Electrophoretic Motion of a Charge-Regulated Sphere Normal to a Plane

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The electrophoresis of a charge-regulated spherical particle normal to a planar surface is analyzed theoretically. The present study extends previous analyses in that the surface of the particle assumes a general charged condition, which simulates biocolloids and particles covered by an artificial membrane, and an arbitrary, double layer thickness κa , κ , and a being respectively the reciprocal Debye length and particle radius. We conclude that the thinner the double layer and/or the larger the particle-surface distance, the greater the mobility of the particle. If the concentration of the dissociable functional groups on particle surface N_s is low, the absolute mobility of a particle decreases with the increase in κa , and the reverse is true if N_s is high. For a medium N_s , the absolute mobility has a local maximum as κa varies. We show that the higher the pH of the liquid phase, the smaller the absolute mobility. © 2001 Academic Press

Key Words: electrophoresis; charge-regulated particle; boundary effect; particle-planar surface interaction.

INTRODUCTION

The electrophoresis of spherical particles has been studied extensively for both dilute dispersions (1–9) and concentrated dispersions (10–14). Reported results range from analytical solution for low potential and thin double layer to numerical solution for arbitrary potential and double layer thickness. The effect of the presence of a boundary on the electrophoretic behavior of a particle has also been examined. Zydeny (15) and Lee *et al.* (16, 17), for example, discussed the electrophoretic behavior of a sphere in a spherical cavity. Morrison and Stukel (18) considered the electrophoresis of a sphere normal to a plane for the case of thin double layer and low surface potential. The problem was solved by adopting bispherical coordinates, and it was concluded that the presence of the plane has the effect of reducing the mobility of the particle, and the closer the distance between the particle and the plane the more significant the effect. Keh and Lien (19), however, pointed out that the analysis of Morrison and Stukel is flawed. Keh and Anderson (20) examined the boundary effect of electrophoresis by considering

three types of problems: a particle moves normal to a plane, a particle moves parallel to a plane, and a particle moves along the axis of a cylindrical cavity. A reflection method was applied, and an analytical solution was derived under the conditions of low electrical potential and infinitely thin double layer. They conclude the following: (a) The flow field has the effect of reducing the electrophoretic velocity of a particle. (b) The presence of a boundary has the effect of changing the electrical interaction between a particle and the applied electric field. (c) If the boundary is charged, an electroosmotic effect will be induced. The analysis was extended by Ennis and Anderson (21) to a finite, double layer thickness, but the double layer is not allowed to reach the boundary. It was found that the double layer surrounding a particle has the effect of reducing its electrophoretic velocity. Feng and Wu (22) considered the electrophoretic motion of an arbitrary prolate body of revolution normal to a conducting plane under the condition of a thin double layer. In a recent study, Shugai and Carnie (23) adopted the method of Teubner (24) to solve the electrophoresis of a colloidal particle normal to a plane for the case of low electrical potential and arbitrary, double layer thickness. Two types of boundary conditions are considered: constant surface potential and constant surface charge density. It was shown that when the double layer is close to the boundary the method adopted is more accurate than the reflection method. As pointed out by Krozal and Saville (25), constant surface potential and constant surface charge density are idealized conditions which represent two limiting cases, and in practice, the charged condition is between these two limits.

In the present study, we consider the electrophoresis of a charge-regulated sphere normal to a planar surface. If the electrical potential is low, the surface condition of the latter can be approximated by a linear combination of constant potential and constant charge. In other words, the results in the literature are extended to a more general case. A typical example of the type of particle examined includes biocolloids, such as biological cells and microorganisms, and particles covered by an artificial membrane layer. In addition to a more detailed understanding about the boundary effect on electrophoresis, the present analysis has potential applications in processes which involve the interaction between a charged entity and a surface—For instance, electrodeposition and cellular adhesion to surfaces, to name a few.

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THEORY

We consider the electrophoresis of a sphere of radius a toward a planar surface. The distance between the center of the particle and the surface is h . The liquid phase contains $z_1 : z_2$ electrolyte, z_1 and z_2 being respectively the valence of cations and that of anions. The electroneutrality in the bulk liquid phase requires that $n_{20} = n_{10}/\alpha$, n_{10} and n_{20} are respectively the bulk concentration of cations and that of anions, and α is constant. Referring to Fig. 1, a uniform electric field $\mathbf{E}_z$ with strength E_z is applied in the Z direction and the particle moves with velocity $\mathbf{U}$. The bispherical coordinates (ξ, η, ϕ) are adopted in which $\eta = 0$ represents the planar surface and $\eta = \eta_0$ represents the surface of the particle. The present coordinates are related to the cylindrical coordinates (ω, θ, Z) by (24)

$$z = c \frac{\sinh \eta}{\cosh \eta - \cos \xi} \quad [1]$$

$$\omega = c \frac{\sin \xi}{\cosh \eta - \cos \xi}, \quad [2]$$

where c is the focal length and $0 \leq \eta \leq \eta_0$, $0 \leq \xi \leq \pi$. Suppose that the liquid phase is incompressible with constant physical properties.

We assume that the electrical potential of the system under consideration can be described by the Poisson equation

$$\nabla^2 \phi = -\frac{\rho}{\varepsilon} = -\sum_{j=1}^2 \frac{z_j e n_j}{\varepsilon}, \quad [3]$$

where ρ and ε are respectively the space charge density and

FIG. 1. The bispherical coordinates (ξ, η, ϕ) adopted, where $\eta = 0$ and $\eta = \eta_0$ represent respectively the plane and particle surface. a is particle radius and h is the distance between the center of the particle and the plane. A uniform electric field $\mathbf{E}_z$ is applied in the Z direction, and the particle moves with velocity $\mathbf{U}$.

the permittivity of the liquid phase, e is the elementary charge, and n_j and z_j are respectively the number concentration and the valence of ionic species j .

Suppose that the flow field can be described by the Navier–Stokes equation in the creeping flow regime

$$\bar{\nabla} \cdot \mathbf{v} = 0 \quad [4]$$

$$\mu \nabla^2 \mathbf{v} - \bar{\nabla} p - \rho \bar{\nabla} \phi = 0, \quad [5]$$

where p and μ are respectively the pressure and the viscosity.

For convenience, ϕ is expressed as a sum of ϕ_1 and ϕ_2 (6). The former represents the potential in the absence of applied electric field, that is, the equilibrium potential, and the latter is a perturbed potential, which arises from the applied electric field. Suppose that the spatial variation of the ionic concentration at equilibrium follows the Boltzmann distribution

$$n_j = n_{j0} \exp\left(-\frac{z_j e \phi_1}{k_B T}\right). \quad [6]$$

If the potential is low, this expression and ρ can be approximated respectively by

$$n_j \cong n_{j0} \left(1 - \frac{z_j e \phi_1}{k_B T}\right) \quad [7]$$

and

$$\rho \cong \sum_{j=1}^2 z_j e n_{j0} \left(1 - \frac{z_j e \phi_1}{k_B T}\right). \quad [7a]$$

Substituting Eq. [7] into Eq. [3] and defining $\phi_j^* = \phi_j / (k_B T / z_1 e)$ and $n_j^* = n_j / n_{10}$, we obtain

$$\nabla^2 \phi_1^* = (\kappa a)^2 \phi_1^*, \quad [8]$$

where the reciprocal Debye length κ and the scaled Laplace operator ∇^2 are defined respectively by

$$\kappa = \left[\sum_{j=1}^2 n_{j0} (e z_j)^2 / \varepsilon k_B T \right]^{1/2} \quad [9]$$

and

$$\nabla^2 = \frac{x^2}{c^{*2}} \left[\frac{\partial^2}{\partial \eta^2} + \frac{\partial^2}{\partial \xi^2} - \frac{\sinh \eta}{x} \frac{\partial}{\partial \eta} + \frac{\cos \xi \cosh \eta - 1}{x \sin \xi} \frac{\partial}{\partial \xi} \right], \quad [10]$$

with $c^* = c/a$ and $x = \cosh \eta - \cos \xi$. Since $\phi = \phi_1 + \phi_2$, Eqs. [3] and [8] lead to

$$\nabla^2 \phi_2^* = \nabla^2 \phi^* - \nabla^2 \phi_1^* = 0. \quad [11]$$

Taking the curl of Eq. [5] and adopting the stream function representation, we obtain

$$\mu \frac{i_\varphi}{\omega} E^4 \psi - \nabla \times (\rho \nabla \phi) = 0, \quad [12]$$

where $\omega = c \sin \xi / x$ is the distance from the symmetric axis. The η - and the ξ -components of the velocity, v_η and v_ξ , can be expressed as

$$v_\eta = \frac{x^2}{c^2 \sin \xi} \frac{\partial \psi}{\partial \xi} \quad [13]$$

$$v_\xi = \frac{-x^2}{c^2 \sin \xi} \frac{\partial \psi}{\partial \eta}. \quad [14]$$

Substituting Eq. [7a] into Eq. [12] and defining the scaled stream function ψ^* as $\psi^* = \psi / (k_B T / z_1 e)^2 \varepsilon a / \mu$, we have

$$E^4 \psi^* = -(\kappa a)^2 \frac{x \sin \xi}{c^*} \left(\frac{\partial \phi_1^*}{\partial \xi} \frac{\partial \phi_2^*}{\partial \eta} - \frac{\partial \phi_1^*}{\partial \eta} \frac{\partial \phi_2^*}{\partial \xi} \right), \quad [15]$$

where $E^4 = E^2 E^2$, and E^2 is defined by

$$E^2 = \frac{x^2}{c^{*2}} \left[\frac{\partial^2}{\partial \eta^2} + \frac{\partial^2}{\partial \xi^2} + \frac{\sinh \eta}{x} \frac{\partial}{\partial \eta} + \frac{1 - \cos \xi \cosh \eta}{x \sin \xi} \frac{\partial}{\partial \xi} \right]. \quad [16]$$

The boundary conditions associated with Eqs. [8], [11], and [15] are assumed as follows. For the equilibrium potential ϕ_1 we assume that it vanishes at a point far away from the sphere; that is,

$$\phi_1^* = 0, \quad \eta = 0, \quad \xi = 0, \quad \xi = \pi, \quad \text{and} \quad \eta = 0. \quad [17]$$

The symmetric nature of the present system requires that

$$\frac{\partial \phi_1^*}{\partial \xi} = 0, \quad \xi = 0, \quad \text{and} \quad \xi = \pi. \quad [18]$$

Suppose that the dissociation of the functional groups on the particle surface can be expressed by (27)

Let K_a be the dissociation constant of this expression. Then

$$K_a = \frac{[A^-][H^+]_S}{[AH]}. \quad [20]$$

The subscript S denotes surface property. Suppose that the

spatial variation of $[H^+]$ follows Boltzmann distribution

$$[H^+]_S = [H^+]_b \exp\left(\frac{-e\phi_1}{k_B T}\right). \quad [21]$$

The subscript b denotes bulk liquid property. If we let N_S be the concentration of dissociable functional groups on the particle surface, then

$$N_S = [A^-] + [AH]. \quad [22]$$

Equations [20]–[22] yield

$$[A^-] = \frac{N_S}{1 + \frac{[H^+]_b}{K_a} \exp\left(-\frac{e\phi_1}{k_B T}\right)}. \quad [23]$$

The surface charge concentration σ can be expressed as

$$\sigma = -e[A^-]. \quad [24]$$

Equations [23] and [24] lead to

$$\sigma = -\frac{eN_S}{1 + \frac{[H^+]_b}{K_a} \exp\left(-\frac{e\phi_1}{k_B T}\right)}. \quad [25]$$

If ϕ_1 is low, this expression can be approximated by

$$\begin{aligned} \sigma &= \sigma|_{\phi_1=0} + \left. \frac{d\sigma}{d\phi_1} \right|_{\phi_1=0} (\phi_1 - 0) \\ &= \frac{-eN_S}{\{1 + [H^+]_b/K_a\}} - \frac{(e^2 N_S / k_B T) \{[H^+]_b / K_a\}}{\{1 + [H^+]_b / K_a\}^2} \phi_1. \end{aligned} \quad [26]$$

We assume that the dielectric constant of the solid phase is much smaller than that of the liquid phase. Then

$$\sigma = -\varepsilon \nabla \phi \cdot \mathbf{n}. \quad [27]$$

In bispherical coordinates, this expression becomes

$$\sigma = \varepsilon \frac{x}{c} \frac{\partial \phi_1}{\partial \eta}. \quad [28]$$

Equations [26] and [28] yield

$$\frac{x}{c^*} \frac{1 + [H^+]_b / K_a}{(e^2 N_S a / \varepsilon k_B T)} \frac{d\phi_1^*}{d\eta^*} + \frac{[H^+]_b / K_a}{1 + [H^+]_b / K_a} \phi_1^* = -1. \quad [29]$$

If we let $A = (e^2 N_S a / \varepsilon k_B T)$ and $B = [H^+]_b / K_a$, then this expression can be rewritten as

$$\frac{x}{c^*} \frac{1 + B}{A} \frac{d\phi_1^*}{d\eta^*} + \frac{B}{1 + B} \phi_1^* = -1. \quad [30]$$

This condition needs to be satisfied on the particle surface. Note that if the surface potential of the particle remains constant, then Eq. [30] becomes

$$\phi_1^* = 1, \quad \eta = \eta_0. \quad [31]$$

For the perturbed potential ϕ_2 , since the electric field $\mathbf{E}_z$ is in the Z direction, we have

$$\frac{\partial \phi_2^*}{\partial \eta} = 0, \quad \eta = \eta_0 \quad [32]$$

$$\frac{\partial \phi_2^*}{\partial \xi} = 0, \quad \xi = 0, \quad \text{and} \quad \xi = \pi \quad [33]$$

$$\phi_2^* = -\mathbf{E}_z^* \cdot \mathbf{r}^*, \quad \eta = 0, \quad \eta = \eta_0, \quad \xi = 0, \quad \text{and} \quad \eta = 0, \quad [34]$$

where $\mathbf{E}_z^* (= \mathbf{E}_z / (k_B T / (z_1 e / a)))$ is the scaled electric field, $\mathbf{r}^*$ is the position vector from the center of the particle, $\mathbf{r}^* = r^* \cdot \mathbf{i}_r$, $r^* = r/a$, r being the distance from the center of the particle. In bispherical coordinates, r^* can be expressed as

$$r^* = c^* \left(\frac{\cosh \eta + \cos \xi}{x} \right)^{1/2}. \quad [35]$$

At steady state the particle moves with velocity $\mathbf{U}$ and the fluid at infinity is stationary; therefore the boundary conditions for the flow field can be assumed as

$$\psi^* = \frac{-1}{2} \omega^{*2} U^* \quad \text{and} \quad \frac{\partial \psi^*}{\partial \eta} = \frac{c^{*2}}{x^3} \sin \xi \sinh \eta U^*, \quad \eta = \eta_0 \quad [36]$$

$$\psi^* = 0 \quad \text{and} \quad \frac{\partial \psi^*}{\partial \xi} = 0, \quad \xi = 0, \quad \text{and} \quad \xi = \pi \quad [37]$$

$$\psi^* = 0 \quad \text{and} \quad \frac{\partial \psi^*}{\partial \eta} = 0, \quad \eta = 0 \quad [38]$$

$$\frac{\psi^*}{r^{*2}} \rightarrow 0, \quad \eta = 0, \quad \xi = 0. \quad [39]$$

In these expressions, $\omega^* = c^* \sin \xi / x$ and $U^* = U / (kT / (ez_1)^2 \varepsilon / \mu a)$.

The forces acting on the particle include the electric force F_{Ez} and the hydrodynamic force F_{Dz} , which can be evaluated by

$$F_{Ez} = 2\pi \int_0^\pi \sigma (-\nabla \phi) \delta S \quad [40]$$

$$F_{Dz} = \mu \pi \int_0^\pi \omega^3 \frac{\partial}{\partial n} \frac{E^2 \psi}{\omega^2} \delta S - \pi \int_0^\pi \omega^2 \rho \frac{\partial \phi}{\partial S} \delta S, \quad [41]$$

where s denotes a tangential surface. Substituting Eq. [28] into

Eq. [40] yields

$$F_{Ez}^* = 2\pi \int_0^\pi -\frac{\sin \xi}{x} \frac{\partial \phi}{\partial \eta} \times \left((1 - \cosh \eta \cos \xi) \frac{\partial \phi^*}{\partial \eta} - \sinh \eta \sin \xi \frac{\partial \phi^*}{\partial \xi} \right) d\xi. \quad [42]$$

Substituting Eq. [7a] into Eq. [41] gives

$$F_{Dz}^* = \pi \int_0^\pi -\omega^{*3} \frac{\partial}{\partial \eta} \frac{E^2 \psi^*}{\omega^{*2}} d\xi + (\kappa a)^2 \pi \times \int_0^\pi \frac{c^{*2}}{x^2} \sin^2 \xi \phi_1^* \frac{\partial \phi^*}{\partial \xi} d\xi. \quad [43]$$

Following the same treatment as that of O'Brien and White (6), the problem under consideration is decomposed into two sub-problems. In the first the particle moves with constant velocity in the absence of the applied electric field, and in the second the particle is held constant when the external electric field is applied. If we let f_1 and f_2 be the net forces acting on the particle in these two problems, then $f_1 = \chi U^*$ and $f_2 = \beta E^*$, where χ and β are constant. Since the net force acting on the particle vanishes at the steady state, we have $f_1 + f_2 = 0$, which implies that the mobility of the particle can be expressed as $U^* / E^* = -\beta / \chi$.

RESULTS AND DISCUSSION

Figure 2 illustrates the variation of the scaled electrophoretic mobility (U^* / E^*) as a function of η_0 at various κa , and the variation of (U^* / E^*) as a function of κa at various η_0 is

FIG. 2. Variation of scaled electrophoretic mobility (U^* / E^*) as a function of η_0 at various, double layer thicknesses κa for the case when the surface potential of particle remains constant at unity.

FIG. 3. Variation of scaled electrophoretic mobility (U^*/E^*) as a function of κa at various distances between the particle and plane η_0 for the case when the surface potential of the particle remains constant.

presented in Fig. 3. Since $\eta_0 = \cosh^{-1}(h/a)$ and h being respectively the radius of a particle and the distance between the center of the particle and the surface, η_0 is as a measure for the relative distance between the particle and the surface. Figure 2 shows that for a fixed κa the scaled mobility increases with the increase in η_0 ; that is, the presence of the surface has the effect of retarding the movement of the particle. Therefore, the larger the η_0 , the greater the distance between the particle and the surface, and the less the influence of the latter on the movement of the former. This is consistent with previous results (21, 22). Apparently, if $\eta_0 \rightarrow \infty$ the mobility of the particle approaches that of an isolated particle. Figure 3 reveals that for a fixed η_0 , (U^*/E^*) increases with κa . This is because the smaller the κa the thicker the double layer surrounding the particle, and the more easily it feels the presence of the surface. Note that if κa is smaller than a certain critical value, (U^*/E^*) becomes independent of κa . This critical value is on the order of the surface-to-surface distance between the particle and the surface.

The variation of the scaled electrophoretic mobility (U^*/E^*) as a function of κa at various A is illustrated in Fig. 4a, and the corresponding variation in the surface potential is presented in Fig. 4b. According to its definition ($=e^2 N_S a / \epsilon k_B T$), A is a measure of the concentration of the dissociable functional groups on the particle surface; the larger the value of A the greater the number of dissociable functional groups. Figure 4a reveals that if A is small, the absolute mobility $|U^*/E^*|$ decreases with the increase in κa . However, if A is large, $|U^*/E^*|$ increases with the increase in κa . For a Medium A , $|U^*/E^*|$ exhibits a local maximum as κa varies. These trends reveal that the mobility is controlled by two competing factors. The first factor is of hydrodynamic nature. It is known that for both an isolated particle and a concentrated dispersion, the thinner the double layer the larger the mobility (6, 17). That is, $|U^*/E^*|$ increases with κa . This is pronounced if the double layer does

not touch the planar surface. As $A \rightarrow \infty$, the particle has a constant potential surface, and therefore the qualitative behavior of $|U^*/E^*|$ as κa varies for the case A is large ($=1000$) similar to that of Fig. 3. Note that a particle is positively charged in the latter and is negatively charged in the former. The second controlling factor is the variation of the surface potential of a particle. As can be seen from Fig. 4b, the absolute surface potential decreases with the increase in κa . This is because the thinner the double layer the smaller the space available for the distribution of dissociated ionic species, which leads to a smaller degree of dissociation of the functional groups on the particle surface. This yields a lower surface charge density and a smaller mobility. This effect is pronounced if A is small. For a medium value of A ($=100$), the competition of the two controlling factors yields a local maximum in $|U^*/E^*|$ as κa varies.

The variation of the scaled electrophoretic mobility (U^*/E^*) as a function of κa at various B is illustrated in Fig. 5. This figure suggests that if κa is constant, $|U^*/E^*|$ decreases with the increase in B . The definition of B ($=[H^+]_b/K_a$) indicates that for a fixed K_a it is a measure of $[H^+]$ in the bulk liquid phase. The larger the value of B the higher the $[H^+]$, which

FIG. 4. Variation of scaled electrophoretic mobility (U^*/E^*), (a), and scaled surface potential, (b), as a function of κa at various A for the case $\eta_0 = 1.317$ and $B = 1$.

FIG. 5. Variation of scaled electrophoretic mobility (U^*/E^*) as a function of κa at various B for the case $\eta_0 = 1.317$ and $A = 1$.

leads to a lower degree of dissociation of the functional groups on particle surface. Therefore the surface charge density becomes lower, and the mobility becomes smaller. On the other hand, if $[H^+]$ is low, the surface charge density is high, and the absolute mobility becomes large, as illustrated in Fig. 5. This figure also shows that if B is very large ($B = 100$), the dissociation of the functional groups on the particle surface is negligible, and the mobility vanishes essentially. On the other hand, if B is small, $|U^*/E^*|$ is large. If $B = 0$, the dissociation of surface functional groups is complete, and the present model reduces to a constant surface charge model. However, as mentioned previously, since the mobility is still influenced by the thickness of a double layer,

FIG. 6. Variation of scaled electrophoretic mobility (U^*/E^*) as a function of η_0 at various A for the case $\kappa a = 10$ and $B = 1$.

$|U^*/E^*|$ starts to decrease with the increase in κa if the latter is sufficiently large.

Figure 6 summarizes the variation of the scaled electrophoretic mobility (U^*/E^*) as a function of η_0 at various A . For a fixed A , the qualitative behavior of the variation of (U^*/E^*) as a function of η_0 is similar to that shown in Fig. 2. For a fixed η_0 , the qualitative behavior of the variation of (U^*/E^*) as a function of A is similar to that illustrated in Fig. 4.

CONCLUSION

Bispherical coordinates coupled with an orthogonal collocation procedure are adopted to solve the problem of the electrophoresis of a charge-regulated sphere normal to a plane. The method used is applicable to an arbitrary, double layer thickness. The charged conditions on particle surface are found to have a profound influence on its qualitative behavior. For instance, for a medium level of concentration of the dissociable functional groups on the surface of a particle, its electrophoretic mobility may have a local maximum as the thickness of the double layer varies.

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