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有限長度之圓柱形粒子在圓柱管中之電泳(2/3)

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計畫執行概要

本計畫目前進展順利，已完成主要的理論推導、主控方程式的求解、及相關之數值模擬與分析。後附資料為目前已獲得之部份結果。

摘要

本文中吾人探討了兩相同圓柱粒子在一不帶電的圓柱管中，在低電位與弱外加電場的條件下延其軸心之電泳。這種幾何形狀的安排使吾人可以同時探討邊界效應與粒子間作用對電泳的影響。數值模擬的結果顯示，當粒子周圍電雙層的厚度與粒子間距離接近時，粒子間的作用對電泳的影響將相當可觀。不過，這個效應又不及邊界效應來的重要。吾人証實，粒子表面帶電不均勻時的邊界效應較粒子表面帶電均勻時者重要。另外，兩圓柱粒子間之作用對其電泳的影響較兩球形粒子間者為顯著。

I. Introduction

the role that a boundary can play in electrophoresis is an interesting but nontrivial problem. Compared with the electrophoresis of an isolated entity, that when a physical boundary is present is of complicated nature in that the entity-boundary interaction needs to be considered which can lead to unintuitive result. In a study of the electrophoresis of a spherical particle in a spherical cavity, for example, Hsu et al.,¹ concluded that while the closer the particle-cavity distance the smaller the electrophoretic mobility of the sphere, the reverse can be true when that distance is shorter than a critical value. The latter was elaborated by that if both particle and cavity are maintained at constant electrical potential, charge reversal might occur on the latter. Keh and Chen² considered the electrophoresis of a sphere parallel to a plane under the conditions of thin double layer. They showed that, for moderate to large separations between particle and plane, the presence of the plane has the effect of reducing the electrophoretic mobility of the particle, but the reverse is true if a particle is sufficiently close to a plane. Similar behavior was also observed by Keh et al.³ in an analysis of the electrophoresis of an infinite cylinder parallel to a plane, where they showed that the closer the cylinder to the plane, the larger its electrophoretic mobility. These observations can be explained by that the presence of the plane affects the electrical force and the viscous force acting on a particle in opposite direction, and the observed phenomenon is the net result of two competing effects. In a study of the electrophoresis of a sphere in spherical cavity, Zydney⁴ concluded that the boundary effect for the case of thick double-layer is much more significant than that for the case of thin double layer. Also, the charge on a cavity could alter the direction of particle movement through inducing charge on its surface and through generating an electroosmotic recirculation flow. Other relevant analyses include, for instance, those

reported by Ennis and Anderson⁵ and Shugai and Carnie⁶ for the cases of a sphere parallel to a plane, normal to a plane, and along the axis of a cylindrical pore. In fact, when a boundary is present the electrophoretic behavior of an entity is influenced by several key factors such as the increase in the viscous drag, the enhancement of the local electric field on particle surface, and the effect of possible electroosmotic flow, which can be significant if the boundary is charged.

Similar to the effect arising from the presence of a boundary, the existence of nearby entities can also have an important influence on the electrophoretic behavior of an entity. The latter occurs, for instance, when electrophoresis is conducted on a concentrated dispersion where the interaction between neighboring entities needs to be considered. Several efforts have been made to interpret the electrophoretic behavior of a particle under the influence of other particles, such as two spheres in an infinite medium⁷⁻¹⁰ and multiple spheres in an infinite medium.¹¹⁻¹⁴ These studies concluded that for thin double layers, particle-particle interaction has a relatively weak effect on the electrophoretic mobility of a particle, but if double-layer polarization is important, it can be significant. Shugai et al.¹⁵ considered the electrophoresis of two spheres, which can have different charged conditions and sizes, in an infinite medium for the case of thick double layers. They concluded that the effect of particle-particle interaction on the electrophoretic mobility of a particle can be significant when the thickness of the double layer surrounding it is comparable to its radius.

The electrophoretic behavior of a particle also depends largely on the distribution of charge over its surface. It is not uncommon in practice that the charged condition on the surface of particle is of non-uniform nature. The charge on the basal plane of a kaolin particle, for example, can have a different sign from that on its facial plane. Apparently, an analysis, which is capable of taking this effect into consideration, is

desirable from practical point of view. In a study of the electrophoresis of a charged circular disk of zero thickness in an infinite medium, Sherwood and Stone¹⁶ found that if the double layer surrounding the particle is thin, the contribution of its edge charge to its electrophoretic velocity is more important than that of its surface charge.

In this study, the effects of particle-particle interaction and particle-boundary interaction on the electrophoretic behavior of a particle are examined simultaneously by considering the electrophoresis of two cylindrical particles with a moderate thick electrical double layer along the axis of an uncharged cylindrical pore under the conditions of low electrical potential and weak applied electric field. The influences of the key parameters of the system under consideration on the electrophoretic mobility of a particle are investigated through numerical simulation. These include the thickness of double layer surrounding a particle, the aspect ratio of a particle, the distance between two particles, and the ratio (particle radius/pore radius). The effect of the charged conditions on a particle is also discussed.

II. Theory

Let us consider the problem illustrated in Figure 1, where two identical, rigid, non-conducting, cylindrical particles of radius a and length $2d$ are located on the axis of a cylindrical pore of radius b . The surface-to-surface distance between particles is $2h$. A uniform electric field E with strength E in the z -direction is applied, and the particles move along the axis of the cylindrical pore with the same velocity. The cylindrical coordinates (r, θ, z) with its origin located at the center between two particles are chosen. Because the present system is symmetric about θ only the (r, z) domain needs to be considered.

The electrophoretic behavior of the particles is described by the electrokinetic equations, which include the equation for the conservation of ions, that for the electrical field, and that for the flow field. At steady state, the conservation of the ions leads to

$$\nabla \cdot \left[n_i \mathbf{u} - D_i \left(\nabla n_i + \frac{e z_i n_i}{k_B T} \nabla \Psi \right) \right] = 0 \quad (1)$$

In this expression, ∇ is the gradient operator, n_i , D_i , and z_i are respectively the number concentration, the diffusivity, and the valence of ionic species i , e is the elementary charge, Ψ is the electric potential, k_B is the Boltzmann constant, T is the absolute temperature, and u is the fluid velocity. It can be shown that on the basis of Gauss law, the electrical field can be described by the Poisson equation

$$\nabla^2 \Psi = -\frac{\rho}{\varepsilon} \quad (2)$$

where ε is the permittivity of the liquid phase, and $\rho = \sum_i z_i e n_i$ is the charge density of mobile ions. For the case of incompressible fluid with constant physical properties at a low Reynolds number, the flow field can be described by

$$\eta \nabla^2 \mathbf{u} - \nabla p = \rho \nabla \Psi \quad (3)$$

$$\nabla \cdot \mathbf{u} = 0 \quad (4)$$

where η is the fluid viscosity, and p is the pressure.

Suppose that the applied electric field is weak and the surface potential of particle is low. In this case the effect of double layer polarization is negligible, the electrophoretic motility of a particle is independent of the drag coefficients of ionic species, and the equations governing the electric field and the flow field can be decoupled. Following the treatment of Henry,¹⁷ Ψ is decomposed into the electrical potential in the absence of the applied electric field, that is, the equilibrium potential arising from the presence of particles, Ψ_1 , and that outside particles arising from the applied electric field, Ψ_2 . In the absence of the applied electric field n_i can be obtained by letting $u=0$ in eq 1 and solving the resultant expression to obtain $n_i = n_{i0} \exp(-ez_i \Psi_1 / k_B T)$, n_{i0} being the bulk number density of ionic species i . Under the conditions of low surface potential, Ψ_1 can be described approximately by the linearized Poisson-Boltzmann equation

$$\nabla^2 \Psi_1 = \kappa^2 \Psi_1 \quad (5)$$

where $\kappa = (e^2 \sum_i z_i^2 n_{i0} / \varepsilon k_B T)^{1/2}$ is the reciprocal Debye length. Similarly, the electrical potential associated with the applied electric field Ψ_2 can be described by the Laplace equation

$$\nabla^2 \Psi_2 = 0 \quad (6)$$

If both the basal and the lateral surfaces of a particle are uniformly charged with densities σ_1 and σ_2 respectively, then the boundary conditions associated with eqs 5 and 6 can be expressed as

$$\mathbf{n} \cdot \nabla \Psi_1 = \frac{-\sigma_1}{\varepsilon} \quad \text{and} \quad \frac{\partial \Psi_2}{\partial z} = 0, \quad |z| = h, \quad |z| = h + 2d, \quad 0 \leq r \leq a \quad (7)$$

$$\mathbf{n} \cdot \nabla \Psi_1 = \frac{-\sigma_2}{\varepsilon} \quad \text{and} \quad \frac{\partial \Psi_2}{\partial r} = 0, \quad r = a, \quad h \leq |z| \leq h + 2d \quad (8)$$

$$\mathbf{n} \cdot \nabla \Psi_1 = 0 \quad \text{and} \quad \frac{\partial \Psi_2}{\partial r} = 0, \quad r = b \quad (9)$$

$$\Psi_1 = 0 \quad \text{and} \quad \nabla \Psi_2 = -\mathbf{E}, \quad |z| \rightarrow \infty, \quad r < b \quad (10)$$

In these expressions $\mathbf{n}$ is the unit normal vector directed into the liquid phase.

Suppose that both the surface of particle and that of pore are no-slip. Then if U is the magnitude of particle velocity in the z -direction, the boundary conditions associated with eqs 3 and 4 can be expressed as

$$\mathbf{u} = U\mathbf{i}_z \quad \text{on particle surface} \quad (11)$$

$$\mathbf{u} = 0, \quad r = b \quad \text{and} \quad |z| \rightarrow \infty, \quad r < b \quad (12)$$

where $\mathbf{i}_z$ is the unit vector in the z -direction.

In the present case, the forces acting on a particle include the hydrodynamic force and the electric force. The axisymmetric nature of the present problem suggests that only the forces in the z -direction need to be considered. The electric force acting on a particle in the z -direction, F_E^Z , can be calculated by

$$F_E^Z = \iint_S \sigma E_Z dS = \iint_S \sigma \left(-\frac{\partial \Psi}{\partial z} \right) dS \quad (13)$$

where S denotes particle surface. The hydrodynamic force acting on a particle in the z -direction, F_D^Z , comprises the viscous force and that arising from the hydrodynamic pressure. We have¹⁸

$$F_D^Z = \iint_S \eta \frac{\partial(\mathbf{u} \cdot \mathbf{t})}{\partial n} t_z dS + \iint_S -pn_z dS \quad (14)$$

where t and n are respectively the unit tangential and unit normal vectors on particle surface, n is the magnitude of n , and t_z and n_z are the z -component of t and that of n respectively. At steady state, the net force acting on a particle vanishes, that is,

$$F_D^Z + F_E^Z = 0 \quad (15)$$

The electrophoretic mobility of a particle can be evaluated through a trial-and-error procedure based on this expression. For a set of specified E , σ_1 , σ_2 , κa , h/a , a/d and a/b , Ψ_1 and Ψ_2 are evaluated by solving eqs 5 and 6, and the results obtained used to calculate F_E^Z by eq 13. An arbitrary U is guessed which is used

to determine the flow field through solving eqs 3 and 4, and F_D^Z is calculated by eq 14. The calculated F_E^Z and F_D^Z are substituted into eq 15 to see if it is satisfied. If it is satisfied, the previously guessed value of U is correct. Otherwise, a new U is guessed, and the whole procedure is repeated.

FlexPDE¹⁹, a differential equation solver based on a finite element method is adopted for the resolution of the governing equations and the associated boundary conditions. This software was found to be efficient and accurate for the electrokinetic phenomena of the present type.²⁰ For convenience, we define the scaled mobility μ^* as $\mu^* = U\eta/\zeta_{ref}\epsilon E$, where $\zeta_{ref} = k_B T / e$ is a reference potential. The applicability of the numerical scheme adopted is justified by Figure 2, where the variation of μ^* as a function of λ ($=a/b$) at various scaled separation distance between two particles h/a is illustrated. This figure shows that the result for the case when only one cylinder is present in pore²⁰ can be recovered as a limiting case of the present analysis as h/a is large.

III. Results and Discussions

For the present problem, if both σ_1 and σ_2 are known, the mobility is a function of κa , h/a , a/d and a/b . Figure 3 shows the variation of the scaled electrophoretic mobility μ^* as a function of the scaled separation distance between two particles, h/a , at various combinations of λ and κa for the case of uniformly charged particles. The corresponding results for the case of non-uniformly charged particles are illustrated in Figure 4. Figure 3 indicates that, if h/a is sufficiently large, μ^* approaches a constant value, which is the result for the case when only a particle is present in the pore. Similar to the result for the case of a pair of identical spheres in

an unbound fluid,¹⁵ the influence of the interaction between the double layers surrounding the particles is inappreciable if κa is large, but becomes significant if κa is small. For example, if $\lambda = 0.1$, the maximum differences of μ^* from that for the case of an isolated sphere in a pore are 12, 15, and 25 % for $\kappa a = 4, 1$, and 0.5 respectively. The dependence of μ^* on h/a is more sensitive to that for the case of two identical spheres in an unbound fluid.¹⁵ This dependence becomes insignificant, however, if λ is large, because the viscous drag arising from the wall dominates. It can be inferred that the dependence of μ^* on h/a for the case of two cylinders in an unbound fluid is much more sensitive to that for the case of two spheres because the interaction between the double layers surrounding two particles is more significant in the former. Figure 3 also indicates that the smaller the κa (thicker double layer), the smaller the μ^* , and the larger the h/a needed to reach a constant value. The former is because the thicker the double layer, the greater the hydrodynamic retardation force arising from the access ions and the boundary, and the latter is because a larger separation distance between two particles is required to avoid double layer interaction. Note that because the double layers surrounding the particles begin to overlap when κh reaches unity, double layer interaction is significant when $\kappa h \leq 1$. In general, μ^* decreases with the increase in λ , which is expected because the larger the λ , the more significant is the viscous drag arising from the presence of cylindrical pore. It is interesting to note that while μ^* decreases with the increase in h/a , in general, a local maximum may occur if κa is sufficiently large and λ is sufficiently small. This phenomenon was also observed for the case of two spheres in an unbounded fluid.¹⁵ The occurrence of the local maximum is the consequence of the competition between two opposing effects: (1) the strength of local electric field on the surface of a particle

$(\partial\Psi_2/\partial z)_{r=a}$, and therefore, the electric force acting on it, decreases with the decrease in h/a , and (2) the hydrodynamic retardation arising from the presence of the other particle decreases with the decrease in h/a , which is also observed in sedimentation. It should be pointed out that as two particles approach each other under the conditions of constant surface charge, the surface potential might violate the low surface potential assumption. For example, if $h/a=0.0625$ and $\kappa a = 1$, the scaled surface potential has a maximum value of 2.23~3.36 for $\lambda=0.1\sim0.8$. As can be seen in Figure 4, the qualitative behaviors of μ^* for non-uniformly charged particles are similar to those of uniformly charged particles. A comparison between Figures 3 and 4 reveals that if λ is small, the rate of decrease of μ^* as h/a increases for non-uniformly charged particles is faster than that for uniformly charged particles. For example, if $\lambda = 0.1$, the maximum differences of μ^* from that for the case of an isolated sphere in a pore are 15, 26, and 29% for $\kappa a = 4, 1$, and 0.5 respectively; these differences are larger than that for the case of uniformly charged particles. This implies that the interaction between two particles for the case of non-uniformly charged particles is stronger than that for the case of uniformly charged particles. If λ is large, because the boundary effect of pore dominates, the influence between particles on μ^* becomes unimportant. A comparison between Figures 3(a) and 4(a) indicates that at $\kappa a = 0.5$, if λ is small (0.1-0.2), μ^* for non-uniformly charged particles is larger than that for uniformly charged particles; but the reverse is true if λ is large (0.4-0.7). This implies that the boundary effect for the case of non-uniformly charged particles is more significant than that for the case of uniformly charged particles. This will be discussed later.

The variations of the scaled electrophoretic mobility μ^* as a function of λ for various combinations of a/d and κa are presented in Figure 5 for the case of

uniformly charged particles. This figure reveals that while μ^* decreases with the increase in λ , in general, μ^* may have a local maximum as λ varies if a/d is sufficiently large. This phenomenon was also observed in the case of the electrophoresis of a single cylinder in a cylindrical pore.²⁰ For the present case, it can be explained by the behaviors of the forces acting on a particle illustrated in Figure 6. As can be seen in Figure 6(a), for uniformly charged particles, the electric force acting on a particle increases monotonically with λ , and the hydrodynamic force acting on it may have a local minimum as λ varies. The former is mainly because that if a particle is close to pore, the squeeze of the applied electric field near particle surface leads to an increase in the electric force. The latter arises from the complicated effects of the combination of the space charged density and the strength of local electric field between particles and boundary on the hydrodynamic force as λ varies. For the case of non-uniformly charged particles, the hydrodynamic force acting on a particle does not have a local minimum, as shown in Figure 6(b), which implies that μ^* decreases monotonically with the increase in λ , which is justified in Figure 7. A comparison between Figures 5 and 7 indicates that the variation of μ^* as λ varies for the case of non-uniformly charged particles is more drastic than that for the case of uniformly charged particles. This is consistent with previous observation, that is, the boundary effect on μ^* for the case of non-uniformly charged particles is more significant than that for the case of uniformly charged particles.

Figure 8 shows the variation of the scaled electrophoretic mobility μ^* as a function of λ for the case of Figure 5. This figure suggests that the larger the a/d the smaller the μ^* . For a fixed a , a large a/d implies a small lateral surface of a particle, and therefore, a small amount of surface charge. The former leads to a small

hydrodynamic retardation by shear force, and the latter yields a small electric force. In the present case, because the decrease in the electric force is larger than that in the hydrodynamic retardation as a/d increases, μ^* decreases accordingly. Figure 9 shows the variation of the scaled electrophoretic mobility μ^* as a function of λ for the case of non-uniformly charged particles. This figure reveals that, if the double layer is sufficiently thick ($\kappa a = 0.5$ and 1), the dependence of μ^* on a/d is similar to that for the case of uniformly charged particles illustrated in Figure 8. However, for a sufficiently thin double layer ($\kappa a = 4$), the behavior of μ^* as a/d varies shows a different result from that described above when λ is small. That is, if λ is small (boundary effect unimportant), μ^* may have a local maximum as a/d varies, and it is expected to decreasing to that of uniformly charged particles as $a/d \rightarrow 0$. The former is a net result of the competing effects between electric force and hydrodynamic retardation described in the discussion on Figure 8 as a/d varies, and the later is due to the fact that the contribution of the charge on the basal surface of a particle can be ignored when its length approaches infinite.

IV. Conclusion

In summary, the boundary effect, the presence of other particle, and the charged conditions on a particle on its electrophoretic behavior are examined simultaneously by considering the electrophoresis of two identical cylindrical particles along the axis of an uncharged cylindrical pore. The results of numerical simulation reveal the followings: (a) The dependence of the mobility of a particle on the separation distance between two particles is more sensitive to that for the case of two identical spheres in an unbound fluid. This dependence becomes insignificant, however, if the boundary

effect is significant. (b) The effect of particle-particle interaction on the mobility of a particle becomes significant when the thickness of the double layer surrounding a particle is comparable to half the separation distance between two particles, but this effect is less significant than the boundary effect, which is closely related to the charged conditions on a particle. The boundary effect for the case of non-uniformly charged particles is more significant than that for the case of uniformly charged particles. (c) While the mobility of a particle decreases with the increase in the separation distance between two particles, in general, a local maximum may occur if the double layer is sufficiently thin and the boundary effect is relatively unimportant. (d) If the boundary effect is relatively unimportant, the rate of decrease in the mobility of a particle as the separation distance between two particles increases for non-uniformly charged particles is faster than that for uniformly charged particles. (e) For uniformly charged particles, while the more significant the boundary effect the smaller the mobility, in general, the mobility may have a local maximum if the particle aspect ratio is sufficiently large. The mobility of a particle increases with the increase in its aspect ratio. (f) For non-uniformly charged particles, if the double layer is sufficiently thick, the dependence of the mobility on the particle aspect ratio is similar to that for the case of uniformly charged particles. However, if the double layer is sufficiently thin and the boundary effect is relatively unimportant, the mobility may have a local maximum as the particle aspect ratio varies.

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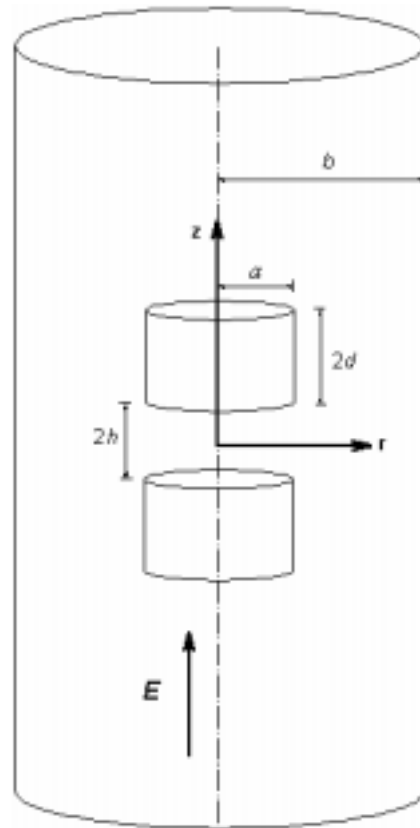


Figure 1. Schematic representation of the problem considered. Two charged cylindrical particles with a separation distance $2h$ are placed on the axis of an infinite cylindrical pore of radius b . An electric field E parallel to the axis of the pore is applied. The cylindrical coordinates (r, θ, z) are adopted with its origin located at the center two particles. $2a$ and $2d$ are respectively the axes of the particle in the r - and in the z -directions.

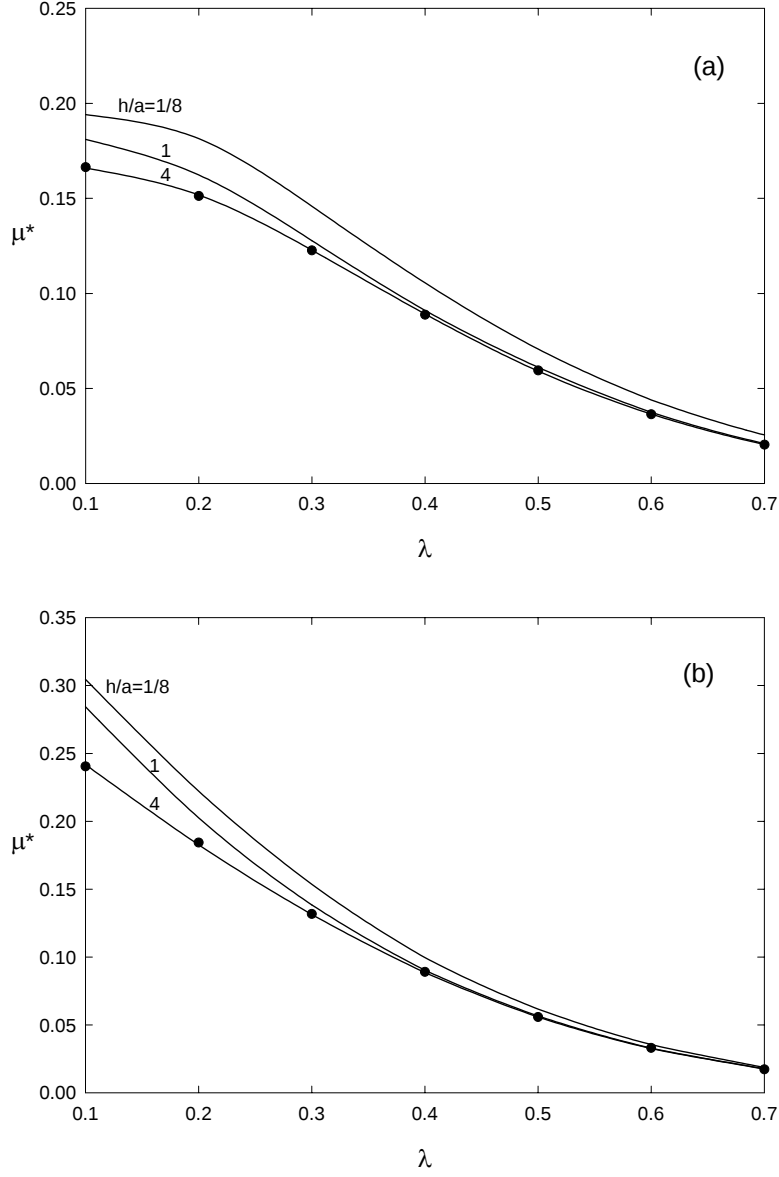


Figure 2. Variation of scaled mobility μ^* as a function of λ at various scaled separation distance between two particles h/a for the case when $\kappa a = 1$, and $a/d = 1$. (a) $\sigma_1 = \sigma_2 = 0.5(\epsilon \kappa k_B T / e)$, (b) $-\sigma_1 = \sigma_2 = 0.5(\epsilon \kappa k_B T / e)$. Discrete symbols are the results for the case when only one cylinder is present in a cylindrical pore.²⁰

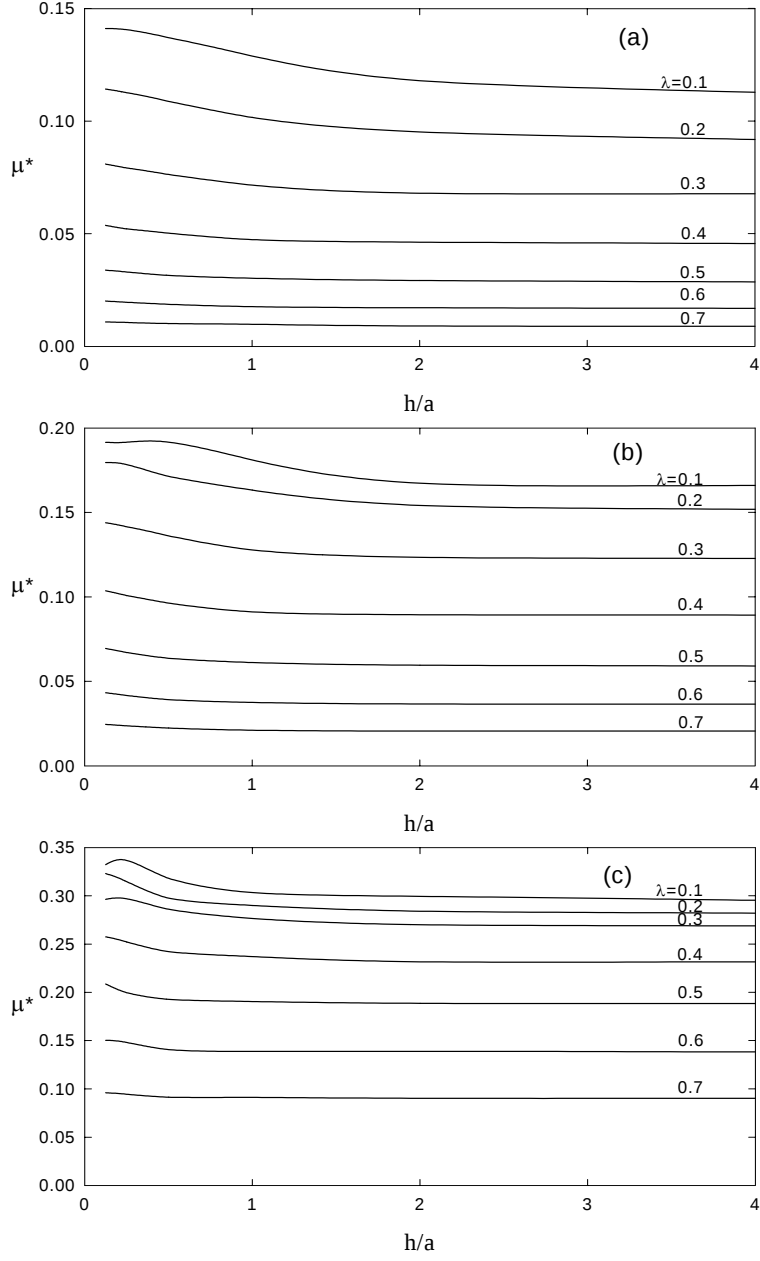


Figure 3. Variation of scaled mobility μ^* as a function of h/a at various λ and κa for the case when $a/d=1$, and $\sigma_1 = \sigma_2 = 0.5(\epsilon\kappa k_B T/e)$. (a) $\kappa a = 0.5$, (b) $\kappa a = 1$, (c) $\kappa a = 4$.

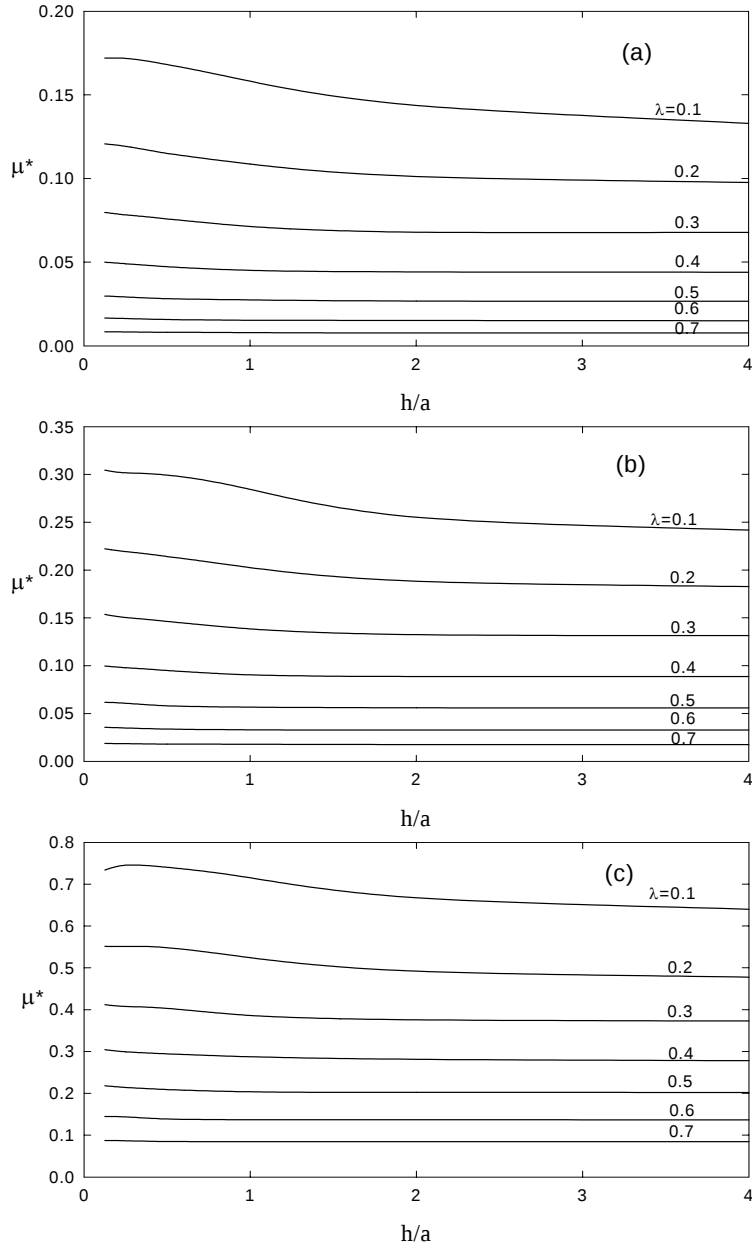


Figure 4. Variation of scaled μ^* as a function of h/a for the case of Figure 3 except that $-\sigma_1 = \sigma_2 = 0.5(\epsilon\kappa k_B T/e)$. (a) $\kappa a = 0.5$, (b) $\kappa a = 1$, (c) $\kappa a = 4$.

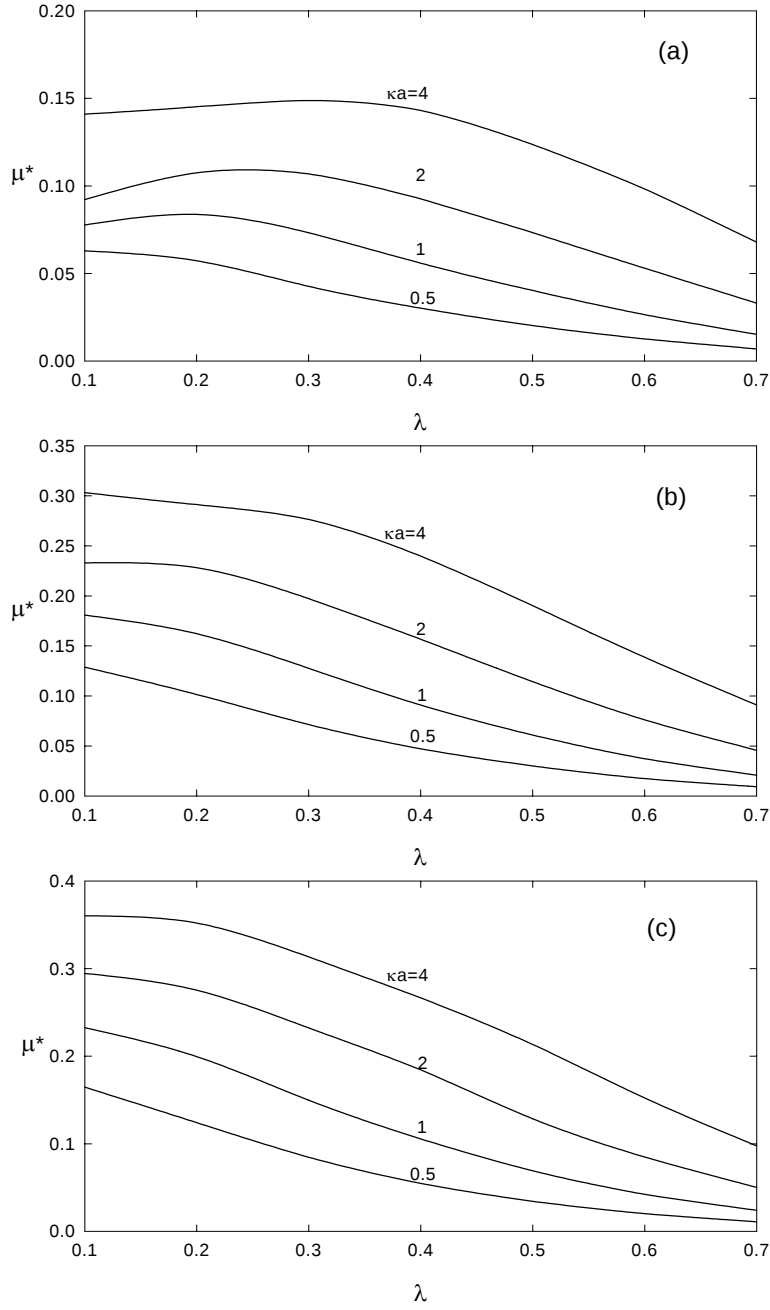


Figure 5. Variation of scaled mobility μ^* as a function of λ at various combinations of a/d and κa for the case when $h/a=1$, and $\sigma_1 = \sigma_2 = 0.5(\epsilon\kappa k_B T / e)$. (a) $a/d=4$, (b) $a/d=1$, (c) $a/d=0.5$.

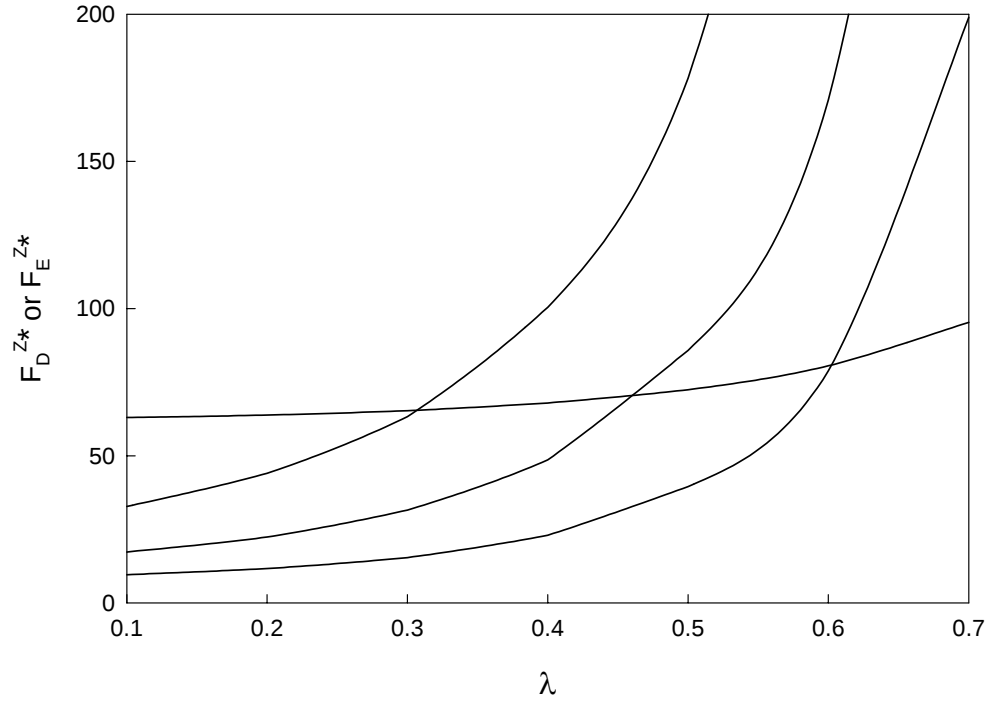


Figure 6. Variation of scaled hydrodynamic force $F_D^Z^*$ ($= F_D^Z / n_{10} k_B T$) and scaled electric force $F_E^Z^*$ ($= F_E^Z / n_{10} k_B T$) acting on a particle at various μ^* as a function of λ for the case when $\kappa a = 4$, $h/a = 1$, and $a/d = 4$. (a) $\sigma_1 = \sigma_2 = 0.5(\epsilon \kappa k_B T / e)$, curves 1, 2, and 3 correspond respectively to $\mu^* = 0.20$, 0.17 , and 0.15 . (b) $-\sigma_1 = \sigma_2 = 0.5(\epsilon \kappa k_B T / e)$, curves 1, 2, and 3 correspond respectively to $\mu^* = 0.4$, 0.2 , and 0.1 .

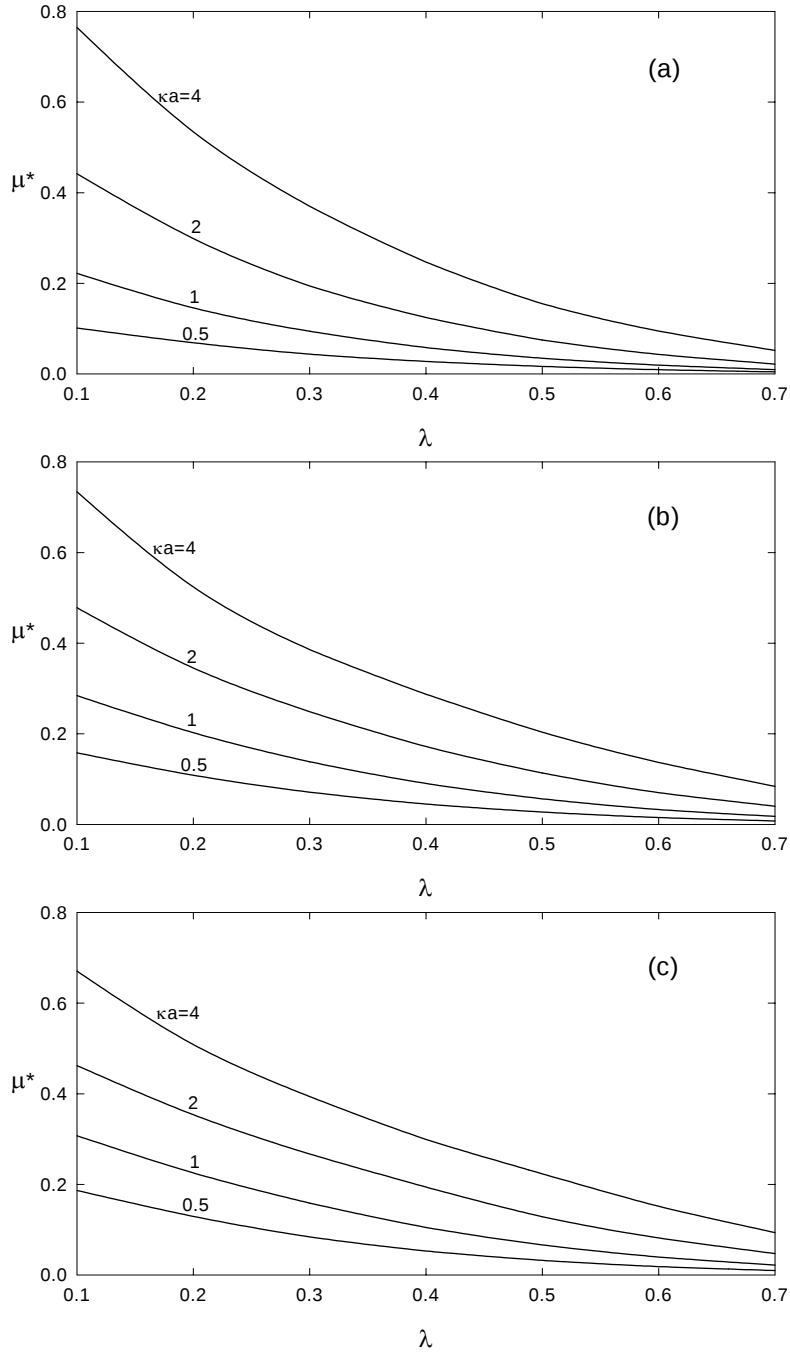


Figure 7. Variation of scaled mobility μ^* as a function of λ for the case of Figure 5 except that $-\sigma_1 = \sigma_2 = 0.5(\varepsilon\kappa k_B T / e)$. (a) $a/d=4$, (b) $a/d=1$, (c) $a/d=0.5$.

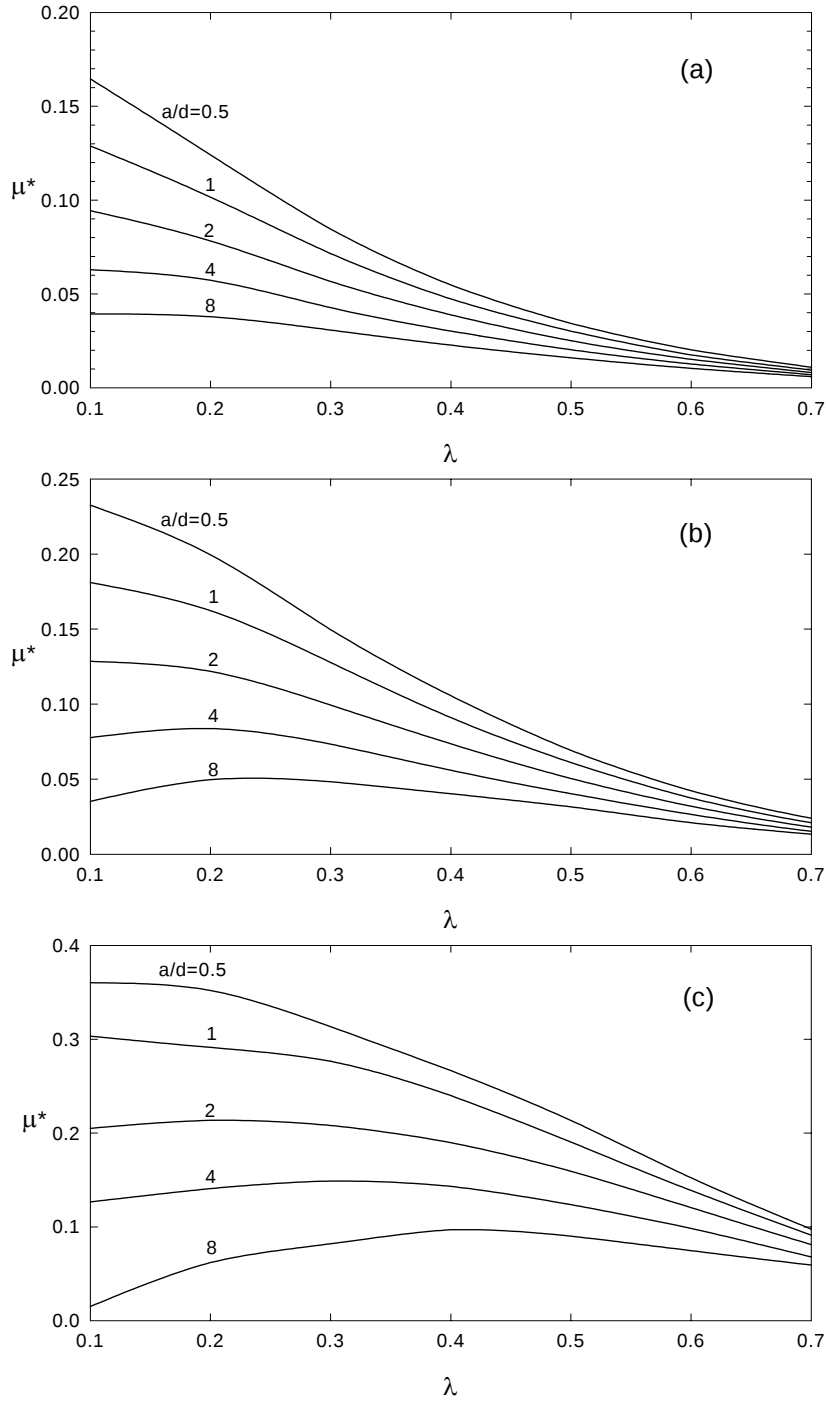


Figure 8. Variation of scaled mobility μ^* as a function of λ at various combinations of a/d and κa for the case when $h/a=1$ and $\sigma_1 = \sigma_2 = 0.5(\epsilon\kappa k_B T/e)$. (a) $\kappa a=0.5$, (b) $\kappa a=1$, (c) $\kappa a=4$.

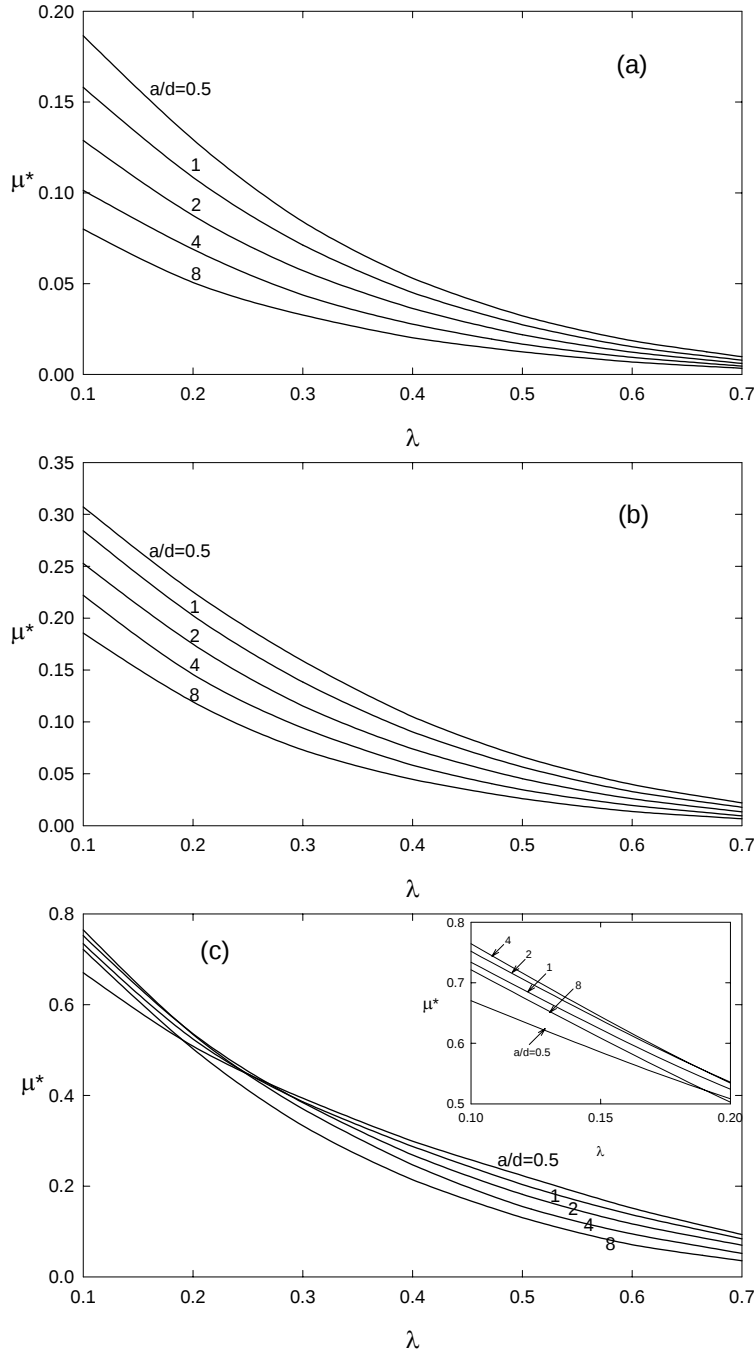


Figure 9. Variation of scaled mobility μ^* as a function of λ for the case of Figure 8 except that $-\sigma_1 = \sigma_2 = 0.5(\epsilon\kappa k_B T/e)$. (a) $\kappa a = 0.5$, (b) $\kappa a = 1$, (c) $\kappa a = 4$.