Interparticle Potential and Sedimentation of Monodisperse Colloid System

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The effect of interparticle potential on the sedimentation of a colloid system has not been solved completely. A key problem is how to deal with the interparticle potential in stable systems. Based on previous works, an interparticle potential function for a stable system that cuts off the potential interaction at the position of the potential barrier (the position where interparticle potential is equal to 15 kT) is presented. Adopting this potential function, the sedimentation coefficients of monodisperse systems containing potential interacting particles are calculated, and the theoretical results are compared with those given by experiments. The potential function for the adopted stable system is more useful and reasonable than previous ones.

Introduction

The sedimentation of a colloid system of particles under gravity has been studied extensively because of its importance in practical application. The problem that has received the greatest attention is the sedimentation of a monodisperse suspension at small Reynolds number.

Batchelor (1972) derived the hindered settling formula for dilute and monodisperse colloid system, which may be written as

$$\overline{U} = U_0(1 - 6.55\phi). \tag{1}$$

Here $\overline{U}$ is the mean hindered settling velocity, U_0 is the Stokes terminal velocity of an isolated particle, ϕ is the volume fraction of the particles, and -6.55 is the sedimentation coefficient S of hard spheres. Numerous experiments have shown that the linear relation between the average change in the mean settling velocity and ϕ is correct, but at that time many experimental sedimentation coefficients were larger than -6.55 and varied from -6 to -4. The main factor causing the deviation is that Batchelor's 1972 theory was proposed for hard spheres. The interparticle potential was given in a very simple form and the statistical distribution of the particles was assumed uniform. In most sedimentation experiments, however, there are complex potential interactions among ordinary particles. The distribution of the particles is usually nonuniform.

Experimental confirmations of Eq. 1 were made only after Batchelor's 1972 work. For example, for fd bacteriophage DNA, Newman et al. (1974) obtained the experimental value $S = -6.7 \pm 0.8$. For silica spheres that behave thermodynamically as hard spheres, the sedimentation coefficient observed by Kops-Werkhoven et al. (1982) was -6.6 ± 0.6. Al-Naafa and Selim (1992) measured $S = -6.51 \pm 0.6$ using a coated silica sphere system. All these data agree with Batchelor's theoretical value quite well. On the other hand, the problem of the sedimentation of ordinary particles with interparticle potential remains unsolved.

Analysis on Previous Works

Sedimentation of ordinary colloid particles is strongly affected by potential interactions. If the repulsion exceeds the attraction, the sedimentation coefficient of the system is smaller than that of the hard spheres; if the attraction exceeds the repulsion, the reverse occurs. A colloid system may even change from a stable one to an unstable one if the attraction is too much stronger than the repulsion, leading to strongly accelerated sedimentation. Although it is important to pay full attention to the effects of the potential interactions when studying sedimentation phenomena, previous workers made limited progress due to the lack of good de-

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scriptions of interparticle potential for a stable system. Previous investigations—including model methods and the method adopting approximate potential expressions—are listed below.

1. "Hard sphere" model (Batchelor, 1972). This model neglects potential interactions unless two particles, whose potential function may be written as

$$\Phi = \begin{cases} \infty & (s = 2) \\ 0 & (s > 2), \end{cases} \tag{2}$$

come in contact. Here s is the center-to-center separation of two particles made dimensionless on the average radius of the particles. Generally speaking, this model is too simple for the ordinary experimental situation.

2. Born cutoff method (Goldstein and Zimm, 1971; Reed and Anderson, 1980). When the potential interactions include the van der Waals attraction, an important problem is how to eliminate the divergence of the attractive potential at the near field. Because of the Born repulsive force among the electron clouds, Goldstein and Zimm and Reed and Anderson gave the small distance $\delta = 0.5$ nm to cut off the potential interactions. The potential function of this method may be expressed as

$$\Phi = \begin{cases} \infty & (s < 2 + \delta/a), \\ V_R + V_A & (s \geq 2 + \delta/a), \end{cases} \tag{3}$$

in which V_R and V_A denote the double-layer repulsive potential and the van der Waals attractive potential, respectively, and a denotes the radii of the particles.

This method was used in those authors' articles to extract the Hamaker constant from sedimentation data observed in Experiment 1 (Table 1). Our study has shown, however, that the Born cutoff method is valid only for small particles. For large particles it cannot interpret the sedimentation data of a stable system because a deep primary minimum probably appears at the near field due to the small absolute thickness of the Born repulsion barrier. But the Born cutoff isn't the only difficulty in this method, as any other kind of repulsion that has an absolute cutoff parameter whose magnitude is most likely to be angstrom (Feke et al., 1984) will cause the same problem. (The radii of the particles used in Experiment 1 were mistakenly used as diameters in the articles of Goldstein and Zimm (1971) and Reed and Anderson (1980).)

Table 1. Two Monodisperse Sedimentation Experiments Made in Polystyrene Suspension

	Experiment 1 Cheng and Schachman (1955)	Experiment 2 Buscall et al. (1982)
T*	290 K	290 K
I	0.098 M	0.001 M
a	0.13 μm	1.55 μm
ξ_0	0.0077	0.0065
S	−5.1	-5.4 ± 0.1

*T is the absolute temperature of the system, I is the electrolyte concentration, and ξ_0 the dimensionless double-layer thickness scaled on the average radius.

Table 2. Numerical Results on the Experiments in Table 1

	Experiment 1 Cheng and Schachman (1955)	Experiment 2 Buscall et al. (1982)
Sedimentation Coefficient S		
Exp. value	−5.1	-5.4 ± 0.1
Theor. value		
Potential function (12)	−5.1	−5.0
Russel's model (5)	−4.5*	−6.5
Double-Layer Thickness ξ_0		
Exp. value	0.0077	0.0065
Theor. value		
Potential function (12)	0.0077	0.008
Batchelor's model (4)	0.02	0.009

*Value given by Russel et al. (1989) is −5.0. However, using their data of s_0 and τ, following their formula of sedimentation coefficient, one can only obtain $S = -4.5$.

3. Multisection model (Batchelor and Wen, 1982). This model made great progress by taking into account both double-layer repulsion and the van der Waals attraction, although some simplifications were made. The potential function of this model may be expressed as

$$\Phi = \begin{cases} \infty & [0 < (s-2) < \xi_0], \\ V_A & [\xi_0 \leq (s-2) < 0.2], \\ 0 & (\xi_0 \geq 0.2). \end{cases} \tag{4}$$

The influence of the double layer was expressed by the variation of the dimensionless double-layer thickness, ξ_0. If the thickness was less than ξ_0, infinite repulsion was assumed to exist; the effect of the van der Waals attraction was assumed to be dominant for $\xi_0 \leq \xi < 0.2$; and for $\xi \geq 0.2$, any potential interactions were neglected. Batchelor and Wen (1982) investigated the two experiments in Table 1 using the multisection model. Their work showed that particles in close pairs, which were caused by van der Waals attraction, fell more quickly than well-separated pairs due to direct hydrodynamic interactions, and therefore a larger number of close pairs could make the sedimentation coefficient larger than −6.55. But the decision to cut off the double-layer repulsion at ξ_0 is a difficult one. The double-layer potential is surely active when $\xi \geq \xi_0$. Thus, the numerical result of this model still deviates from experimental data (see Table 2).

4. Baxter model (Russel et al., 1989). This model is the so-called adhesive excluded shell model, which may be expressed as

$$\frac{\Phi}{kT} = \begin{cases} \infty & (s < s_0), \\ -\ln \dfrac{\delta(s - s_0)}{6\tau} & (s = s_0), \\ 0 & (s > s_0). \end{cases} \tag{5}$$

Here k is the Boltzmann constant and δ is the delta function. Potential interactions in this model were assumed to focus on a shell where $s = s_0$. s_0 is the shell radius and $1/\tau$ is the stickiness. The two parameters are determined by the integrals of the interparticle potential, namely,

$$\frac{s_0^3}{2} = 4 + \frac{3}{2}\int_2^{\sigma} [1 - \exp(-\Phi/kT)] \cdot s^2 \, ds \qquad (6)$$

and

$$\frac{1}{\tau} = \frac{6}{s_0^2}\int_{\sigma}^{\infty} [\exp(-\Phi/kT) - 1] \cdot s^2 \, ds. \qquad (7)$$

Here σ is a specific distance as opposed to $\Phi(\sigma) = 0$.

Taking advantage of its strange form, the adhesive excluded shell model was used to deduce an analytical form of the sedimentation coefficient, but it didn't provide a clear physical meaning and couldn't actually separate from the detailed potential function. Russel et al. (1989) used the model to investigate the two experiments in Table 1. As is indicated in Table 2, their numerical results of the sedimentation coefficient still deviate from experimental data.

5. Potential function including only double-layer repulsion and neglecting van der Waals attraction (Petsev and Denkov, 1992; Nägele, 1996). Adopting a mean force repulsive potential, Petsev and Denkov (1992) derived a simple analytical formula for the sedimentation coefficient. We can easily find that the sedimentation coefficient derived from their formula (Petsev and Denkov, 1992, Eq. [2.37]) can only be smaller than -6.55, and that it cannot explain why the experimental sedimentation coefficient of ordinary particles like that used in Experiments 1 and 2 is larger than -6.55. This result agrees with Batchelor and Wens' (1982) conclusion—which was confirmed by Russel et al. (1989)—that without the van der Waals attraction, the sedimentation coefficient of a monodisperse system would certainly be smaller than -6.55. The investigations made by Nägele (1996), who adopted a repulsive Yukawa potential and neglected the attractive potential of strongly charged colloid particles, cannot interpret the experimental data of Experiments 1 and 2 either.

Practical Way to Deal with Interparticle Potential in Stable Systems

Most sedimentation experiments have been made in a hydrosol system. Particles in a hydrosol system hold like charges on their surface, and each particle is surrounded by a counterion layer. This structure is the well-known double layer. Besides the repulsive electrical double-layer potential, van der Waals attraction exists between two neighboring hydrosol particles. The growth and decline of the relative strength of the two different type potentials determines whether sedimentation coefficient is larger or smaller than -6.55 of the hard spheres.

In monodisperse systems, a commonly used double-layer repulsive potential has the form (Russel et al., 1989)

$$V_R = 2\pi\epsilon\epsilon_0 a\psi_S^2 \ln[1 + \exp(-\kappa(s-2))], \qquad (8)$$

where ϵ is the relative dielectric constant of the medium, ϵ_0 is the dielectric constant of the vacuum, ψ_S is the surface potential, and κ is the reciprocal of the dimensionless double-layer thickness ξ_0. It is known that Eq. 8 is valid for only the small double-layer thickness and the low constant surface potential (Zeichner and Schowalter, 1977). At room temperature, for a hydrosol system containing 1-1 electrolyte, such as NaCl or KCl, it demands that $\psi_S < 51.4$ mV. Since the numerical investigations in this artical involve high surface potential, a new expression of double-layer potential advanced by Wang and Jin (1996) is used for comparison with experimental data. It is

$$V_R = 32\pi\epsilon\epsilon_0\left(\frac{kT}{ze}\right)^2 a \cdot \frac{2}{s} \cdot \left[\gamma^2\left(1 - \frac{2}{3}\gamma^2\right)\ln(1 + e^{-\kappa(s-2)}) + \frac{2}{3}\gamma^4 e^{-\kappa(s-2)} + \frac{\gamma^4(1+2\gamma^2)}{12(1-\gamma^2)}\ln(1 + e^{-4\kappa(s-2)})\right], \qquad (9)$$

where $\gamma = \tanh(e\psi_S/4kT)$, e is the electron charge, and z is the valence of the electrolyte. In fact, Eq. 9 approaches Eq. 8 if the surface potential is very low. The data given by Wang and Jin showed that Eq. 9 was accurate for a moderate potential as high as 102.8 mV.

The Hamaker expression of the van der Waals attractive potential is

$$V_A = A \cdot \tilde{V}_A = -\frac{A}{6}\left[\frac{2}{(s^2-4)} + \frac{2}{s^2} + \ln\frac{(s^2-4)}{s^2}\right], \qquad (10)$$

where A is the composite Hamaker constant and $\tilde{V}_A$ represents the dimensionless van der Waals attractive potential. According to DLVO theory, one obtains the total interparticle potential by adding Eq. 9 to Eq. 10,

$$\Phi = V_R + V_A. \qquad (11)$$

In order to calculate the effects of the interparticle potential on the sedimentation coefficient, one should first solve the divergence problem of the interparticle potential at near field. As we know from Eq. 11, when $(s-2) \to 0$, the van der Waals potential approaches negative infinity and double-layer potential approaches a finite positive value, so the total interparticle potential approaches negative infinity. As a consequence, the pair distribution function and the sedimentation integration (see Eqs. 15 and 16) are all divergent at near field.

Our study has shown that the idea of introducing strong Born repulsion into the interparticle potential at the molecular scale was limited. A more reasonable way is: in a stable system, cutoff the potential interactions at the position where $\Phi = 15kT$, provided that a high potential barrier exceeding $15kT$ exists. The existence of a high potential barrier is the condition of a system's stability. Particles in a stable system cannot cross the high potential barrier and be caught by the primary minimum. The potential function of this method has the form

$$\Phi = \begin{cases} \infty & (s < s_{15kT}), \\ V_R + V_A & (s \geq s_{15kT}), \end{cases} \qquad (12)$$

in which s_{15kT} is the position where $\Phi = 15kT$, and V_R and V_A are formulated by Eqs. 9 and 10, respectively. In fact, it is important to eliminate the influence of the primary mini-

mum, but which position should be chosen as the cutoff position—s_{15kT} or s_{20kT}—brings only slight deviation to the numerical result of the sedimentation coefficient. Also, $\Phi = 15kT$ can be treated as the typical value of the potential barrier to ensure the stability of a colloid system.

Without the absolute cutoff distance, the potential function (Eq. 12) can be freely used in studying the sedimentation of interacting particles, whether the particles are small or large. Other potential function curves that were discussed previously are shown in Figure 1, in which curve 1 shows the behavior of the potential function (Eq. 12) presented in this article.

Numerical Results and Discussion

In the study of the effects of the interparticle potential on the sedimentation of colloid particles, the monodisperse case is the simplest and the most appropriate one to show the nature of the action of the interparticle potential. An important equation, which describes the statistical distribution of the particles in a colloid system, is the equation for the pair-distribution function p_{ij} (Wen, 1996), namely,

$$\frac{\partial p_{ij}}{\partial t} = -\nabla\cdot(\boldsymbol{V}_{ij}p_{ij}) + \nabla\cdot\{p_{ij}\boldsymbol{D}_{ij}\cdot\nabla(\Phi_{ij}/kT)\} + \nabla\cdot(\boldsymbol{D}_{ij}\cdot\nabla p_{ij}). \tag{13}$$

Here the pair-distribution function, p_{ij}, is defined as the probability that the center of a particle j (with radius a_j and density ρ_j) lies within the unit volume at position $\boldsymbol{r}$ relative to the center of the test particle i (with radius a_i and density ρ_i); $\boldsymbol{V}_{ij}$ is the relative velocity of the two particle centers due to the imposed gravitational force; $\boldsymbol{D}_{ij}$ is the relative Brownian diffusivity tensor of the two particles; and Φ_{ij} denotes the interparticle potential.

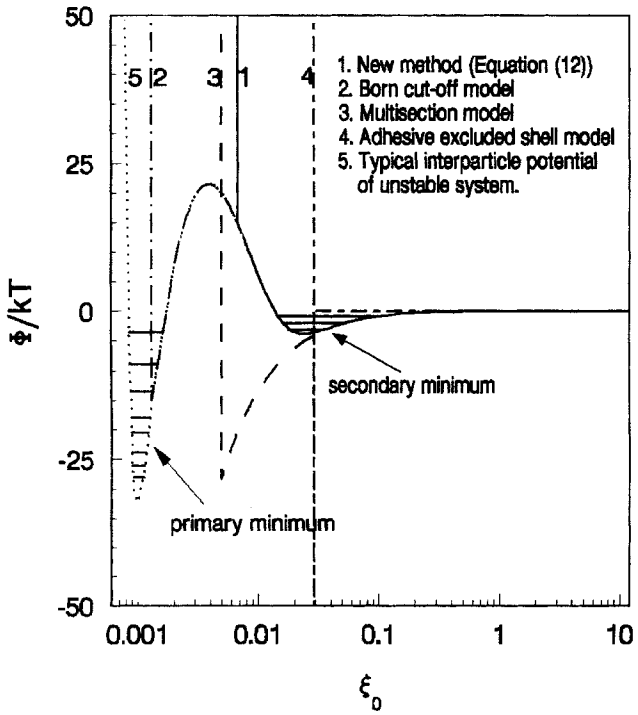

Figure 1. Various potential functions.

Under the special condition of monodisperse stable system, Eq. 13 reduces to

$$\nabla\cdot\{p_{ij}\boldsymbol{D}_{ij}\cdot\nabla(\Phi/kT)\} + \nabla\cdot(\boldsymbol{D}_{ij}\cdot\nabla p_{ij}) = 0, \tag{14}$$

where Φ is a special form of the interparticle potential, Φ_{ij} in the monodisperse system. The strict solution of this differential equation for a stable system is the Boltzmann distribution

$$p_{ij} = \exp(-\Phi/kT). \tag{15}$$

According to Batchelor and Wen's (1982) theory, the sedimentation coefficient can be written as

$$S = -6.55 + \int_2^{\infty}(A_{11} + 2B_{11} + A_{12} + 2B_{12} - 3)_{\lambda=1} \times\{\exp(-\Phi/kT) - 1\}\cdot s^2\, ds, \tag{16}$$

where the integral on the righthand side denotes the deviation term from the sedimentation coefficient of the hard spheres, and A_{11}, A_{12}, B_{11} and B_{12} are the four longitudinal (A) and transverse (B) mobility functions, respectively, given by Jeffrey and Onishi (1984).

We have used the two monodisperse sedimentation experiments given in Table 1 to examine our potential function (Eq. 12), and the numerical results are shown in Table 2. The composite Hamaker constant of polystyrene–water suspension takes the average value 7×10^{-21} J of different experimental data (Zeichner and Schowalter, 1979; Russel et al., 1989). The surface potential takes the values assumed by Russel et al. (1989), that is, -75 mV for Experiment 1 and -25 mV for Experiment 2.

Obviously, the results shown in Table 2 indicate that the potential function (Eq. 12) suggested in this article is better than the potential functions mentioned earlier. As we have found, the key point to solving the sedimentation problem in stable systems was finding a suitable size for the potential interactions. Analysis of Experiments 1 and 2 indicates that s_{15kT} is about 0.025 (3.25 nm for Experiment 1 and 39 nm for Experiment 2) from the particle surface, and that the position of the secondary minimum is about 0.05 (6.5 nm for Experiment 1 and 78 nm for Experiment 2) from the particle surface. The Born cutoff distance of method 2 is much smaller than that given by Eq. 12 for large particles. The double-layer cutoff distance assumed by model 3 is also smaller than that given by Eq. 12, and eventually the assumptions of this model overestimate the van der Waals attraction, so the theoretical thickness of the double layer deduced from it is surely larger than experimental data. The adhesive and excluded shell radius of model 4 does not provide a clear physical meaning, and the numerical result deduced from this model did not agree with the experimental data very well.

Further calculations have been carried out to show the effects of the interparticle potential on the sedimentation phenomena. Monodisperse polystyrene suspension is chosen as

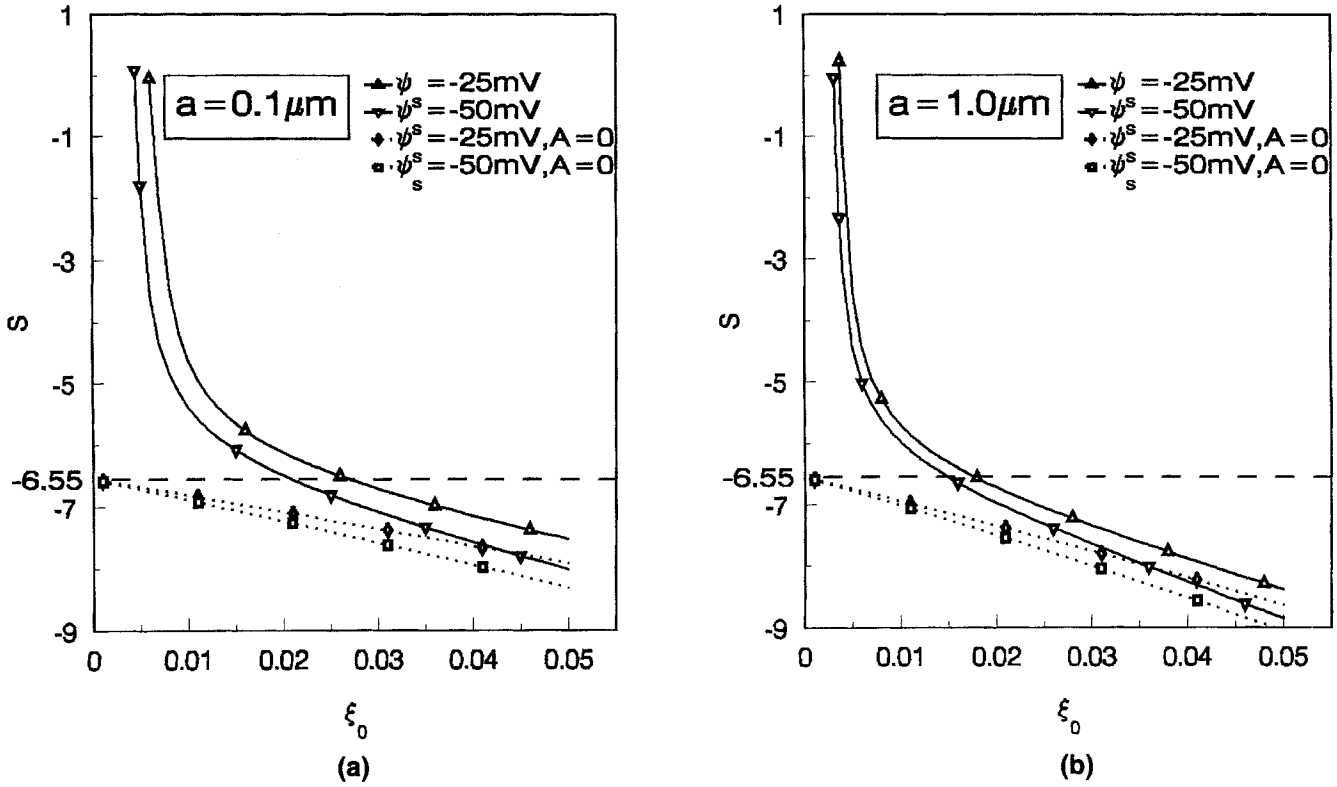

Figure 2. Sedimentation coefficient as a function of the dimensionless double-layer thickness ξ_0 on the condition of fixed ψ_S according to the new method.

the sample system, and the calculations involve two different particle sizes, 0.1 μm and 1.0 μm. In Figure 2 we show the variations of the sedimentation coefficient on the dimensionless double-layer thickness on the condition of the fixed surface potential. The sedimentation coefficient increases sharply for $\xi_0 < 0.01$, because many particles in close pairs, which fall more rapidly than well-separated pairs, appear in the colloid system. If $S = 0$ is used as the stability criterion, the values of ξ_0 giving rise to this result can be regarded as the critical dimensionless double-layer thickness $\xi_{0,C}$. When ξ_0 is smaller than the critical value $\xi_{0,C}$, the stable system becomes unstable, hindered settling becomes enhanced settling, and the particles are separated from the dispersion system much more rapidly than in the case of $\xi_0 > \xi_{0,C}$. The critical dimensionless double-layer thickness of the system seen in Figure 2 are given in Table 3. The $S \sim \xi_0$ curves neglecting the van der Waals attractive potential (namely, $A = 0$) are also shown in Figure 2. And, as we can see, these sedimentation coefficient data are all smaller than those of the hard spheres—just as has been discussed by Batchelor and Wen (1982). These curves show once again that the potential model that neglects the van der Waals attractive potential cannot explain why the experimental sedimentation coefficients of ordinary particles can be larger than -6.55.

Curves of $S \sim \psi_S$ for fixed ξ_0 are shown in Figure 3. The variation in the sedimentation coefficients is shown only in the limited range of ξ_0 and ψ_S, since a ξ_0 or ψ_S that is too small can lead to the disappearance of the potential barrier and hence an unstable system. As we see in Figure 3, most sedimentation coefficients are larger than -6.55 when ξ_0 or ψ_S is small enough. Figure 2 also shows us that S increases with the decrease of ξ_0 for fixed ψ_S, and in Figure 3 we see that S decreases with the increase of ψ_S for fixed ξ_0. It is in keeping with common sense that when ψ_S is fixed, a thinner double layer means an increased attraction and more close pairs falling more quickly than well-separated pairs, which means the sedimentation coefficient is larger; when ξ_0 is fixed, the larger surface potential means a stronger repulsion and fewer close pairs, which means the sedimentation coefficient is smaller.

Table 3. Critical Double-Layer Thickness

Radius a	0.1 μm		1.0 μm	
Surface potential ψ_S	-25 mV	-50 mV	-25 mV	-50 mV
Critical $\xi_{0,C}$	0.006	0.005	0.004	0.003

Conclusion

A new and practical way of dealing with the interparticle potentials in stable colloid systems, whose potential function was expressed by Eq. 12, has been put forward in this article. The new method is clear in its physical meaning, and theoretical values of the sedimentation coefficient deduced by the new method agreed with the experimental data better than previous investigations. The validity of the potential function

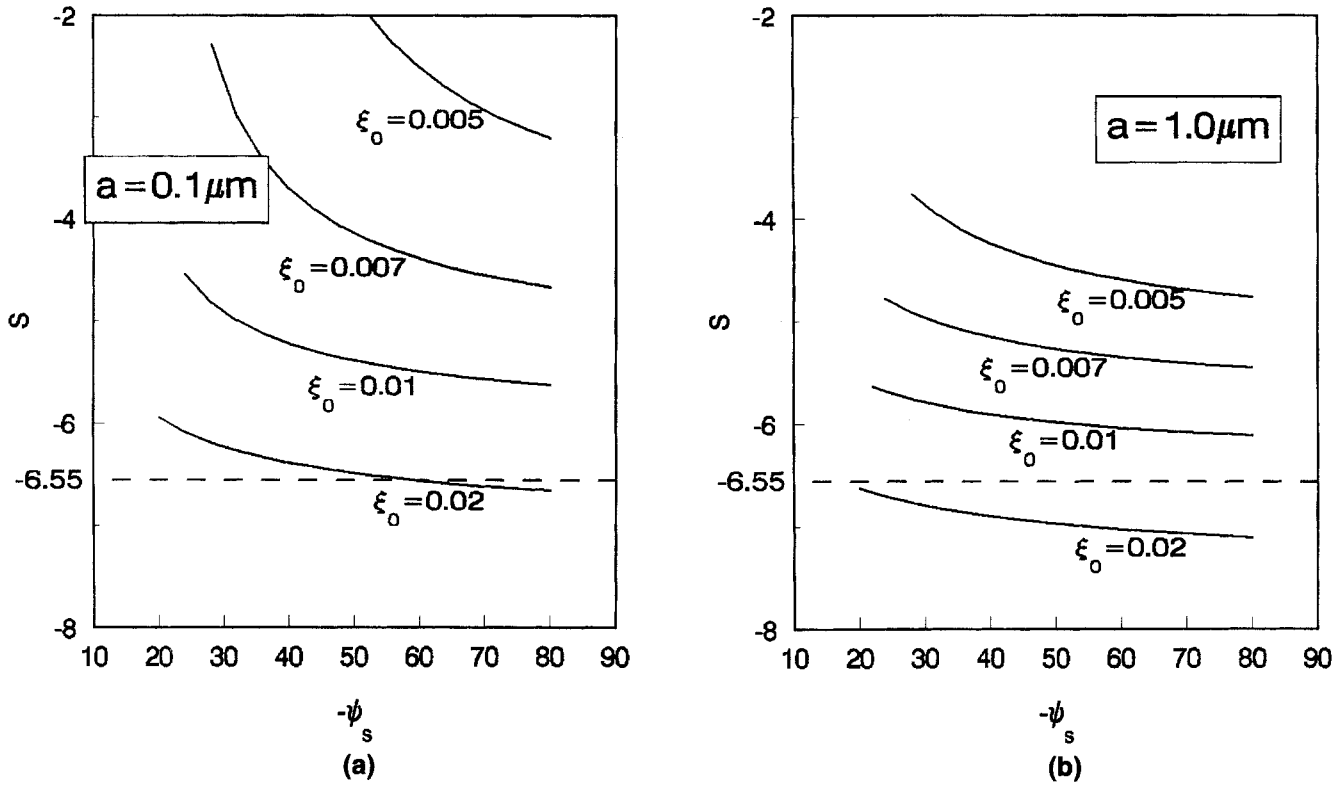

Figure 3. Sedimentation coefficient as a function of the surface potential ψ_S on the condition of fixed double-layer thickness ξ_0 according to the new method.

(Eq. 12) implies that, although it is important to particle coagulation in unstable systems, any complex potential interactions between the particle surface and the high potential barrier do not affect the sedimentation phenomena in stable systems. In this sense, one can more easily consider the effects of the interparticle potential for a stable system than that for an unstable system. The potential function (Eq. 12) is believed also to be valid for further research on the sedimentation of a polydisperse system.

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