

Electrophoresis of a Nonrigid Entity in a Spherical Cavity

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The electrophoretic behavior of a nonrigid entity, e.g., liquid entity or bubble, in a spherical cavity is analyzed theoretically taking the effect of double-layer polarization into account. We show that the electrophoretic mobility of the entity decreases as the ratio (viscosity of surrounding fluid/viscosity of medium inside entity) decreases and its surface potential decreases. If the surface potential is sufficiently high, double layer polarization has the effect of producing a local minimum in the electrophoretic mobility as double layer thickness varies. If the double layer is thick, depending upon the level of surface potential, the limiting electrophoretic mobility when the viscosity ratio is much larger than unity (e.g., gas bubble in liquid) is about 5–10 times of that when the viscosity ratio is much smaller than unity (e.g., rigid particle in liquid). For a thin or medium-thick double layer, the limiting electrophoretic mobility when the viscosity ratio is much larger than unity is about 3 times of that when the viscosity ratio is much smaller than unity. The presence of cavity wall has the effect of retarding the electrophoretic mobility.

1. Introduction

The application of an external electrical field on a dispersion of colloid particles in an electrolyte solution has the influence on both the distribution of ions in the double layer surrounding a particle and its kinetic behavior, the so-called electrokinetic phenomena.^{1–4} The movement of a charged particle relative to the surrounding fluid as a response to the applied electric field, for example, is termed electrophoresis, and the terminal velocity of the particle per unit strength of the electric field is defined as its electrophoretic mobility. In general, the equations describing electrophoresis are coupled, nonlinear partial differential equations, and solving them analytically is almost impossible. To recover an explicit expression for electrophoretic mobility, drastic assumptions are often made in practice. These include, for example, simple geometry, low surface potential, limiting thickness of double layer, isolated entity, and negligible boundary effect. The original derivation for electrophoresis was conducted by Smoluchoski,^{5,6} in which it was shown that under the conditions of infinitely thin double layer, low surface potential, and weak applied electric field the electrophoretic mobility of a rigid entity is linearly dependent on its zeta potential. The analysis of Smoluchoski was extended by Henry⁷ to a finite double layer thickness. Relevant studies about electrophoresis are ample in the literature, and the original derivations of Smoluchoski were extended to the cases of, for example, double-layer relaxation which is significant, arbitrary level of electrical potential, strong applied electric field, presence of a boundary, and concentrated dispersions.^{8–12}

The electrophoresis of nonrigid entities, such as liquid drops and bubbles, is also of practical significance. The problem is more complicated than that of a rigid entity for several reasons. First, because the flow field inside an entity might influence that outside it, the former needs to be considered, in general. Second, if the fluid phase inside the entity contains electrolyte, the electric field inside it might also be important and needs to

be considered. Third, the deformation of the entity surface may be significant. Under the conditions of negligible double layer relaxation, weak applied electric field, and low surface potential, Booth¹³ was able to derive the relation between the electrophoretic velocity of an entity as a function of its viscosity and double layer thickness. Levich¹⁴ also analyzed the same problem through a different approach, and a result different from that of Booth was obtained. Levine and O'Brien¹⁵ concluded that the difference between the two is appreciable for the case of a mercury entity; the latter is found to be more accurate than the former, which does not consider the phenomenon of surface dipole. Levine¹⁶ examined the electrophoresis of a drop taking this phenomenon into account. Ohshima et al.¹⁷ found that if the surface potential of a mercury drop is sufficiently high its electrophoretic behavior is the same as that of the corresponding rigid particle, the so-called solidification phenomenon of a liquid entity. Baygents and Saville¹⁸ investigated the electrophoretic behavior of an isolated liquid or gas entity for the case of weak applied electric field and arbitrary surface potential; both the nonconducting and conducting entity were considered. It was concluded that for a nonconducting entity its electrophoretic velocity increases with the increase in surface potential and has a local maximum, but it decreases with the increase in double layer thickness and has a local minimum. The solidification phenomenon occurs at a high surface potential. Compared with that of a nonconducting entity, the electrophoretic behavior of a conducting entity is more complicated. For example, the numerical result reveals that if the double layers inside and outside an entity is very thin, it may move to a direction which is inconsistent with the sign of its surface charge. This was attributed to the complicated flow behavior of a buffer zone, which appeared on the entity surface.

In the present study, the boundary effect on the electrophoretic behavior of a nonrigid entity is studied. In particular, that of a nonconducting entity in a spherical cavity is analyzed taking the effects of surface potential, double layer thickness, double-layer polarization, and relative magnitude of the viscosity of the entity and that of the surrounding fluid into account.

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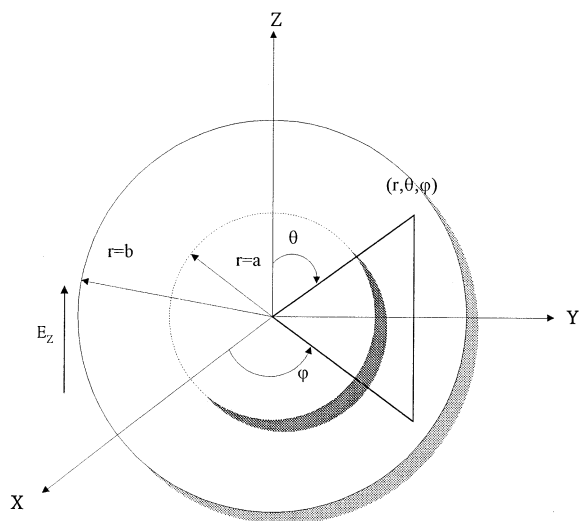


Figure 1. Schematic representation of the problem under consideration. An entity of radius a is placed at the center of a spherical cavity of radius b . An electric field \mathbf{E}_z , which has magnitude E_z and in the Z direction is applied, and \mathbf{U} is the terminal velocity of the particle. The origin of the spherical coordinates (r, θ, φ) is placed at the center of the cavity.

2. Theory

Referring to Figure 1, we consider the case where a nonconductive entity of radius a is located at the center of a spherical cavity of radius b , which is filled with a certain fluid. An electric field \mathbf{E}_z , with magnitude E_z is applied in the Z direction. The spherical coordinates (r, θ, φ) with their origin located at the center of the cavity are adopted. Suppose that the physical properties of the entity and those of the fluid assume their macroscopic values, that the shape of the entity remains spherical, and that the effect of the surface tension of the entity is negligible. For simplicity, we consider the case where the interior of the entity is electrolytes free. We assume that the electric potential can be described by the Poisson equation

$$\nabla^2 \phi = -\frac{\rho_E}{\epsilon} \quad (1)$$

where $\rho_E = \sum n_i z_i e = \sum n_{i0} z_i e \exp(-z_i e \phi / k_B T)$ is the space charge density in the fluid phase, n_i and n_{i0} are respectively the concentration of ionic species i and its bulk concentration, z_i is the valence of ionic species i , k_B and T are respectively the Boltzmann constant and the absolute temperature, e is the elementary charge, ϕ is the electrical potential, $\Delta \phi$ is the difference between the bulk electric potential inside the entity and that of the fluid,¹⁹ ∇ is the gradient operator, and ϵ and ρ_E are respectively the dielectric constant and the space charge density of mobile ions.

At steady state, the conservation of ionic species j implies that

$$\nabla \cdot \mathbf{f}_j = 0 \quad (2)$$

The flux of ionic species j , \mathbf{f}_j , can be expressed by

$$\mathbf{f}_j = -D_j \left[\nabla n_j + \frac{z_j e n_j}{k_B T} \nabla \phi \right] + n_j \mathbf{v} \quad (3)$$

where D_j is the diffusion coefficient of ionic species j , and \mathbf{v} is the velocity of the fluid. Equations 2 and 3 yield

$$\nabla^2 n_j + \frac{z_j e}{k_B T} (\nabla n_j \cdot \nabla \phi + n_j \nabla^2 \phi) - \frac{1}{D_j} \mathbf{v} \cdot \nabla n_j = 0 \quad (4)$$

For the case of an incompressible Newtonian fluid with constant physical properties, the flow field in the creeping flow regime can be described by

$$\mu \nabla^2 \mathbf{v} - \nabla p - \rho_E \nabla \phi = 0 \quad (5)$$

$$\nabla \cdot \mathbf{v} = 0 \quad (6)$$

where p is the pressure and μ is the viscosity of the fluid. These expressions are also applicable to the flow field inside the entity, except that the physical properties of the entity should be used.

Suppose that the applied electric field is weak so that the electric potential ϕ can be approximated by the sum of the potential in the absence of the applied electric field (or the equilibrium potential), ϕ_1 , and a perturbed potential due to the applied electric field, ϕ_2 , with $|\phi_2| \ll |\phi_1|$. If the applied electric field is weak, the problem under consideration is of a linear nature, and it can be decomposed into two virtual problems.¹¹ In the first one, an entity moves in the absence of the applied electric field, and in the second one, the entity is held fixed when the electric field is applied.

Instead of solving directly the governing equations for the flow field, they are transformed first into a stream function representation. If we let ψ be the stream function, then the r and the θ component of \mathbf{v} can be expressed respectively as $v_r = -(1/r^2 \sin \theta) (\partial \psi / \partial \theta)$ and $v_\theta = (1/r \sin \theta) (\partial \psi / \partial r)$. Taking curl on both sides of eq 6 yields

$$E^4 \psi = \frac{1}{\mu} \left[\frac{\partial \rho_E}{\partial r} \frac{\partial \phi}{\partial \theta} - \frac{\partial \rho_E}{\partial \theta} \frac{\partial \phi}{\partial r} \right] \sin \theta \quad (7)$$

where $E^4 = E^2 E^2$ with

$$E^2 = \frac{\partial^2}{\partial r^2} + \frac{\sin \theta}{r^2} \frac{\partial}{\partial \theta} \left(\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \right) \quad (8)$$

Suppose that the equilibrium potential ϕ_1 can be described by the Poisson–Boltzmann equation

$$\nabla^2 \phi_1 = -\frac{\rho_E^0}{\epsilon} \quad (9)$$

where $\rho_E^0 = \sum n_i^0 z_i e = \sum n_{i0} z_i e \exp(-z_i e \phi_1 / k_B T)$. Because $\phi = \phi_1 + \phi_2$, the perturbed potential ϕ_2 can be described by

$$\nabla^2 \phi_2 = \nabla^2 \phi - \nabla^2 \phi_1 = -\left(\frac{\rho_E}{\epsilon} - \frac{\rho_E^0}{\epsilon} \right) \quad (10)$$

The distribution of the space charge density of mobile ions are influenced by the applied electric field. This can be taken into account by assuming¹¹

$$\rho_E = \sum_i n_i z_i e = \sum_i z_i e n_{i0} \exp \left(-\frac{z_i e (\phi_1 + \phi_2 + g_i)}{k_B T} \right) \quad (11)$$

where g_i is a perturbed potential for ionic species i . Because the applied electric field is weak, eq 11 can be approximated by

$$\rho_E \approx \sum_i z_i e n_{i0} \exp \left(-\frac{z_i e \phi_1}{k_B T} \right) \left[1 + \frac{z_i e}{k_B T} (\phi_2 + g_i) \right] \quad (12)$$

Substituting this expression into eq 4 yields

$$\nabla^2 g_i - \frac{z_i e}{k_B T} \nabla \phi_1 \cdot \nabla g_i = \frac{1}{D_i} \mathbf{v} \cdot \nabla \phi_1 \quad (13)$$

The entity is nonconductive, and its interior is electrolyte free; therefore, the boundary conditions at the entity–fluid interface can be expressed as

$$\frac{\partial \phi_1}{\partial r} = 0, \quad r = a \quad (14)$$

$$\frac{\partial g_j}{\partial r} = 0, \quad r = a \quad (15)$$

The last expression arises from the fact that \mathbf{f}_j vanishes at the entity–fluid interface. For convenience, we assume that the fluid is fixed, and the cavity moves with velocity $-\mathbf{U}$. Suppose that the cavity surface is remained at constant potential ζ_b , that the perturbed potential vanishes at cavity surface, and that the cavity surface is nonslip. These lead to the following boundary conditions on cavity surface:

$$\phi_1 = \zeta_b, \quad r = b \quad (16)$$

$$\frac{\partial \phi_2}{\partial r} = -E_Z \cos \theta, \quad r = b \quad (17)$$

$$g_j = -\phi_2, \quad r = b \quad (18)$$

$$\psi = -\frac{1}{2} U r^2 \sin \theta \quad \text{and} \quad \frac{\partial \psi}{\partial r} = -U r \sin^2 \theta, \quad r = b \quad (19)$$

Equation 17 describes the magnitude of the electric field at cavity surface, and the last expression arises from the fact that $v_r = U \cos \theta$ and $v_\theta = -U \sin \theta$. The symmetric nature of the problem requires that

$$\frac{\partial \phi_1}{\partial r} = \frac{\partial \phi_2}{\partial r} = \frac{\partial g_1}{\partial r} = \frac{\partial g_2}{\partial r} = \frac{\partial \psi}{\partial r} = 0, \quad r = 0 \quad (20)$$

Equations 7, 9, 10, and 13 can be rewritten in scaled forms as

$$\nabla^{*2} \phi_1^* = -\frac{1}{(1+\alpha)} \frac{(\kappa a)^2}{\phi_r} [\exp(-\phi_r \phi_1^*) - \exp(\alpha \phi_r \phi_1^*)] \quad (21)$$

$$\begin{aligned} \nabla^{*2} \phi_2^* - \frac{(\kappa a)^2}{(1+\alpha)} (\exp(-\phi_r \phi_1^*) + \alpha \exp(\alpha \phi_r \phi_1^*)) \phi_2^* = \\ \frac{(\kappa a)^2}{(1+\alpha)} (\exp(-\phi_r \phi_1^*) g_1^* + \alpha \exp(\alpha \phi_r \phi_1^*) g_2^*) \end{aligned} \quad (22)$$

$$E^{*4} \psi^* = \frac{(\kappa a)^2}{(1+\alpha)} \left[\left(\frac{\partial g_1^*}{\partial \theta} n_1^* + \frac{\partial g_2^*}{\partial \theta} \alpha n_2^* \right) \frac{\partial \phi_1^*}{\partial r^*} \right] \sin \theta \quad (23)$$

$$\nabla^{*2} g_1^* - \phi_r \nabla^* \phi_1^* \cdot \nabla^* g_1^* = \phi_r^2 P e_1 \mathbf{v}^* \cdot \nabla^* \phi_1^* \quad (24)$$

$$\nabla^{*2} g_2^* + \alpha \phi_r \nabla^* \phi_1^* \cdot \nabla^* g_2^* = \phi_r^2 P e_2 \mathbf{v}^* \cdot \nabla^* \phi_1^* \quad (25)$$

In these expressions, $g_j^* = g_j / \zeta_a$, $\phi_1^* = \phi_1 / \zeta_a$, $\phi_2^* = \phi_2 / \zeta_a$, $\psi^* = \psi / (U_s a^2)$, $\phi_r = \zeta_a z_1 e / k_B T$, and $\alpha = z_2 / z_1$, where ϕ_r is the scaled surface potential, $P e_j$ is the Peclet number of ionic species j , and κa is the scaled entity radius, with κ being the reciprocal Debye length defined by

$$\kappa = \left[\sum_{j=1}^2 n_{j0} (e z_j)^2 / \epsilon k_B T \right]^{1/2} \quad (26)$$

n_1^* and n_2^* are the scaled ionic concentrations defined by

$$n_1^* = \exp(-\phi_r \phi_1^*) [1 - \phi_r (\phi_1^* + \phi_2^* + g_1^*)] \quad (27)$$

$$n_2^* = \exp(\alpha \phi_r \phi_1^*) [1 + \alpha \phi_r (\phi_1^* + \phi_2^* + g_2^*)] \quad (28)$$

The problem under consideration can be solved by the method of separation of variables. Let $\phi_2^* = \Phi_2(r^*) \cos \theta$, $g_1^* = G_1(r^*) \cos \theta$, $g_2^* = G_2(r^*) \cos \theta$, and $\psi^* = \Psi(r^*) \sin^2 \theta$. The symmetric nature of the problem implies that ϕ_1^* is a function of r^* only. Then the governing equations, (21–25), become

$$\begin{aligned} L^2 \Phi_2 - \frac{(\kappa a)^2}{(1+\alpha)} (\exp(-\phi_r \phi_1^*) + \alpha \exp(\alpha \phi_r \phi_1^*)) \Phi_2 = \\ \frac{(\kappa a)^2}{(1+\alpha)} (\exp(-\phi_r \phi_1^*) G_1 + \alpha \exp(\alpha \phi_r \phi_1^*) G_2) \end{aligned} \quad (29)$$

$$L^2 G_1 - \phi_r \left(\frac{\partial \phi_1^*}{\partial r} \frac{\partial G_1}{\partial r} \right) = P e_1 \left(\frac{-2\Psi}{r^2} \right) \frac{\partial \phi_1^*}{\partial r} \quad (30)$$

$$L^2 G_2 - \alpha \phi_r \left(\frac{\partial \phi_1^*}{\partial r} \frac{\partial G_2}{\partial r} \right) = P e_2 \left(\frac{-2\Psi}{r^2} \right) \frac{\partial \phi_1^*}{\partial r} \quad (31)$$

$$D^4 \Psi = -\frac{(\kappa a)^2}{(1+\alpha)} \left[(n_1^* G_1 + \alpha n_2^* G_2) \frac{\partial \phi_1^*}{\partial r^*} \right] \quad (32)$$

where the operators L and D are defined by

$$L^2 = \frac{d^2}{dr^{*2}} + \frac{2}{r^*} \frac{d}{dr^*} - \frac{2}{r^{*2}} \quad (33)$$

$$D^4 = D^2 D^2 = \left(\frac{d^2}{dr^{*2}} - \frac{2}{r^{*2}} \right)^2 \quad (34)$$

In problem 1, an entity moves with velocity \mathbf{U}^* without the presence the external electric field. In this case, the magnitude of the net force acting on it F_1 is

$$F_1 = F_{D1} + F_{E1} \propto U^* \quad (35)$$

In problem 2, an electric field \mathbf{E}_Z^* is applied, but the entity is held fixed. Therefore, the magnitude of the net force acting on the entity F_2 can be expressed as

$$F_2 = F_{D2} + F_{E2} \propto \mathbf{E}_Z^* \quad (36)$$

where F_{Di} and F_{Ei} are respectively the magnitudes of drag force and electric force in problem i . Because the net force acting on the entity should vanishes, the scaled electrophoretic mobility U_m^* ($= U_m / U_s$) can be expressed as

$$U_m^* = \frac{U^*}{E^*} = -\frac{F_2}{F_1} \quad (37)$$

3. Results and Discussion

Figures 2 and 3 show the variation of the scaled electrophoretic mobility U^* as a function of the scaled double layer thickness κa at various scaled surface potential of the entity for

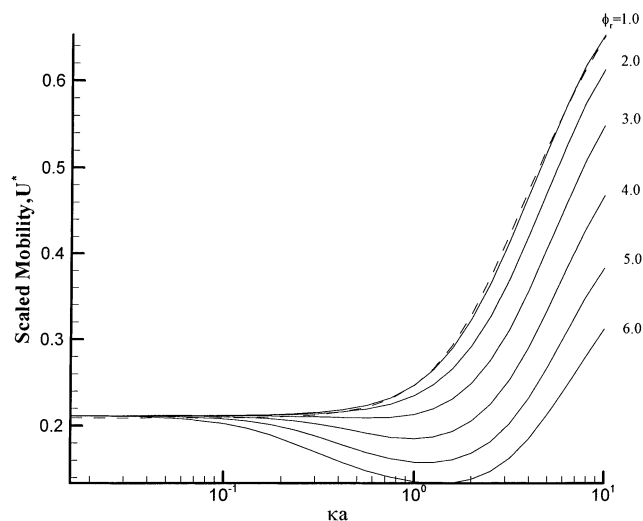


Figure 2. Variation of scaled mobility as a function of κa at various surface potential ϕ_r for the case $\lambda = 0.5$ and $\mu/\bar{\mu} = 0.01$. Solid line, present model; dashed line, result for rigid sphere.¹² Key: $Pe_1 = 0.01$, $Pe_2 = 0.01$, $\alpha = 1$.

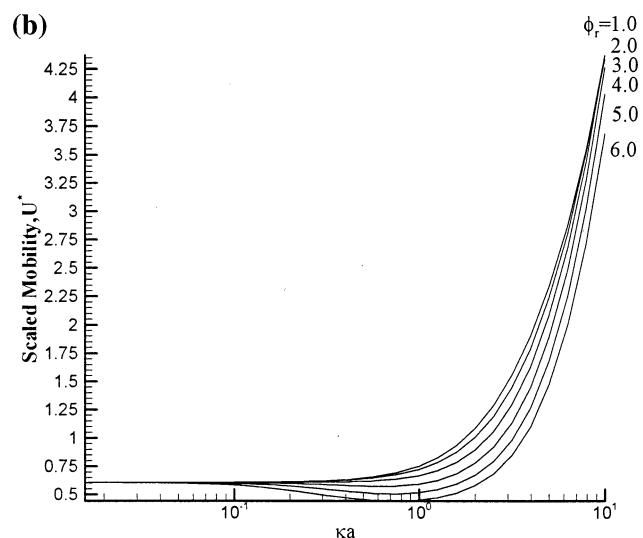
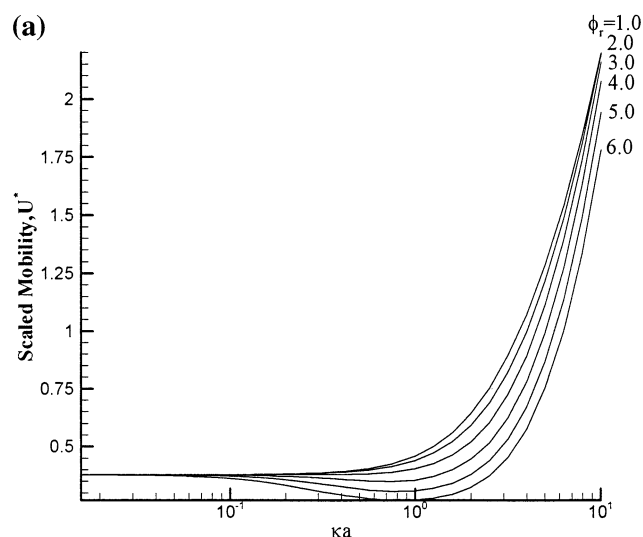


Figure 3. Variation of scaled mobility as a function of κa at various surface potential ϕ_r for the case $\lambda = 0.5$, $\mu/\bar{\mu} = 1.0$, (a); $\mu/\bar{\mu} = 100$, (b). Key: same as Figure 2.

several values of the viscosity ratio $\mu/\bar{\mu}$ (viscosity of fluid/viscosity of entity). The typical variation in the stream function

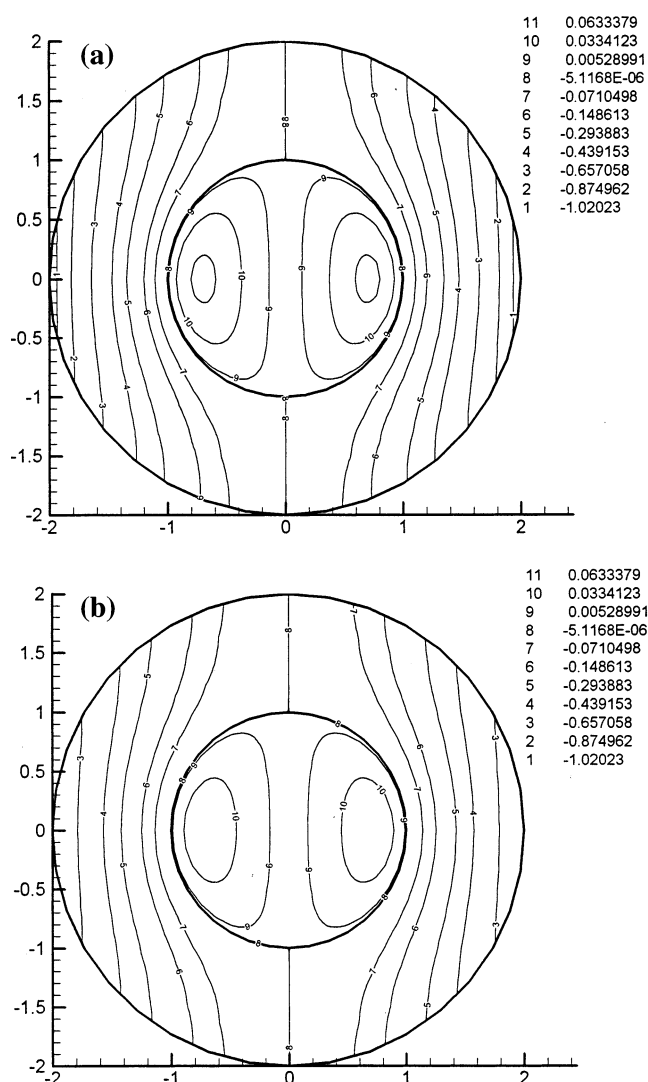


Figure 4. Variation of stream function Ψ for the case $\lambda = 0.5$, $\mu/\bar{\mu} = 2.0$, and $\kappa a = 1.0$. (a) $\phi_r = 1.0$; (b) $\phi_r = 4.0$. Key: same as Figure 2.

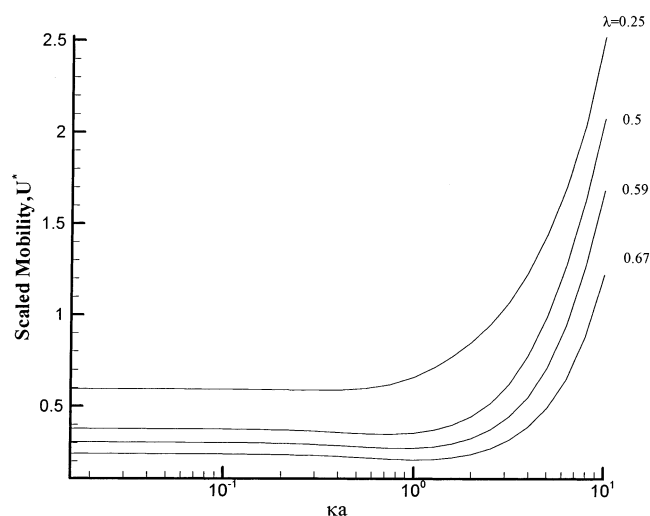


Figure 5. Variation of scaled mobility as a function of κa at various λ for the case $\phi_r = 4.0$ and $\mu/\bar{\mu} = 1.0$. Key: same as Figure 2.

Ψ for two levels of surface potential is illustrated in Figure 4. In Figure 2, $\mu/\bar{\mu} \ll 1$, which simulates a rigid entity. For comparison, the result of Zydnev,¹² which models the behavior of a rigid sphere in a spherical cavity at a low electrical potential is also presented in Figure 2. As can be seen from Figure 2,

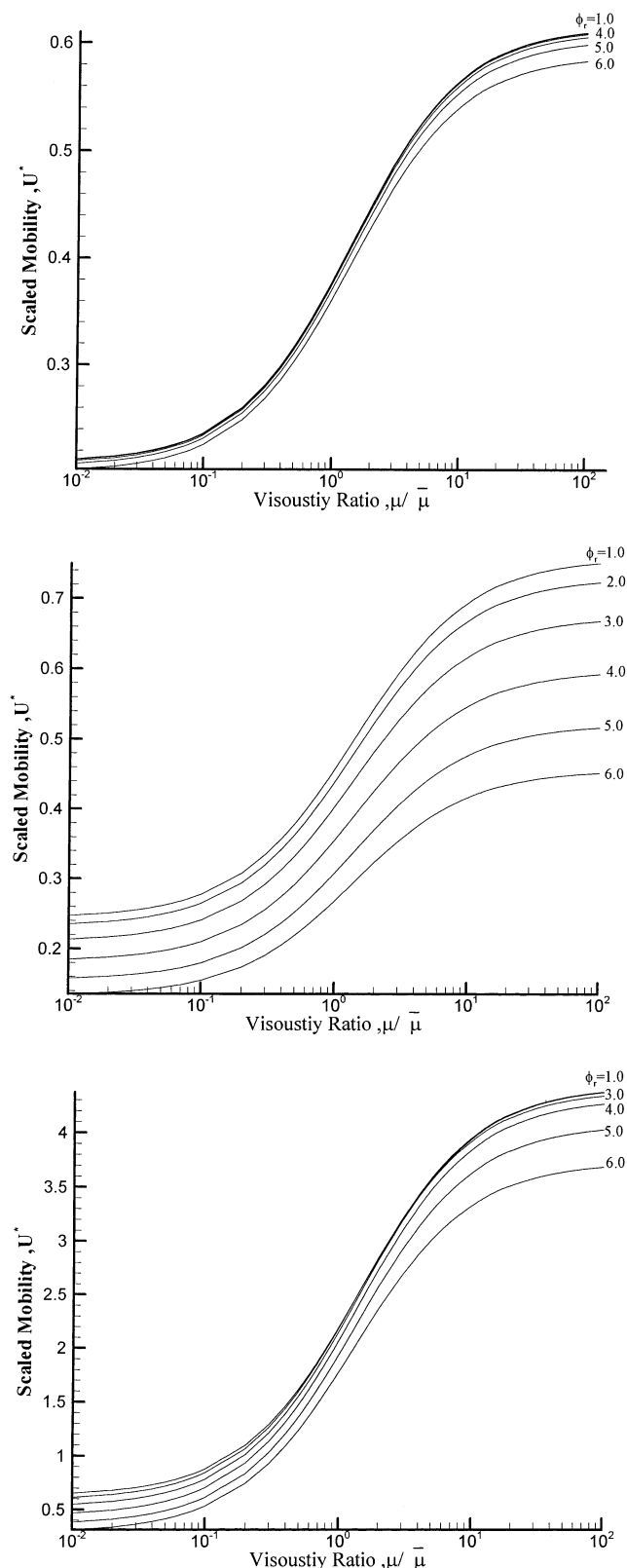


Figure 6. Variations of the scaled mobility U^* as a function of the viscosity ratio $\mu/\bar{\mu}$ at various scaled surface potential ϕ_r for three levels of double layer thickness κa . (a) $\kappa a = 0.1$, (b) $\kappa a = 1.0$, (c) $\kappa a = 10.0$. Key: same as Figure 2.

Zydney's result can be recovered as a special case of the present study by assuming a small ϕ_r . Figure 2 reveals that if κa is small, that is, the double layer is thick, U^* is insensitive to the variation in ϕ_r . This is because, if the double layer reaches the wall of the cavity, the hydrodynamic effect dominates, and the

effect of surface potential and double layer polarization on the electrophoretic mobility becomes unimportant. As κa gets large, the effect of double layer polarization becomes important, and U^* decreases with the increase in ϕ_r . In this case, neglecting the effect of double layer polarization will overestimate the electrophoretic mobility. Note that, if ϕ_r is sufficiently high, U^* has a local minimum as κa varies. This is because, if κa is large, the resistance due to hydrodynamic effect becomes insignificant, and the gradient of electrical potential is large, and therefore, the electrophoretic mobility is large. However, if surface potential is high, the effect of double layer polarization is important. Because this effect induces an internal electric field, which is in the inverse direction as that of the applied electric field, the electrophoretic mobility becomes small. The value of $\mu/\bar{\mu}$ is unity in Figure 3a and is much larger than unity in Figure 3b; the former and the latter simulate respectively a liquid drop and a gas bubble in a liquid medium. The qualitative behavior of the results shown in Figure 3 is similar to that presented in Figure 2. Figures 2 and 3 suggest that the smaller the viscosity of an entity the larger its electrophoretic mobility. This is because the smaller the viscosity of an entity the smaller the drag force it experienced. For the case $\phi_r = 1.0$ and a medium κa , $U^*(\mu/\bar{\mu} \gg 1)$ can be more than three times larger as $U^*(\mu/\bar{\mu} \ll 1)$. Figure 4 indicates that the flow of the medium inside and that outside of an entity are in the inverse direction, as expected.

Figure 5 shows the variation of the scaled mobility of an entity U^* as a function of κa at various radius ratio $\lambda = a/b$. This figure reveals that, for a fixed κa , the larger the λ the smaller the mobility of the entity. This is because the larger the λ the more significant the hydrodynamic effect of cavity wall on the movement of the entity. As can be seen from Figure 5, if λ is small, U^* increases monotonically with κa , but if λ becomes large, U^* has a local minimum as κa varies. This can be explained by the result of Zydney,¹¹ in which it was concluded that, if κa is small, the electrophoretic mobility is proportional to λ and is proportional to λ^3 if it is large. That is, the presence of a boundary will influence the qualitative behavior of the electrophoretic mobility.

The variations of the scaled mobility U^* as a function of the viscosity ratio $\mu/\bar{\mu}$ at various scaled surface potential ϕ_r for three levels of double layer thickness κa are presented in Figure 6. This figure reveals that if the double layer is thick, depending upon the level of surface potential, $U^*(\mu/\bar{\mu} \gg 1)$ is about 5–10 times of $U^*(\mu/\bar{\mu} \ll 1)$. For a thin or medium thick double layer $U^*(\mu/\bar{\mu} \gg 1) \approx 3U^*(\mu/\bar{\mu} \ll 1)$.

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