliable relationships have not been available. This paper is concerned with development of methods for predicting the settling velocity-voidage relationship in suspensions consisting of discrete as well as continuous mixtures of particle sizes. The method is based on knowledge of the sedimentation behavior of the individual size fractions within the mixture each settling alone in the suspending liquid. Interaction effects among particles of different sizes are accounted for by viewing the sedimentation of a given size fraction to take place in a matrix composed of the suspending fluid and the more slowly settling particles of smaller sizes.

## **CONCLUSIONS AND SIGNIFICANCE**

Operations involving relative motion between a fluid and suspended particles arise often and include sedimentation, fluidization and co- or countercurrent solid-liquid operations.

The hydrodynamics of slow vertically flowing liquid-particle mixtures, involving noncolloidal particles of uniform size and density, are understood in the dilute limit and can now be reliably treated empirically in the intermediate and concentrated limits. The presence of particles of mixed sizes enhances in-

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terparticle interaction because the faster settling larger particles in a sedimenting system must displace suspending fluid as the smaller particles which they overtake during their descent.

In addition the presence of a mixture of particle sizes enhances particle packing, and this affects suspension behavior very strongly outside the dilute limit. The dilute limit is one in which interparticle interactions are presumed to be negligible, and therefore particle size is irrelevant in this limit; particle volume fraction suffices. Natural and industrial processes generally involve many particles with wide distributions of sizes. Sedimentation in such systems results in particle classification by size, and models capable of describing settling in such concentrated mixed particle size system are needed in assessing industrial operations such as separation, and particle fractionation and natural processes involving sedimentation.

A model for sedimentation in binary suspensions, based on viewing individual particle settling to be governed solely by the local voidage surrounding it, without regard to the sizes of neighboring particles or to whether they are moving relative to it or not has been proposed (Lockett and Al-Habbooby, 1973).

The model overpredicts sedimentation velocities because it does not account for the additional retarding effects of the more slowly settling smaller particles as they are overtaken by the faster settling larger particles. An extension of this model, incorporating a correction factor equal to the voidage raised to the power 0.4, results in improved prediction for binary and ternary suspended mixtures but its limitations reside in its failure to distinguish among particles of different sizes.

In the present work a model is developed which describes sedimentation in mixed particle size suspensions in which the role of particle size is clearly distinguished. The model is tested against published and also newly collected experimental data on binary and ternary suspension sedimentation, and also on suspensions containing continuous particle size distributions. The new model predicts these low Reynolds number sedimentation processes very well, and is also capable of representing experimental data on binary countercurrent operations for which the representative particle Reynolds numbers are much higher.

## PREVIOUS WORK

There are many correlations in the literature for predicting the sedimentation or fluidization velocities in nonflocculating solid-liquid suspensions. For equi-sized particles, Garside and Al-Dibouni (1977) compared the accuracy of the available correlations in representing experimental results. The correlations suggested by Richardson and Zaki (1954), Barnea and Mizrahi (1973), and Garside and Al-Dibouni (1977) were found to be the most accurate. Among these, the Garside and Al-Dibouni correlation gives the most reliable estimates and the closest agreement with the experimental results. This correlation may be written as:

$$U_s/U_t = \epsilon^{n-1} \tag{1}$$

where n is given by

$$\frac{5.1 - n}{n - 2.7} = 0.1N_{Re}^{0.9} \tag{2}$$

In Eqs. 1 and 2,  $U_s$  is the fluid-particle relative velocity,  $U_t$  is the terminal velocity of a single-particle settling in the same fluid and container as the suspension,  $\epsilon$  is the bed voidage or porosity, and  $N_{Re} = \rho_f U_t d/\mu_f$  is the particle Reynolds number. The relative velocity  $U_s$  is also referred to in the literature as the slip velocity. For fluidization,  $U_s = U_o/\epsilon$  where  $U_o$  is the superficial velocity of the fluid. In sedimentation experiments, the average velocity of the particles relative to the container  $U_c$  is measured. The settling velocity  $U_c$  is related to the slip velocity by  $U_s = U_c/\epsilon$ .

Because of the possible significance of the container wall in laboratory-scale experiments, the terminal velocity including the container wall effect,  $U_t$ , rather than  $U_{t\infty}$ , is used in Eq. 1.  $U_{t\infty}$ , the terminal velocity of a sphere in an infinite fluid, is calculated from the standard drag coefficient curve (Perry and Chilton, 1973). Several expressions are available in the literature which relate  $U_t$  to  $U_{t\infty}$ . Garside and Al-Dibouni (1977) recommend the following expressions:

$$\frac{U_{t^{\infty}}}{U_t} = \left[\frac{1 - 0.475(d/D)}{1 - (d/D)}\right]^4 (N_{Re} < 0.2) \tag{3}$$

$$\frac{U_{t^{\infty}}}{U_{t}} = 1 + 2.35(d/D) (0.2 < N_{Re} < 10^{3})$$
 (4)

$$\frac{U_{t^{\infty}}}{U_t} = 1/[1 - (d/D)^{3/2}] (10^3 < N_{Re} < 3 \times 10^3)$$
 (5)

Equation 3 is due to Francis (1933), Eq. 4 is due to Garside and Al-Dibouni (1977), and Eq. 5 is due to Munroe (1888) according to Garside and Al-Dibouni.

Because of the simplicity and accuracy of the Garside and Al-

Dibouni correlation, Eqs. 1 and 2, we shall use it as the basis of our model for the sedimentation of multisized-particle systems.

The principal investigations on sedimentation of multisized particles are those due to Smith (1965, 1967), Lockett and Al-Habbobby (1973, 1974), and Mirza and Richardson (1979). Smith (1965, 1967) presented a theoretical model for the sedimentation of multisized particles. The model is an extension of the spherical fluid model originally proposed by Happel (1958) for the settling of equisized particles. Predictions from the model, however, were in poor agreement with the experimental data acquired by Smith (1965) for binary suspensions. Lockett and Al-Habbobby (1973, 1974) carried out sedimentation and countercurrent solid-liquid vertical flow experiments with binary particle mixtures. They found that the Richardson and Zaki correlation, originally proposed for monodisperse systems, could be applied to particles of each of the two sizes in a binary suspension. Mirza and Richardson (1979) extended the Lockett and Al-Habbobby model to the sedimentation of multisized particle systems.

Both models, however, ignore interparticle interactions during sedimentation; that is, the relative velocities of the particles of different sizes are assumed not to affect the settling velocity of any individual particle. As a result, both models were found to overpredict experimental sedimentation velocities. Mirza and Richardson (1979) applied a correction factor of (voidage)<sup>0.4</sup> to the predicted sedimentation velocities to obtain a better representation of the experimental data. The correction factor being empirical can only be used within the range of experimental conditions on which it was based

In the present paper, the Lockett and Al-Habbobby model for binary suspensions and its extension by Mirza and Richardson for multisized systems will be modified to account for interparticle interactions. Evidently, interparticle interactions become increasingly important both as the total solids concentration increases and as the relative velocity of the particles increases. The new model is tested against published and newly collected sedimentation data for suspensions with discrete and continuous particle size distributions. In addition, the present model is tested against published data on countercurrent solid-liquid vertical flow operations.

## PHYSICAL MODEL

In general, segregation of particles has been observed during the sedimentation of suspensions of two or more sizes of particles. Distinct sedimenting zones are formed due to complete segregation of particles when the size ratio of the two closest sizes of particles is greater than 1.6. Partial segregation without distinct zone for-

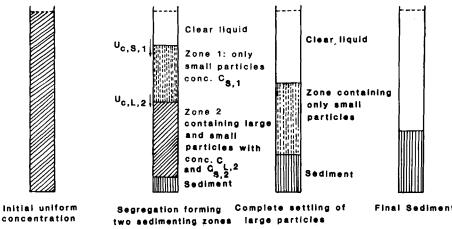


Figure 1. Segregation and zone-formation during sedimentation of a binary suspension.

mation has been observed even at a particle size ratio as low as 1.19. When a uniform suspension of two distinct sizes of particles starts settling, the segregation gives rise to two sedimenting zones; a lower zone in which particles of both sizes are settling and an upper zone in which only smaller size particles are settling (Fig. 1). Sedimentation of a suspension of three sizes of particles gives rise to three distinct zones of sedimentation, the lowest where all three sizes of particles are settling, the middle where medium and small sizes of particles are settling and the top where only small particles are settling. The same physical phenomenon occurs in a suspension of m sizes of particles; the occurrence of segregation and the formation of m sedimenting zones (Figure 2).

Lockett and Al-Habbooby (1973) developed a physical model for binary suspensions and Mirza and Richardson (1979) extended their model to multisized suspensions. Both groups of workers assumed that within any sedimenting zone the settling velocities of larger particles were unaffected by the sizes and velocities of surrounding particles. This assumption is physically unacceptable and results in higher predicted settling velocities for the larger particles.

It has been observed that, when a large particle settles in a suspension of smaller particles, it displaces not only the fluid but also the smaller-size particles. Thus, sedimentation velocities of large particles in a multisized-particle system are lower compared to their velocities in a suspension of their own kind at the same total concentration. It is clear, however, that interparticle interaction cannot be ignored in multisized-particle systems. These interactions are taken into account by considering the buoyancy effects of all particles having sizes less than those of particles in size group i on the terminal falling velocity of particles in size group i. Thus, the interaction effects among particles of different sizes are accounted for simply by computing the terminal settling velocity of a given particle as if it were settling in an effective fluid having the same viscosity as the pure fluid and a density equal to a mixture of the pure fluid and all of the more slowly settling particles locally present. We use this idea to derive first the model for binary suspensions, and then to extend it to the case of suspensions with multisized particles. Clearly, the range and accuracy of the model must rely on experimental data for verification.

## **BINARY SUSPENSIONS**

In general, a suspension of two distinct sizes of particles will give rise to four zones during the course of sedimentation (Figure 1). From the top downwards these will consist of: clear liquid, suspension of predominantly smaller particles, suspension of particles of both sizes with concentration equal to initial concentration, and lastly, sediment.

Consider sedimentation in the zone containing both sizes of particles. Let suffixes S and L designate the small and large particles, respectively, and suffixes 1 and 2 apply to upper and lower

sedimenting zones. To account for particle interactions in the lower sedimenting zone, the terminal falling velocity for the larger particles is calculated as if they were settling in a suspension consisting only of the smaller-size particles. In the Stokes' Law range the terminal falling velocity of a large particle  $U_{t^{\infty},L,2}$  is given as:

$$U_{t \infty, L, 2} = \frac{d_L^2(\rho_p - \rho_s)g}{18\mu_f} \tag{6}$$

where  $\rho_s$  is the density of a suspension consisting only of the small-size particles. The density  $\rho_s$  may be written as:

$$\rho_s = \frac{(1 - C_{L,2} - C_{S,2})\rho_f + C_{S,2}\rho_p}{(1 - C_{L,2})}$$
(7)

The terminal falling velocity for the small-size particles  $U_{t\infty,S,2}$  may be written as:

$$U_{t^{\infty},S,2} = \frac{d_S^2(\rho_p - \rho_f)g}{18\mu_f}$$
 (8)

To account for the container wall effect in Eqs. 6 and 8 we use the following equation by Francis (1933):

$$\frac{U_{t,i,2}}{U_{t^{\infty},i,2}} = \left[ \frac{1 - 0.475 \, d_i/D}{1 - d_i/D} \right]^{-4} \tag{9}$$

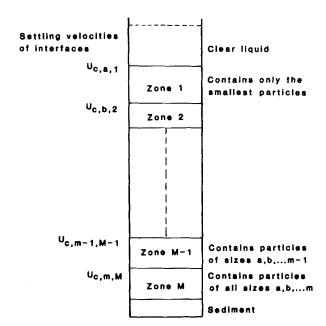


Figure 2. Segregation and zone-formation during sedimentation of a multisized particle suspension.

where i = L, S. Now Eq. 1 may be written for the small- and large-size particles in this zone to give:

$$U_{s,L,2} = U_{t,L,2} \epsilon_2^{nL,2^{-1}} \tag{10}$$

$$U_{s,S,2} = U_{t,S,2} \epsilon_2^{nS,2^{-1}} \tag{11}$$

The  $n_{L,2}$  and  $n_{S,2}$  may be computed from Eq. 2 for Garside and Al-Dibouni (1977). In sedimentation, the upward flow rate of liquid is equal to the downward flow rate of particles at any cross section. This gives:

$$U_{f,2}\epsilon_2 = U_{C,L,2}C_{L,2} + U_{c,S,2}C_{S,2}$$
 (12)

The fluid-particle relative velocity,  $U_s$ , is related to the fluid velocity,  $U_f$ , and the sedimentation velocity,  $U_c$ , by the equation

$$U_s = U_f + U_c \tag{13}$$

A plus sign is used in Eq. 13 since the direction of  $U_f$  is opposite to that of  $U_c$ . Equation 13, applied to the large and small particles in zone 2, gives:

$$U_{s,L,2} = U_{f,2} + U_{c,L,2} \tag{14}$$

$$U_{s,S,2} = U_{f,2} + U_{c,S,2} \tag{15}$$

Introduction of Eqs. 14 and 15 into Eq. 12 gives:

$$U_{f,2}\epsilon_2 = (U_{s,L,2} - U_{f,2})C_{L,2} + (U_{s,S,2} - U_{f,2})C_{S,2}$$
 (16)

Since  $\epsilon_2 + C_{L,2} + C_{S,2} = 1$ , Eq. 16 may be rearranged to give

$$U_{f,2} = U_{s,L,2}C_{L,2} + U_{s,S,2}C_{S,2}$$
 (17)

Now,

$$U_{c,L,2} = U_{s,L,2} - U_{f,2}$$

$$= U_{s,L,2} - (U_{s,L,2}C_{L,2} + U_{s,S,2}C_{S,2})$$

$$= U_{s,L,2}(1 - C_{L,2}) - U_{s,S,2}C_{S,2}$$
(18)

Substituting Eqs. 10 and 11 into Eq. 18 we get:

$$U_{c,L,2} = U_{t,L,2} \epsilon_2^{nL,2^{-1}} (1 - C_{L,2}) - U_{t,s,2} \epsilon_2^{nS,2^{-1}} C_{S,2}$$
 (19)

By the same procedure we get

$$U_{c,S,2} = U_{s,S,2} - U_{f,2}$$

$$= U_{s,S,2} - (U_{s,L,2}C_{L,2} + U_{s,S,2}C_{S,2})$$

$$= U_{s,S,2}(1 - C_{s,2}) - U_{s,L,2}C_{L,2}$$

$$= U_{t,S,2}\epsilon_2^{nS,2-1} (1 - C_{S,2}) - U_{t,L,2}\epsilon_2^{nL,2-1}C_{L,2}$$
(20)

Since all the terms on the righthand side of Eqs. 19 and 20 are known,  $U_{c,L,2}$  and  $U_{c,S,2}$  can be calculated. Here,  $U_{c,L,2}$  corresponds to the observed rate of fall of the interface between the two sedimenting zones.

The concentration of the particles in the upper zone 1 is not directly known but can be calculated using a mass balance. The volumetric rate at which the small particles pass from the lower zone to the upper zone is  $(U_{c,L,2}-U_{c,s,2})C_{S,2}A_t$ , where  $A_t$  is the cross sectional area of the container. The rate of increase in the volume of the upper zone is  $(U_{c,L,2}-U_{c,S,1})A_t$ . Thus, the concentration of particles in the upper zone,  $C_{S,1}$ , is given by

$$C_{S,1} = \frac{(U_{c,L,2} - U_{c,S,2})C_{S,2}}{(U_{c,L,2} - U_{c,S,1})}$$
(21)

Since this zone contains only uniformly-sized particles, Eq. 1 can be directly applied to give  $U_{c,S,1}$ 

$$U_{c,S,1} = U_{t,S,1}(1 - C_{S,1})^{n_{S,1}}$$
(22)

where  $U_{t,S,1} = U_{t,S,2}$  and also  $n_{S,1} = n_{S,2}$ . Equations 21 and 22 can be solved simultaneously to calculate  $U_{c,S,1}$  and  $C_{S,1}$ . Clearly,  $U_{c,S,1}$  corresponds to the observed rate of fall of the interface separating the suspension and the clear liquid.

In a typical case, one starts with a uniform suspension of known concentrations for the large- and small-size particles. These initial

concentrations are identical to  $C_{L,2}$  and  $C_{S,2}$ , respectively. With these known,  $U_{c,L,2}$  and  $U_{c,S,2}$  can be calculated from Eqs. 19 and 20. The latter quantities are then used in Eqs. 21 and 22 to calculate  $U_{c,S,1}$ . In this manner the rates of fall of the upper and lower interfaces, i.e.,  $U_{c,S,1}$  and  $U_{c,L,2}$  are determined from the model. These predicted values may be further checked against experimentally-measured values of the interface velocities.

## **MULTISIZED SUSPENSIONS**

Consider a suspension of m different-size particles  $(a,b,\ldots,m)$ ; size a being the smallest); this will give rise to M(=m) zones of settling suspensions,  $(1,2,\ldots,M)$  counting from top), with clear liquid above and a sediment layer at the bottom. All zones will exist at the beginning of the sedimentation (Figure 2) and each zone will disappear in turn during the sedimentation process as its upper boundary coincides with that of the sediment layer.

The model developed for binary suspensions may be extended to the sedimentation of multisized particle systems. Three suffixes will be used for the particle velocity U; the first suffix refers to the type of velocity (settling, slip, terminal), the second designates the size of particle (size a, size b, . . . , size m), and the third refers to the sedimenting zone under consideration (zone 1, zone 2, . . . , zone M). The symbol for concentration C has two suffixes; the first suffix designates the size of the particle and the second refers to the zone under consideration.

## Velocity of Particles in the Lowest Zone (M)

This zone contains all sizes of particles and their velocities can be written using Eq. 1 as:

$$U_{s,i,M} = U_{t,i,M} \epsilon_M^{n_i,M-1} \quad i = a,b,...,m$$
 (23)

As there is no material crossing the sediment zone, the upward flow rate of the liquid must equal the downward flow rate of the settling particles. Writing this in velocity terms, we get:

$$U_{f,M}\epsilon_{M} = U_{c,a,M}C_{a,M} + U_{c,b,M}C_{b,M} + \dots + U_{c,m,M}C_{m,M}$$
$$= \sum_{i=1}^{m} U_{c,i,M}C_{i,M}$$
(24)

The slip velocity of any size particle may be written as:

$$U_{s,i,M} = U_{f,M} + U_{c,i,M} \quad i = a,b,...,m$$
 (25)

Substituting  $U_{c,i,M}$  from Eq. 25 into Eq. 24, we get:

$$U_{f,M}\epsilon_M = \sum_{i=a}^m (U_{s,i,M} - U_{f,M})C_{i,M}$$
 (26)

Equation 26 may be rearranged to give:

$$U_{f,M} = \sum_{i=a}^{m} U_{s,i,M} C_{i,M}$$
$$= \sum_{i=a}^{m} U_{t,i,M} \epsilon_{M}^{n_{i}M-1} C_{i,m}$$
(27)

Substitution of  $U_{s,i,M}$  from Eq. 23 and  $U_{f,M}$  from Eq. 27 into Eq. 25 gives:

$$U_{c,i,M} = U_{t,i,M} \epsilon_M^{n_i,M-1} - \sum_{j=a}^m U_{t,j,M} \epsilon_M^{n_j,M-1} C_{j,m}$$

$$i = a \ b \qquad m \quad (28)$$

Since concentrations in this zone are the same as the initial concentrations, all the terms on the righthand side of Eq. 28 are known and the settling velocity of each size particle can be calculated directly. Evidently,  $U_{c,m,M}$  corresponds to the rate of fall of the interface separating the zones M and M-1. However, it should be kept in mind that the terminal falling velocity of a particle of size i is calculated as if it were settling in a suspension consisting only of particles of sizes smaller than size i. Accordingly, we may write

$$U_{t = s,i,M} = \frac{d_i^2 g(\rho_p - \rho_{s,i,M})}{18\mu_f} \quad i = b,c,...,m$$
 (29)

where  $\rho_{s,i,M}$  is the density of a suspension consisting of all particle sizes smaller than size i in zone M, and may be written as:

$$\rho_{s,i,M} = \frac{\left(1 - \sum_{j=a}^{m} C_{j,M}\right) \rho_{j} + \rho_{p} \sum_{j=a}^{i-1} C_{j,M}}{\left(1 - \sum_{j=i}^{m} C_{j,M}\right)} \quad i = b, c, \dots, m$$
(30)

For the smallest size particles (i = a), the terminal falling velocity may be written as

$$U_{t^{\infty},a,M} = \frac{d_{ag}^{2}(\rho_{p} - \rho_{f})}{18\mu_{f}}$$
 (31)

To account for wall effects in Eqs. 29 and 31 we use, as before, the equation

$$\frac{U_{i,i,M}}{U_{t^{\infty},i,M}} = \left[\frac{1 - 0.475d_i/D}{i - d_i/D}\right]^{-4} \quad i = a, b, \dots, m$$
 (32)

Equation 28 in combination with Eqs. 29, 30, 31 and 32 may be used to compute the settling velocities  $U_{c,i,m}$  of all different size particles in zone M.

#### Velocities of Particles in Zone M-1

The particles present in this zone are of sizes  $a, b, \ldots, m-1$ . Concentrations and settling velocities of particles of all sizes present are unknown and must be calculated.

The rate per unit area at which particles of size i cross the interface from zone M to zone M-1, say x, is given by

$$x = (U_{c,m,M} - U_{c,i,M})C_{i,M} \quad i = a,b,...,m-1$$
 (33)

The rate per unit area at which the volume of zone M-1 is increasing, say y, is clearly the difference in rate of fall of the interfaces forming this zone. This is, therefore, given by:

$$y = U_{c,m,M} - U_{c,m-1,M-1} \tag{34}$$

Dividing the quantity of Eq. 33 by that in Eq. 34, we get the concentration  $C_{i,M-1}$  as:

$$C_{i,M-1} = \frac{(U_{c,m,M} - U_{c,i,M})C_{i,M}}{(U_{c,m,M} - U_{c,m-1,M-1})} \quad i = a,b,\dots,m-1 \quad (35)$$

For this zone, equations analogous to Eqs. 28 through 32 may be listed as follows:

$$U_{c,i,M-1} = U_{t,i,M-1} \epsilon_{M-1}^{n_{i,M}-1-1} - \sum_{j=a}^{m-1} U_{t,j,M-1} \epsilon_{M-1}^{n_{j,M}-1-1} C_{j,M-1}$$

$$i = a, b, \dots, m - 1$$
 (36)

$$U_{t = i, t, M-1} = \frac{d_i^2 g(\rho_p - \rho_{s, i, M-1})}{18\mu_f} \quad i = b, c, \dots, m-1 \quad (37)$$

$$\rho_{s,i,M-1} = \frac{\left(1 - \sum_{j=a}^{m-1} C_{j,M-1}\right) \rho_f + \rho_p \sum_{j=a}^{i-1} C_{j,M-1}}{\left(1 - \sum_{j=1}^{m-1} C_{j,M-1}\right)}$$

$$i = b, c, \dots, M - 1$$
 (38)

$$U_{t \infty, a, M-1} = \frac{d_a^2 g(\rho_p - \rho_f)}{18\mu_f}$$
 (39)

$$U_{t^{\infty},a,M-1} = \frac{d_a^2 g(\rho_p - \rho_f)}{18\mu_f}$$

$$\frac{U_{t,j,M-1}}{U_{t^{\infty},i,M-1}} = \left[\frac{1 - 0.475d_i/D}{1 - d_i/D}\right]^{-4} \quad i = a,b,\dots,m-1$$
(40)

The terminal velocities  $U_{t,j,M-1}$   $(j=a,b,\ldots,m-1)$  are first computed from Eqs. 37 and 39. The densities  $\rho_{s,t,M-1}$  are required in Eq. 37. These are obtained from Eq. 38. An iterative scheme is, however, required since Eq. 38 contains the unknown concentrations  $C_{j,M-1}$  (j = a, b, ..., m-1). The iteration scheme proceeds

- (i) An initial set of values of concentrations  $C_{j,M-1}$  (j = a, $b, \ldots, m-1$ ) is assumed and used in Eq. 38 to compute  $\rho_{s,i,M-1}$  (i  $= b, c, \ldots, m-1$ ). A reasonable guess for this set is the corresponding concentrations in the lower zone.
- (ii) The terminal falling velocities  $U_{t\infty,i,M-1}$  (i = a, b, ..., m m1) are computed using Eqs. 37 and 39 together with Eq. 40 to account for the wall effect if necessary.
- (iii) Equations 35 and 36 are then solved simultaneously for the concentrations  $C_{i,M-1}$  and the velocities  $U_{c,i,M-1}$  (i = a, b, ..., m
- (iv) The values obtained in step (iii) for the concentrations  $C_{i,M-1}$  (i = a, b, ..., m-1) are used in step (i) to recalculate  $\rho_{s,i,M-1}$   $(i=b,c,\ldots,m-1)$  and the cycle is repeated. In the calculations, the iteration cycle is checked for completion by the requirements that

$$|C_{i,M-1}^{(n+1)} - C_{i,M-1}^{(n)}| < \delta_1$$

and

$$|U_{c,m-1,M-1}^{(n+1)} - U_{c,m-1,M-1}^{(n)}| < \delta_2$$

hold for all values of i(i = a, b, ..., m - 1) where  $\delta_1$  and  $\delta_2$  are prescribed error values.

The simultaneous solution of Eqs. 35 and 36 can be obtained in a direct manner by first writing Eq. 36 for the largest particle size in that zone. For the present zone (zone M-1), the largest particle size is m-1. The resulting equation takes the form:

$$U_{c,m-1,M-1} = f(C_{a,M-1}, C_{b,M-1}, \dots, C_{m-1,M-1})$$
 (41)

which indicates that  $U_{c,m-1,M-1}$  is a function only of all concentrations present in zone M-1. As Eq. 35 indicates, each of these concentrations is a function of  $U_{c,m-1,M-1}$  alone, i.e.,

$$C_{a,M-1} = f_a(U_{c,m-1,M-1})$$

$$C_{b,M-1} = f_b(U_{c,m-1,M-1})$$

$$\vdots \qquad \vdots \qquad \vdots \qquad \vdots$$

$$C_{m-1,M-1} = f_{m-1}(U_{c,m-1,M-1})$$
(42)

When expressions for these concentrations are substituted from Eq. 42 into Eq. 41, there results a nonlinear equation for  $U_{c,m-1,M-1}$ . The latter may be solved for  $U_{c,m-1,M-1}$  using the Newton-Raphson method or the Reguli-Falsi method (Lapidus, 1962). With  $U_{c,m-1,M-1}$  known, all unknown concentrations may be obtained directly from Eq. 35. Substitution of the resulting values for the concentrations into Eq. 36 gives the remaining unknown velocities and thereby the solution is complete.

Similar equations can be written for zones M-2, M-3,..., etc; and zone by zone computations can be carried out to calculate the concentrations and the settling velocities in all zones.

The simultaneous solution of Eqs. 35 and 36 may also be carried out iteratively. An initial value for  $U_{c,m-1,M-1}$  is taken as  $U_{c,m-1,M}$ , the settling velocity of the same size particle in the zone below the present zone. Substitution of  $U_{c,m-1,M-1}$  in Eq. 35 gives the concentrations of all particle sizes in this zone. These new values of concentrations are used to calculate  $U_{c,m-1,M-1}$  from Eq. 36. This value of  $U_{c,m-1,M-1}$  may be used to calculate new concentrations from Eq. 35. The iteration continues until the difference between two successive velocity values is less than a prescribed error bound. A computer program which carries out these computations for an arbitrary preassigned number of zones was written. A sample program is available (Kothari, 1981).

## **EXPERIMENTAL WORK**

To evaluate the present model, and to compare it with others, experimental data on suspensions with mixed particle sizes were taken, since available data in the literature on mixed sizes are insufficient. The particles used were glass spheres which were obtained in ten different size ranges,

TABLE 1. PROPERTIES OF GLASS SPHERES

Particle Size in Range Diameter Dens	
No. Sieve Numbers (micron, $\mu$ m) (micron, $\mu$ m) (kg/s	
$\frac{1}{1}$ $\frac{-35 + 40}{-35 + 40}$ $\frac{500 - 420}{500 - 420}$ $\frac{35 - 35}{460}$	
2 - 40 + 45 420 - 354 387 249	20
3 - 45 + 50 $354 - 297$ $326$ $245$	60
4 - 50 + 60 $297 - 250$ $274$ $244$	0
5 - 60 + 70 $250 - 210$ $230$ $248$	30
6 - 70 + 80 $210 - 177$ $194$ $247$	0
7 - 80 + 100   177 - 149   163   244	10
8 -100 + 120   149 - 125   137   246	60
9 $-120 + 140$ $125 - 105$ $115$ $239$	00
10  -170 + 200  88 - 74  81  234	10

TABLE 2. PROPERTIES OF SUSPENDING LIQUIDS

	Density at	Viscosity at
Suspending Medium	$297 \text{ K, kg/m}^3$	297 K, Pa-s
Ethylene Glycol	1,108	0.0184
Diethylene Glycol	1,115	0.0302
60% Aqueous Glycerol	1,165	0.0135

each representing the fraction between two consecutive sieves as shown in Table 1. An arithmetic average of the sieve openings was taken as the average diameter for each size fraction; the size ratios for pairs of openings being equal to  $\sqrt[4]{2}$  = 1.19. The densities of the particles were determined using a pycnometer, and an average value of 2,430 kg/m³ was used for all sizes since density variations among sizes were small, Table 1. To observe particle segregation, and to distinguish motions of various size fractions, the particles were colored using a process originally developed for dyeing glass fibers (Hyde, 1941). The dyeing had no discernible effect on particle size or density (Kothari, 1981).

Ethylene glycol, diethylene glycol, and 60% aqueous glycerol were used as suspending media. The densities of the liquids and their viscosities (measured over the range 293–299 K using a Brookfield viscometer) are given in Table 2.

Sedimentation experiments were carried out in a vertical flat-bottomed Plexiglas tube 3.2 cm in diameter and 76 cm long. During the experiment the tube was held in a vertical position using a metal stand inside a thermostated chamber, which was kept at a constant temperature of  $297 \pm 0.5$  K. The slurry of glass spheres was prepared by mixing known weights of liquid and the appropriate sizes of solids in the sedimentation tube, whose capacity was  $596~{\rm cm}^3$ . After the suspension had reached ambient temperature within the chamber, the suspension was agitated sufficiently to insure an initially uniform concentration throughout the tube, and the tube was replaced within the constant temperature chamber for the start of the sedimentation experiments.

Varying proportions of each particle size were used to prepare 39 different suspensions; 30 suspensions comprising mixes of two sizes of particles, three suspensions of three particle size mixtures, and six suspensions containing continuous particle size distributions. Sedimentation data were taken at five different concentrations for each suspension. The total concentration of solids ranged from 12% to 45% by volume. Altogether 195 data points were collected. Each data point was replicated once and reproducibility was within 3%. The averages of the two measurements were used in the final calculations. Additional details on the experimental work and the listing of all the data are given by Kothari (1981).

Aside from the data collected in our work, the only other available data which could be used here were the data on sedimentation in binary systems due to Smith (1965) and Mirza and Richardson (1979), and the sedimentation and countercurrent flow data in binary suspensions due to Lockett and Al-Habbooby (1973). Smith's binary data totalled 85 points and Mirza and Richardson's data consisted of 45 data points, all of which were used here. Lockett and Al-Habbooby's sedimentation data concerned the initial sedimentation rates for binary suspensions and could not be used with the present model which predicts average settling rates. Smith (1965) also presented five data points on ternary systems, but these could not be used because of the absence of information regarding concentration. The binary and ternary sedimentation data of Davies (1968) were also excluded because properties of the solids and suspending liquid were not given in his paper. Like Lockett and Al-Habbooby (1973), we were unable to reproduce Davies' (1968) experiments because settling rates were so rapid with the types of suspensions he used that we could not obtain an initially uniform concentration.

#### **RESULTS AND DISCUSSION**

Sedimentation data are usually expressed as interface velocity versus voidage in the suspension. We present in Figures 3 to 6 typical and representative velocity-voidage plots which compare our model with our experimental data on binary suspensions. To conserve space, only a small selection from the total number of such velocity-voidage plots we developed are presented here. The complete set of plots and the data are given in Kothari (1981). Figure 7 presents a comparison between the present model and the binary sedimentation data taken by Smith (1965). Figure 8 provides a comparison between our and Mirza and Richardson's model using Mirza and Richardson's (1979) binary sedimentation data. Lockett and Al-Habbooby's (1973), Mirza and Richardson's (1979) and the present models are compared in Figs. 9 and 10.

Aside from the graphical comparisons in Figures 3 to 10 we present in Table 3 the average absolute percent deviations between predicted and measured values for interface velocity for these models. Although not shown in this table, deviations equal to or exceeding 20% occur for only twelve of the data points for the present model, while the number of data points with deviations outside the 20% band is 77 for Mirza and Richardson's model, and 218 for Lockett and Al-Habbooby's model. It is clear from the graphical comparisons and from the numerical values in Table 3. which is based on 238 data for each of the interfaces, that the present model does a superior job of prediction for binary sedimentation than previously proposed models for both interfaces. Figures 9 and 10 demonstrate that the Lockett and Al-Habbooby model considerably overpredicts the settling velocities for both interfaces; no doubt for the reasons stated at the outset. Although these same figures show that predictions using the Mirza and Richardson model are not unsatisfactory for the upper interface, significant deviations from experimental data are obtained for the lower interface.

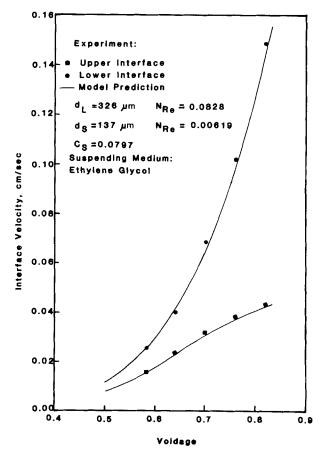


Figure 3. Comparison of experimental results with model for  $d_L=326~\mu m$ ,  $d_S=137~\mu m$ , and  $C_S=0.0797$ .

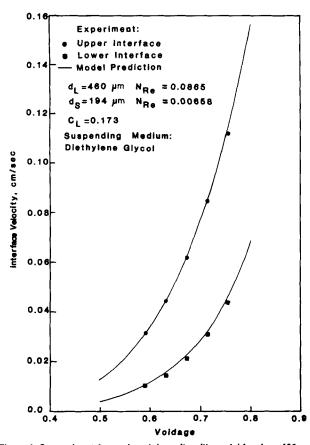


Figure 4. Comparison of experimental results with model for  $d_L=460~\mu{\rm m},$   $d_S=194~\mu{\rm m},$  and  $C_L=0.173.$ 

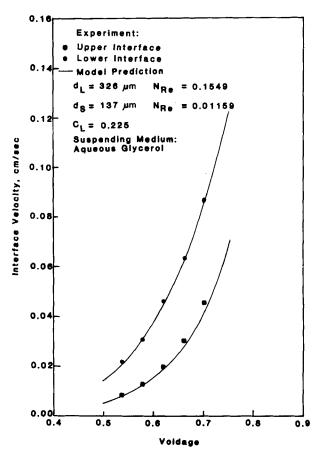


Figure 6. Comparison of experimental results with model for  $d_L=326~\mu{\rm m},$   $d_S=137~\mu{\rm m},$  and  $C_L=0.225.$ 

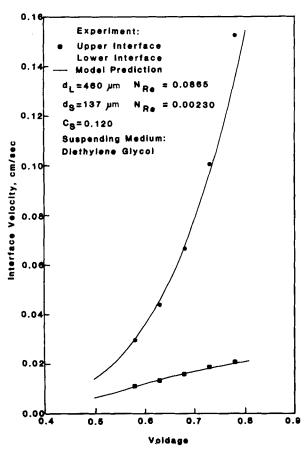


Figure 5. Comparison of experimental results with model for  $d_L=460~\mu{\rm m},$   $d_S=137~\mu{\rm m},$  and  $C_S=0.120.$ 

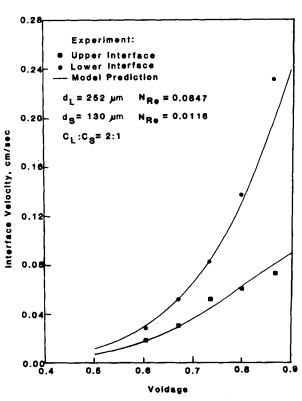


Figure 7. Comparison of the present model with experimental data of Smith.

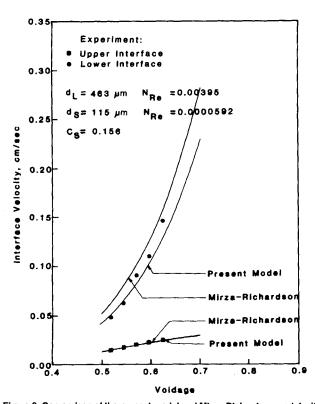


Figure 8. Comparison of the present model and Mizra-Richardson model with experimental data of Mizra and Richardson.

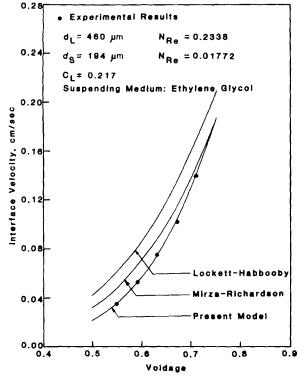
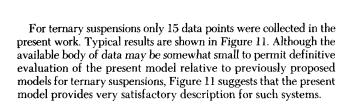


Figure 9. Comparison of models with new experimental data for the lower interface.



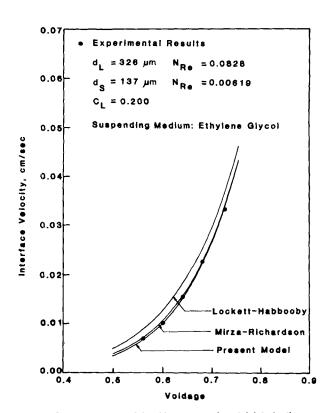


Figure 10. Comparison of models with new experimental data for the upper interface.

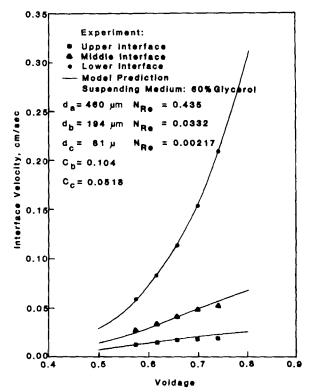


Figure 11. Comparison of the present model with new experimental data for ternary suspensions.

The sedimentation studies described in the foregoing provide tests of the efficacy of the present model for settling at low Reynolds numbers, which is of course a very important range in practice. Data for higher values of Reynolds number may be obtained from co- or countercurrent solid-liquid vertical flows. Lockett and Al-Habbooby's (1973) counter-current flow experiments covered the

TABLE 3. COMPARISON OF PRESENT AND PREVIOUSLY PUBLISHED MODELS WITH EXPERIMENTAL DATA ON BINARY SEDIMENTATION AND COUNTERCURRENT SOLID-LIQUID VERTICAL FLOWS

	Absolute	Average I	Percent D	eviation
	Binary		Countercurrent	
	_Sedime	ntation	$_{-}$ $_{-}$ Flo	oww
	Lower	Upper	Large	Small
Model	Interface	Interface	Particles	Particles
Lockett and Al-Habbooby (1973)	34.2	12.1	40.1	22.2
Mirza and Richardson (1979)	15.5	6.6	21.8	21.3
Present	5.8	6.1	6.8	16.8

Reynolds number range from 79 to 546. Unlike sedimentation such continuous binary operations do not result in distinct zones, but the operation may be viewed to be similar to that occurring in the lower zone in a sedimenting binary suspension. The slip velocities for each size fraction may be calculated using Eqs. 10 and 11. The terminal settling velocities for the individual particles settling alone, for this higher range of Reynolds numbers, are calculated using the drag coefficient-Reynolds number correlation given by Turian et al. (1971). This correlation is given by

$$\begin{split} \log_{10} N_{Re^{\infty}} &= -1.38 + 1.94 \log_{10} \Lambda \\ &- 8.60 \times 10^{-2} (\log_{10} \Lambda)^2 - 2.52 \times 10^{-2} (\log_{10} \Lambda)^3 \\ &+ 9.19 \times 10^{-4} (\log_{10} \Lambda)^4 + 5.35 \times 10^{-4} (\log_{10} \Lambda)^5 \end{split} \tag{43}$$

where

$$\Lambda = N_{Re} C_D^{1/2} = \left[ \frac{4}{3} \frac{g d^3 \rho_f (\rho_p - \rho_s)}{\mu_f^2} \right]^{1/2}$$
 (44)

Equation 43 is valid up to  $N_{Re^\infty} \leq 1.5 \times 10^5$ . The wall effect is included using Eq. 4 due to Garside and Al-Dibouni (1977). The various models and the experimental data of Lockett and Al-Habbooby on binary counter-current suspended flow were compared, and the results are given in Table 3. These comparisons demonstrate that the present model does a much more satisfactory job of representing such data than previous models, and they further provide indications of the validity of the model over broader ranges of Reynolds number than those pertaining to sedimentation experiments alone.

The applicability of the present model for describing sedimentation in suspensions with continuous particle size distributions was also tested. Unlike suspensions containing discrete particle size fractions, no distinct zones are observed in such suspensions but partial segregation of particles by size does occur. To test suspensions containing continuous size distributions using the present methods, we consider such a mixture to consist of a mixture of closely sized fractions, each covering a narrow range of the size spectrum. Then the rate of fall of the interface separating suspension from clear liquid in the partially segregated suspensions is calculated using the model developed for segregated suspensions. In making progressive settling computations with the distribution broken into various size fractions, Smith (1966) found that it was sufficient to have size fractions in which the largest to smallest particle size spread corresponded to a size ratio of 1.2, which coincides with the ratio for two successive screen openings in the U.S. Sieve Series ( $\sqrt[4]{2} = 1.19$ ). Accordingly two kinds of mixtures were prepared: the first consisted of two successive size fractions, and the second of three successive size fractions of particles. Figures 12 and 13 depict typical plots comparing predicted and observed interface velocities for a two-fraction and a three-fraction suspension, respectively. These comparisons demonstrate that the present model is also applicable to suspensions with continuous size distributions.

Although the present model predicts the sedimentation rate for a wide range of particle size ratios and concentrations, it deviates substantially in two limiting cases. The first corresponds to suspensions with nearly equal-sized particles. In this case, depending on concentrations, the model may incorrectly predict the smaller particles to settle faster than the larger ones in the zone containing

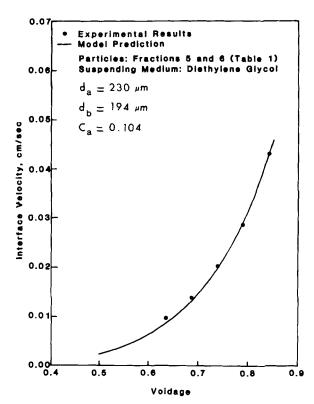


Figure 12. Comparison of the present model with experimental data on suspensions with continuous size distribution.

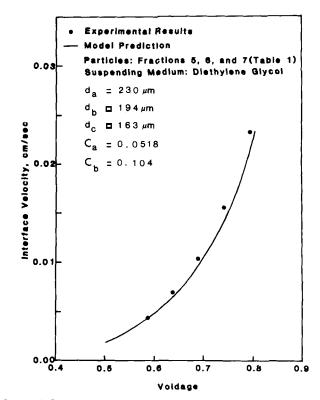


Figure 13. Comparison of the present model with experimental data on suspensions with continuous size distribution.

both species. This limitation is attributable to the fact that the model presumes the occurrence of segregation and distinct zone formation which do not arise in suspensions of nearly equal-sized particles. In fact Coe and Clevenger (1916) have long ago indicated that such suspensions settle with uniform concentration and no segregation (so-called *en masse* settling). The second limiting case corresponds to the opposite limit when there are very large particles settling

amongst much smaller ones. In this case the larger particles settle much faster than the smaller ones, which virtually remain stagnant during the entire duration of fall of the large particles. Thus the larger particles would behave as if they were settling in a homogeneous suspension with an effective viscosity larger than that of the pure fluid alone.

#### **ACKNOWLEDGMENT**

This work was supported by the National Science Foundation through Grant CPE-7827000, and by the International Fine Particle Research Institute, Inc.

#### **NOTATION**

$A_t$	= cross-sectional area of vessel or tube
$\boldsymbol{C}$	= concentration in volume fraction
D	= diameter of vessel or tube
d	= diameter of spherical particle
g	= acceleration due to gravity
n	= exponent of $\epsilon$ in Eq. 1
$U_c$	= settling velocity of particle in suspension
$U_f$	= velocity of displaced fluid
Ú.	= superficial velocity of fluid

 $U_{\mathfrak{s}}$ = slip velocity (relative velocity between particles and fluid)

 $U_t$ = terminal velocity of a single particle in finite fluid = terminal velocity of a single particle in an infinite fluid medium

ion)

## **Greek Letters**

$\epsilon$ = voidage (fluid fraction in sus	spensi
$\mu_f$ = viscosity of fluid	
$\rho_f$ = density of fluid	
$\rho_p$ = density of particles	
$\rho_s$ = density of suspension	

## **Dimensionless Groups**

$N_{Re}$	= particle Reynolds number $dU_t \rho_f / \mu_f$
$N_{Re^{\infty}}$	= particle Reynolds number $dU_{t\infty}\rho_f/\mu_f$

## Subscripts

L	= large particle
S	= small particle
a - m	= sizes of particles (smallest to largest)
1-M	= settling zones (top to bottom)

# LITERATURE CITED

Barnea, E., and J. Mizrahi, "A Generalized Approach to the Fluid Dynamics of Particulate Systems. Part 1: General Correlation for Fluidization and Sedimentation in Solid Multiparticle Systems," Chem. Eng. J., 5, 171 (1973).

- Batchelor, G. K., "Sedimentation in a Dilute Dispersion of Spheres," J. Fluid. Mech., 52, 245 (1972).
- Burgers, J. M., "On the Influence of the Concentration of a Suspension Upon the Sedimentation Velocity," Proc. Kon Nederl. Akad., Wetenschappen (Amsterdam), 44, 1045, 1117 (1941); 45, 9, 126 (1942).
- Coe, H. S., and G. H. Clevenger, "Method for Determining the Capacities of Slime-Settling Tanks," *Trans. AIME*, **55**, 356 (1916).
- Davies, R., "The Experimental Study of the Differential Settling of Particles in Suspension at High Concentrations," Powder Tech., 2, 43 (1968/
- Francis, A. W., "Wall Effect in Falling Ball Method for Viscosity," Physics, 4, 403 (1933).
- Garside, J., and M. R. Al-Dibouni, "Velocity-Voidage Relationship for Fluidization and Sedimentation in Solid-Liquid Systems," Ind. Eng. Chem., Process Des. Dev., 16, 206 (1977).
- Happel, J., "Viscous Flow in Multiparticle Systems: Slow Motion of Fluids Relative to Bed of Spherical Particles," AIChE J., 4, 197 (1958).
- Hasimoto, H., "On the Periodic Fundamental Solutions of the Stokes Equations and Their Application to Viscous Flow Past a Cubic Array of Spheres," J. Fluid Mech., 5, 317 (1959). Hyde, J. F., "Method of Coloring Glass Fibers," U.S. Patent 2,245,783
- (1941).
- Kawaguti, M., "A Hydrodynamical Model for the Sedimentation," J. Phys. Soc. Japan, 13, 209 (1958). Kothari, A. C., "Sedimentation of Multisized Particles," M.S. Thesis, Texas
- Tech University (1981).
- Lapidus, L., "Digital Computation for Chemical Engineers," McGraw-Hill, New York (1962).
- Lockett, J. M., and H. M. Al-Habbooby, "Differential Settling by Size of Two Particle Species in a Liquid," Trans. Inst. of Chem. Eng., 51, 281
- Lockett, J. M., and H. M. Al-Habbooby, "Relative Particle Velocities in Two-Species Settling," Powder Tech., 10, 67 (1974).
- McNown, J. S., and P. N. Lin, "Sediment Concentration and Fall Velocity, Proc. 2nd Midwestern Conf. Fluid Mechanics, reprint in Eng. No. 109, Iowa State University, 401 (1952).
- Mirza, S., and J. F. Richardson, "Sedimentaton of Suspension of Particles of Two or More Sizes," *Chem. Eng. Sci.*, 34, 447 (1979). Munroe, H. S., "The English versus the Continental System of Jigging—Is
- Close Sizing Advantageous?" Trans. Am. Inst. Min. Eng., 17, 637 (1888).
- Perry, R. H., and C. H. Chilton, "Chemical Engineer's Handbook," 5th Ed., McGraw-Hill, New York (1968).
- Pyun, C. W., and M. Fixman, "Frictional Coefficient of Polymer Molecules in Solution," J. Chem. Phys., 41, 937 (1964). Richardson, J. F., and W. N. Zaki, "Sedimentation and Fluidization: Part
- I," Trans. Inst. Chem. Eng., 32, 35 (1954).
- Smith, T. N., "The Differential Sedimentation of Particles of Two Different Species," *Trans. Inst. Chem. Eng.*, 43, 769 (1965).
- Smith, T. N., "The Sedimentation of Particles Having a Dispersion of Sizes," Trans. Inst. Chem. Eng., 44, T153 (1966).
  Smith, T. N., "The Differential Sedimentation of Particles of Various
- Species," Trans. Inst. Chem. Eng., 45, T311 (1967).
- Smoluchowski, M. S., "On the Practical Applicability of Stokes' Law of Resistance, and the Modifications of It Required in Certain Cases," Proc. 5th Intern. Congr. Math., 2, 192 (1912).
- Turian, R. M., T. F. Yuan, and G. Mauri, "Pressure Drop Correlation for Pipeline Flow of Solid-Liquid Suspensions," AIChE J., 17, 809 (1971).
- Uchida, S., "Slow Viscous Flow Through a Mass of Particles," Report Inst. Sci. Technol. Univ. Tokyo, 3, 97 (1949); Happel, J., Abstract, Ind. Eng. Chem., 46, 1194 (1954).

Manuscript received May 10, 1982; revision received December 21, and accepted January 6, 1982.