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# Suspensions and Sediments. Part I. Particle-Particle Interactions in Dilute Solid-Liquid Suspensions

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# REVIEW

# Suspensions and Sediments. Part I. Particle–Particle Interactions in Dilute Solid–Liquid Suspensions

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## Abstract

This article gives a general review of sedimentation behavior relating to dilute suspensions of solids in liquids. There is a strong assumption that partial-fluid association is the factor responsible for variations of sedimentation behavior from one suspension system to another. Studies are therefore cited to acquire an understanding of the hydrodynamic interactions between the solids and liquids within suspensions. Behavior of colloidal suspensions is also included, and the effects of such factors as liquid dielectric constant, charge density, and electrical potential on the interaction of particles are discussed.

## INTRODUCTION

A number of industrial processes deal essentially with the movements of solids in fluids. The phenomenon is of extreme theoretical and practical importance in processes such as differential settling, sedimentation, thickening, and filtration. The behavior of particles in a very dilute suspension (Reynolds number Re < 1) was studied by Stokes in 1851 when he determined the limiting (i.e., maximum) velocity $V_s$ of an isolated spherically shaped particle in a viscous liquid by

$$V_s = \frac{2gr^2(\rho_s - \rho_l)}{9\eta} \tag{1}$$

where $g$ = acceleration due to gravity (cm/s$^2$)
$r$ = radius of particle (cm)
$\rho_s$ = density of the solid particle (g/cm$^3$)
$\rho_l$ = density of the liquid (g/cm$^3$)
$\eta$ = absolute viscosity of liquid (g/cm · s)

A brief description of Reynolds number is given in the Appendix. Under the Stokes' conditions it is assumed that resistance to the motion of a spherical particle $R$ is directly proportional to the size and velocity of the falling particle and is given by

$$R = 6\pi\eta r v \tag{2}$$

in which $r$ is the radius of the sphere and $v$ is the velocity of fall of the particle. This is true of large or small spheres provided $v$ is small enough. The fact should be emphasized that Stokes' law, although holding for low velocities of all moving spheres, holds for the terminal velocity of smaller spheres, but fails for the terminal velocity of large spheres (*1*).

Under Newton-Rittinger conditions, the resistance to the motion of a sphere is

$$R = \frac{C_R \pi \rho_l r^2 v^2}{2} \tag{3}$$

in which $C_R$ is known as the coefficient of resistance. This equation deals with motion under turbulent resistance conditions.

In suspensions of higher solid concentrations, there is a real possibility that the fall of any single particle will be hindered by the other particles in its path. It is found that solid-liquid and solid-solid interaction causes departure from Stokes' law at particle concentrations higher than a certain value, and, therefore, the settling behavior of a single particle in a concentrated suspension is dependent on the neighboring solid particles and on the relative increase in the density of the suspension.

The degree of these interactions is controlled by the overall interfacial area between the liquid and the solid, which increases by the introduction of more and more solid particles into the suspension. Small particles will give relatively larger interfacial areas for unit mass and will thus result in a higher degree of interaction.

For larger particles the interaction will be reduced and, thus, their settling will be relatively more influenced by other physical factors, such as size, shape, specific gravity, and concentration of particles in the suspension.

## MOVEMENT OF PARTICLES IN DILUTE SUSPENSIONS

If a number of particles are settling simultaneously in a liquid, the conditions are different from those in which a single particle is settling. For a particle falling in a liquid, there must be an appreciable displacement of fluid in the direction opposite to that of the motion of the particle. In the case of particles falling in a "still" liquid, the liquid ceases to be "still" as a first approximation if the particles are present in large numbers.

Hall (*2*) and Boardman and Kaye (*3*) studied the settling rate of a single particle in the presence of another particle or even a container wall and showed it to be effected in such a way that when the particle is settling near the wall its velocity is reduced, but when it is brought near other particles its velocity is increased. This degree of acceleration was also reported by Happel (*4*) who attributed it to be a function of interparticle distance and the geometric locations of the particles.

The interparticle distance varies constantly, so the actual distance cannot be assumed. From the foregoing observations, a suitable wall correction can be calculated from the Munroe equation (*5*) which relates the settling velocity ($V_s$) of a particle to the ratio of the radii of the particle and the cylinder:

$$V_s = 1 - \left(\frac{r}{r'}\right)^{3/2} \frac{4}{3C_R}\left(\frac{\rho_s - \rho_l}{\rho_l}\right)\mathrm{rg} \tag{4}$$

where $r$ and $r'$ are the radii of the particle (sperical) and the cylinder, respectively.

The determination of an appropriate particle radius is still doubtful under these conditions due to the fact that in Munroe's experiments, the "cylinder wall" is supposed to be a real stationary boundary, but it is an imaginary and moving boundary in the case of the application of this equation to the movement of particles "en masse." The problem is the same with the application of Francis' equation for wall correction (*6*):

$$V_s = \frac{2}{9}\left(1 - \frac{r}{r'}\right)2.25\,\frac{(\rho_s - \rho_l)r^2g}{\eta} \tag{5}$$

Francis' correction has been applied to the calculation of settling velocities of various size glass ballotinis settling in aqueous glycerol. Table 1 shows a comparison of the calculated, observed, and corrected settling velocities and the corresponding particle radii.

The wall correction factor will be minimal for fine particles settling in a large diameter container because the ratio $r/r'$ will become much smaller.

In an observation by Kaye and Boardman (*3*), the particle acceleration effect was found to be due to particle–particle interaction in dilute suspensions. This has been explained by the formation of clusters enabling groups of particles to fall more quickly than individual particles. The mean observed settling rate, therefore, increases above the Stokes' limiting value $V_s$ with an increase of solids concentration up to some maximum value. On a further increase of concentration, however, the velocity falls rapidly because the phenomenon of return flow counteracts the cluster formation and hindered settling is dominant after that. These observations, therefore, lead to a general form of the sedimentation curve of the type shown in Fig. 1.

The variation in settling time may be used as a measure of the degree of clustering. The interaction between particles leads to the formation of doublets, triplets, quadruplets, and even larger clusters.

Gotoh et al. (*7*) and Kamel et al. (*8*) emphasized that the formation and

TABLE 1

Calculated, Observed, and Corrected (by Francis' correction factor) Settling Rates ($V_s$, cm/s) for Glass Ballotini in 3:1 (v/v) Glycerol:Water, and Corresponding Mean Particle Radii ($r$, μm)

| Grade of glass ballotini | A | B | C | D | E | F | G |
|---|---|---|---|---|---|---|---|
| $V_s$ calculated: From nominal radius | 3.97 | 1.90 | 1.15 | 0.99 | 0.71 | 0.53 | 0.18 |
| From weighted mean radius | 3.80 | 2.07 | 1.10 | 1.02 | 0.68 | 0.57 | 0.16 |
| $V_s$ observed (corrected for wall effect) | 3.78 | 2.18 | 1.05 | 1.02 | 0.74 | 0.59 | 0.19 |
| Mean radius $r$ (μm): Nominal | 650 | 450 | 350 | 325 | 275 | 238 | 138 |
| Weighted from microscopy | 637 | 468 | 343 | 330 | 270 | 246 | 130 |
| From $V_s$ observed (corrected) | 635 | 482 | 334 | 330 | 281 | 251 | 142 |

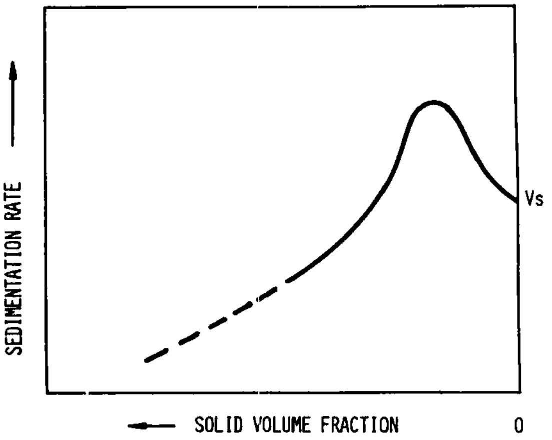

FIG. 1. Dependence of sedimentation rate on concentration of suspensions of closely sized spheres ($V_s$ = Stokes' velocity).

stability of a cluster depends on the size ratio of the component spheres, and Hall (*2*) reported that in a pair of spheres of different sizes, a stable doublet will not be formed.

Kay and Boardman's observations on the formation of clusters in dilute suspensions have been confirmed by studies by Slack (*9*), Hocking (*10*), Johne (*11*), Koglin (*12–15*), and Bhatty et al. (*16*, *17*). Studies by Maude and Whitmore (*18*) and Bhatty et al. (*16*) considered a cluster of particles as a single large particle of appropriate chemistry but of reduced rigidity. This may very well be attributed to the attached immobile liquid within the cluster which causes it to settle with a lower velocity.

Hawksley (*19*) earlier pointed out that a suitable correction factor could be applied to those formations in which particles are close enough so that the interparticle distance is smaller than their distance from the container wall. The resultant effect observed was a greater terminal velocity of the cluster due to reduction in drag on the individual particles. In a theoretical study on particle–particle interaction for a pair of spheres, Happel (*4*) calculated the ratio of the drag force exerted on either sphere to that exerted on a single sphere against $L/D$, the ratio of interparticle separation, where $L$ is the center-to-center distance between spheres of diameter $D$ (spheres touching when $L = D$). These calculations have been carried out for cases of a falling sphere (a) parallel and (b) perpendicular to their center lines. The results are shown in Table 2. For $L/D$ greater than 10, this interaction becomes negligible.

TABLE 2
Ratio of Drag Force Exerted on the Plane of Spheres to That Exerted on a Single Sphere for Cases of Spheres Falling (a) Parallel and (b) Perpendicular to Their Center Lines

| *L/D* | 1 | 2 | 3 | 4 | 5 | 6 | 7 |
|---|---|---|---|---|---|---|---|
| (a) | 0.65 | 0.73 | 0.80 | 0.83 | 0.86 | 0.80 | 0.91 |
| (b) | 0.70 | 0.83 | 0.87 | 0.91 | 0.93 | 0.94 | 0.95 |

Oseen (*20*) showed that for two identical spheres falling in a vertical line, the retardation on the following sphere is smaller than on the leading sphere, so that they move toward each other. A large sphere falling in a vertical line in the vicinity of a small sphere may pick up the small sphere so that it revolves around the large sphere as a satellite.

In cases where large spheres settle in a suspension of fine spheres, the fine particles act as a part of the fluid. The particle segregation occurs up to a limiting concentration when an interlocking mechanism prevents further segregation, and the suspension then settles "en masse." Lockett and Hobbooley (*21*), while working on polydispersed suspensions, reported segregation of particles, resulting in the formation of multiple layers within the suspension which correspond to different particle sizes. Later Bhatty et al., while working on polydispersed suspensions of glass ballotinis in aqueous glycerol, confirmed this finding (*22*).

While dealing with dilute suspension, Einstein (*23*) found that the change in settling velocity of a particle may be due to an increase in the viscosity of suspending liquid from $\eta$ to $\eta_s$:

$$\eta_s = \eta[1 + 2.5(1 - \varepsilon)] \tag{6}$$

where $(1 - \varepsilon)$ is the solid volume fraction, i.e., the volumetric fraction of the suspension occupied by the solid. According to Kynch (*24*, *25*), this formula requires modification for suspensions where $(1 - \varepsilon) > 1\%$.

For microsized particles, known as colloidal particles, interactions of a physical nature are far less significant because the particles are so small that the electrical forces at the particle surfaces become responsible for the particle–particle and particle–liquid interactions.

## INTERACTION OF COLLOIDAL SIZE PARTICLES

There are two principal forces which can be held responsible for the interaction between two particles in the colloidal size range: Coulombic and van der Waals. Coulombic forces are due to the net electrical charges of both particles and they may result in attraction or repulsion of the particles. These forces decrease with the square of the distance between the particles. Van der Waals forces are also based on electrical interactions. They do not involve two net charges, are always attractive, and decrease more rapidly with distance. They are due to the interaction of instantaneous asymmetric charge distribution on and about the particles.

When two molecules, each carrying a dipole, are close to one another, they mutually influence their orientation so that a net attraction results. This causes the particles to cluster into flocs. The magnitude of this force has been successfully worked out by London (*26*) and by Slater and Kirkwood (*27*) on the basis of wave mechanics. According to them, the potential of attraction between the two like molecules varies inversely as the sixth power of the distance. From the basic implication of wave mechanics treatment, the summation of the attractive potential between atomic planes of two particles consisting of many molecules can be achieved. In considering this evaluation for two spheres, each of radius $r$, at a nearest distance $D$, the attractive potential energy per pair of spheres, $w_A$, according to Verwey and Overbeek (*28*) is

$$w_A = -\frac{Ar}{12D} \tag{7}$$

where $A$ is a constant value which depends upon the natures of the material and the separating medium.

Bowman and Hughes (*29*) considered the effect of any dispersing medium on the interparticle reaction to be negligible, whereas according to Weisberg (*30*) the dielectric constant of the medium is of importance when the effect on interaction energy among neutral particles is considered. Davies, Dollimore, and Sharp (*31*) mentioned that there is no simple relationship between dielectric constant and flocculating behaviour of the medium; but for organic liquids, simple electrostatic considerations would suggest that the tendency to flocculation would be inversely proportional to dielectric constant.

The simplest expression for the charge interaction is given by the Poisson equation:

$$\nabla^2\psi = -\frac{4\pi d_c}{D_e} \tag{8}$$

where $\nabla^2$ = the Laplace operator $\frac{d^2}{dx^2} + \frac{d^2}{dy^2} + \frac{d^2}{dz^2}$

$\psi$ = electrical potential
$d_c$ = charge density in the double layer
$D_e$ = liquid dielectric constant

The rate of decrease in $\psi$ with distance from the particle surface is thus inversely related to the liquid dielectric constant, and (other things being equal) the range of the effect of the electrostatic charge will be greater in liquids of higher $D_e$. Thus the electrostatic hindrance is expected to be at a maximum with charged or polar particles of large surface area per gram in polar solvents, and at a minimum with uncharged or nonpolar solids of small specific surface in nonpolar solvents. For instance, the behavior of calcium carbonate suspensions in liquids of different dielectric constants ($D_e$) has been presented by Davies (*32*) who noted the change in the flocculation factor $n$ as a function of $D_e$. Factor $n$ is given in a generalized Richardson and Zaki's (*33*, *34*) expression as

$$Q = V_s \varepsilon^n \tag{9}$$

where $Q$ is the settling rate (cm/s) of the suspension, $V_s$ is the Stokes' terminal velocity (cm/s), and $\varepsilon$ is the suspension porosity. An increase in $n$ corresponds to an increase in flocculation of particles in suspension.

Data for calcium carbonate suspension in various liquids is shown in Table 3. Liquids with higher dielectric constants ($D_e$) generally show higher values of $n$, and therefore have a higher tendency to flocculate. However, higher values of $n$ with low $D_e$ liquids such as benzene, ether, and ethyl acetate have revealed even further complexities. The correla-

TABLE 3
Values of Flocculation Factor $n$ as a Function of Dielectric Constants of Different Liquids Used in Calcium Carbonate Suspensions

| Liquids | Dielectric constant ($D_e$) | Flocculation factor $n$ |
|---|---|---|
| Benzene | 2.3 | 48.6 |
| Diethyl ether | 4.3 | 35.6 |
| Ethyl acetate | 6.4 | 36.1 |
| Ethyl acetoacetate | 15.9 | 38.2 |
| Acetone | 21.4 | 24.4 |
| Water | 78.0 | 62.2 |
| 0.137 *M* NaCl | >78.0 | 73.9 |
| 0.915 *M* NaCl | >78.0 | 74.2 |

tion between the dielectric constant and the hydrodynamic interaction between the particles is therefore not straightforward, and it requires further investigations. Davies (*31*) suggested that most probably the mechanism of interaction observed with higher $D_e$ liquids in calcium carbonate suspensions is of electrostatic origin.

## CONCEPT OF DOUBLE LAYER

The boundary between a solid particle and the surrounding liquid is assumed to be the location of physical and chemical forces, known as boundary forces, which depend upon the properties of both phases. A solid particle in an aqueous environment acquires a surface charge due either to the disassociation of surface groups or the preferential adsorption of ions. This charge is then balanced by an opposite charge of counterions. This leads to the existence of a double layer, first suggested by Helmholtz (*35*), whose views were later modified by Gouy (*36*) and Chapman (*37*) who considered the second layer as a diffused one in which the ions of opposite charge (the counterions) are concentrated near the surface while the ions with similar charges (the co-ions) are repelled. Hence, there is an excess of counterions over co-ions in the vicinity of the surface, but this ion concentration decreases toward the bulk of the solution where it becomes uniform. Stern (*38*) further modified this theory by introducing a correction to the ionic thickness of the layer immediately adjacent to the surface (the Stern layer). This layer is considered to be held firmly to the surface through the electrostatic forces, and it is strong enough to overcome thermal agitations. Thus, the resulting electrical double layer consists of a compact Stern layer adjacent to the surface and a diffused layer, called the Gouy layer, immediately outside it.

Only this diffuse layer of ions is free to move. A free particle will tend to fall through its diffuse layer, leading to a polarization of charge distribution (Fig. 2) with a consequent diminution in the sedimentation rate. The electric potential gradients on and near a solid–liquid interface are represented schematically in Fig. 3, which is based on the work of Stern (*38*). The Stern layer is a layer of specifically adsorbed ions and of ions located within a distance from the solid surface. Following Kruyt (*39*), it is assumed that the Stern layer is immovably attached to the particle, moving with it during particle movement. It is also assumed that the zeta potential measures the electrical potential at the effective slipping plane between the particle (and associated fluid) and the bulk fluid.

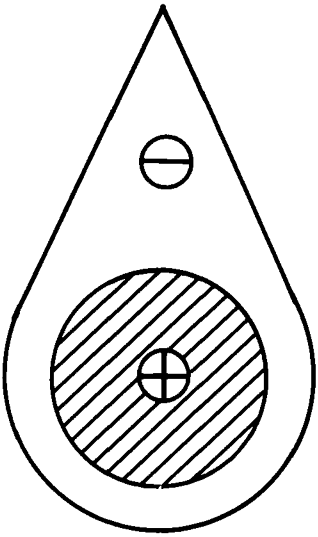

FIG. 2. Sedimentation of a solid particle through its diffuse layer of charge (schematic).

Analysis of the electroviscous problem for suspensions is much more complex than for single particles because the effects of electrostatic interactions between the particles, and of flow round the particles, must be taken into account. Sengupta (*40*) indicated that if the double layer thickness is much greater than the mean separation between particles and there is laminar flow, the suspension may be regarded as an ordinary electrolyte, the particles being merely exceptionally large and highly charged ions.

For the case where the double layer thickness is small compared to the mean separation, and again with laminar flow, the potential near any given particle will be undisturbed by electrical effects from other particles. However, the retardation field of the particle, arising from the polarization of the otherwise symmetrical field by fluid flow, will be supplemented by an additional consideration representing the interactions between electrostatic fields and the effect of the fluid flows round the particles.

The potential between the Stern layer and the diffused layer (Gouy layer) is probably very close to the electrokinetic potential, frequently called the zeta potential ($\zeta$), i.e., the potential difference between the rigid solution layer and the mobile part of the solution adjacent to the bulk

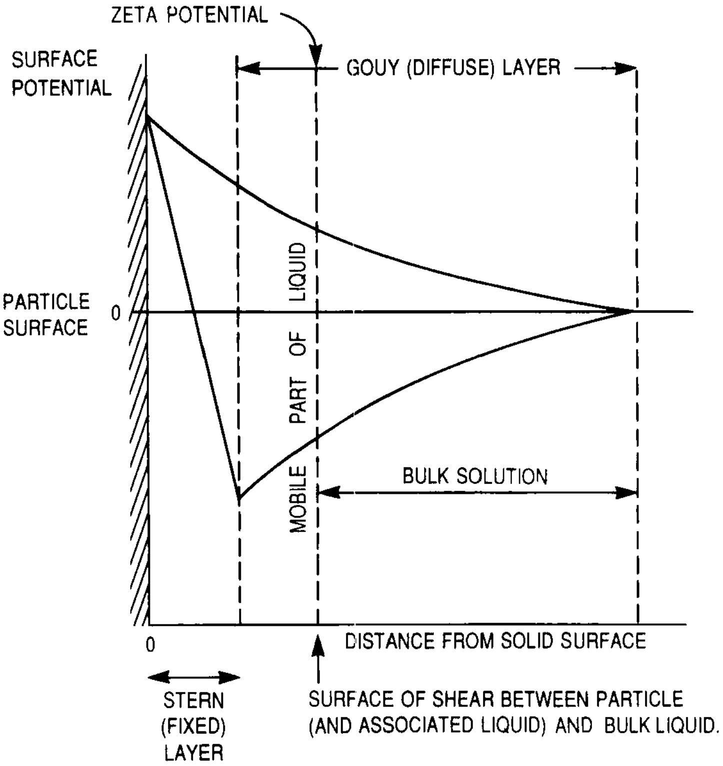

FIG. 3. The electrical potential gradients on and near a solid-liquid interface of a suspended particle (schematic).

solution. The important application of the zeta potential is the estimation of the surface charge of solid particles in an electrolyte solution.

A change in the composition of the medium results in a change in the zeta potential; sometimes in an increase, sometimes in a decrease, and sometimes even a reversal of sign.

If an ion is preferentially adsorbed by the solid particle from the medium or is preferentially diffused from the solid into the medium, this is a potential-determining ion of particular significance. The charge on the particle is therefore sensitive to a change in the ionic concentration of the medium, which may easily be brought about by changing the pH of the medium. At a certain pH value the zeta potential is nil and the surface is neutral; this is at zero point of charge (z.p.c.), which differs for different materials.

## APPENDIX

The Reynolds number, Re, is an absolute number lacking in physical dimensions. Dimensionally:

$$\mathrm{Re} = \frac{2\nu \upsilon \rho_l}{\eta}$$

where $r$ = radius of the solid particle
$\upsilon$ = velocity of the particle
$\rho_l$ = density of the liquid
$\eta$ = viscosity of the liquid

This was derived by Osborne Reynolds and has since become indespensible in fluid mechanics. Stokes' law is accurately valid up to Re = 0.6. In contrast, the Newton-Rittinger relationship is approximately valid from Re = 800 to Re = 200,000. From Re = 0.6 to Re = 800, no theoretical formula is valid.

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