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Electrophoresis of a Rigid Sphere in a Carreau Fluid Normal to a Large Charged Disk

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The electrophoresis of a rigid sphere in a Carreau fluid normal to a large disk is analyzed theoretically under the conditions of low surface potential and weak applied electric field. Previous analyses are extended to the case where a disk can be charged, and a more realistic electrostatic force formula is applied. We show that the qualitative behavior of a sphere depends largely on its distance from a disk, the thickness of double layer, and the nature of a fluid. In general, the presence of a disk has the effect of increasing the conventional hydrodynamic drag on a sphere, and a decrease in the thickness of the double layer surrounding a sphere has the effect of enhancing the shear-thinning effect. However, this might not be the case if a sphere is uncharged and a disk is charged, where the osmotic pressure field and the induced charge on the sphere surface can be significant. The shear-thinning effect is important only if the thickness of double layer is sufficiently thick. This result can play a significant role in practice such as in electrophoretic deposition, where the deposition electrode is charged and the fluid medium is usually of shearing-thinning nature.

1. Introduction

Since the report of the classic analytical result of electrophoresis by Smoluchowski,[1] this electrokinetic phenomenon has been studied extensively in the past. Up to now, modifications of the original model so that it is capable of describing, more realistically, the conditions in practice are still one of the important issues in relevant fields. Electrophoresis has been applied widely in various areas serving both as an analytical tool to characterize the surface properties of an entity and as an operation to separate/differentiate entities of different natures. One of the key effects in conducting electrophoresis in practice is the boundary effect. The influence of a boundary can be significant, for instance, when electrophoresis is conducted in a relatively narrow space and/or when the behavior of a particle near a surface is of primary concern. In these cases, the governing electrokinetic equations need to be solved under appropriately defined boundary conditions. A considerable amount of theoretical efforts has been made in the past to simulate various types of boundary effect on electrophoresis. One of these is the electrophoresis of a particle normal to a boundary. Electrophoretic deposition (EPD), for example, involves this type of problem. Depending upon the geometry considered, previous results can roughly be divided into three categories: the electrophoresis of a particle normal to a plane,[2−10] to a disk,[11,12] and to a cavity.[13−15] In these cases, if a boundary is uncharged, its influence on the behavior of a particle is twofold: the increase in the viscous drag acting on a particle and the distortion of the local electric field near the particle. The situation becomes more complicated if a boundary is charged. In this case, several other factors need to be considered, for example, an electroosmotic flow field[6,7,15−24] and an osmotic pressure field[6,7,12,15−24] will be established near the boundary, and charge will be induced on the particle surface as it approaches the boundary. These factors can have a profound influence on the electrophoretic behavior of a particle.[6,7,12,15−24]

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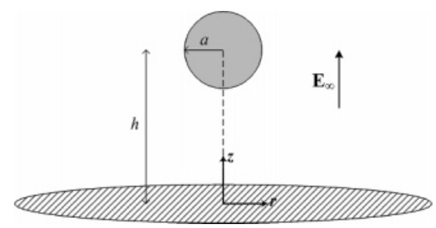

Figure 1. Schematic representation of the problem considered where a rigid sphere of radius *a* moves in a Carreau fluid normal to a large disk as a response to an applied uniform electric field $\mathbf{E}_\infty$ parallel to the *z* direction. The cylindrical coordinates (r, θ, z) with the origin at the center of the disk are adopted; the disk lies on the plane $z = 0$; and *h* is the distance between the center of the sphere and the disk.

Another factor which can be of practical significance is the nature of fluid medium. The introduction of surfactant and polymer into a colloidal dispersion to improve its stability,[25] for example, can yield a non-Newtonian dispersion. These materials are also utilized often as additives to the electrolyte solutions employed in protein or DNA capillary zone electrophoresis for minimizing protein or DNA-capillary wall interactions, for improving selectivity and resolution, and for controlling the electroosmotic flow.[26,27] A dispersion can also be of non-Newtonian nature if the concentration of the dispersed phase becomes appreciable.[28] Several attempts were made recently to model the electrophoresis of a particle in a shear-thinning Carreau fluid. The geometry considered includes a sphere at the center of a spherical cavity,[29] a sphere at an arbitrary position in a spherical cavity,[30] a concentrated spherical dispersion,[31,32] a sphere normal to a plane,[33] and a sphere along the axis of a cylindrical pore.[34] In general, the shear-thinning nature of a fluid is found to be advantageous to the movement of a particle, and the thinner the double layer surrounding a particle the more significant the effect of shear thinning.[29−34]

In this study, the influence of a charged boundary on the electrophoretic behavior of a particle in a Carreau fluid is investigated by considering the electrophoresis of a spherical particle normal to a large disk under the conditions of low surface potential and weak applied electric field. This extends

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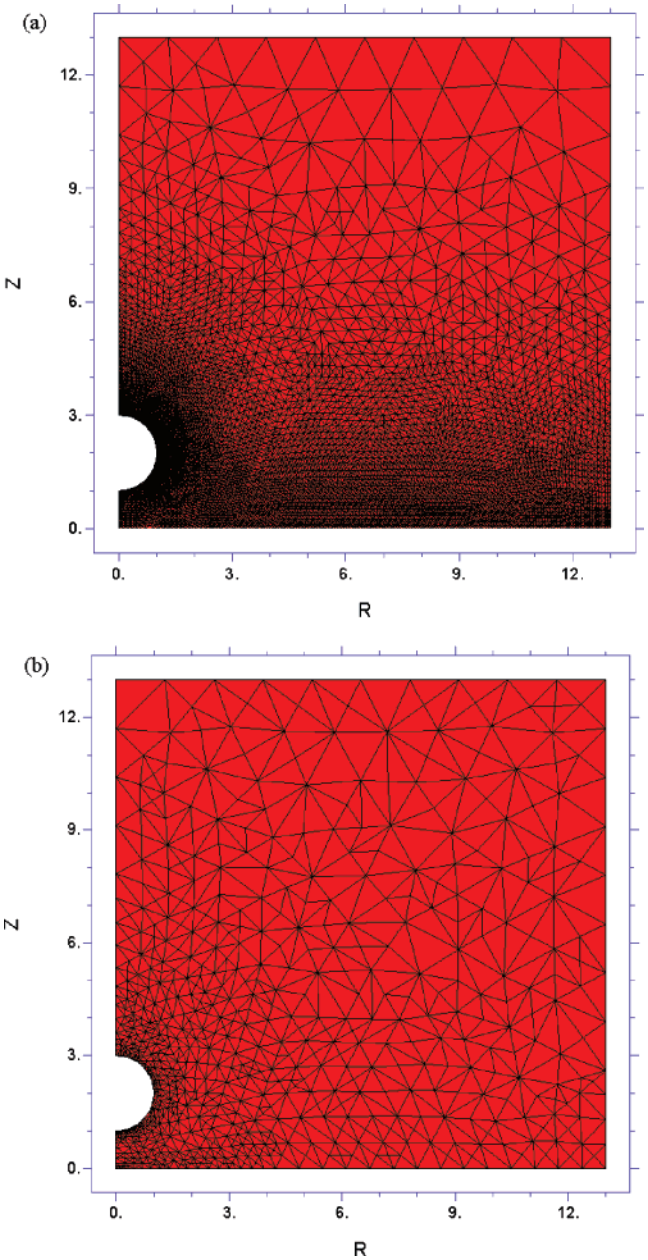


Figure 2. Typical mesh used in the resolution of the electric field (a) and that of the flow field (b) for a Carreau fluid with $N = 0.8$ and $\alpha U/a = 0.8$. Other parameters used are $\zeta_p^*(= \zeta_p/(k_BT/e)) = 1$, $\zeta_w^*(= \zeta_w/(k_BT/e)) = 1$, $h/a = 2$, and $\kappa a = 1$.

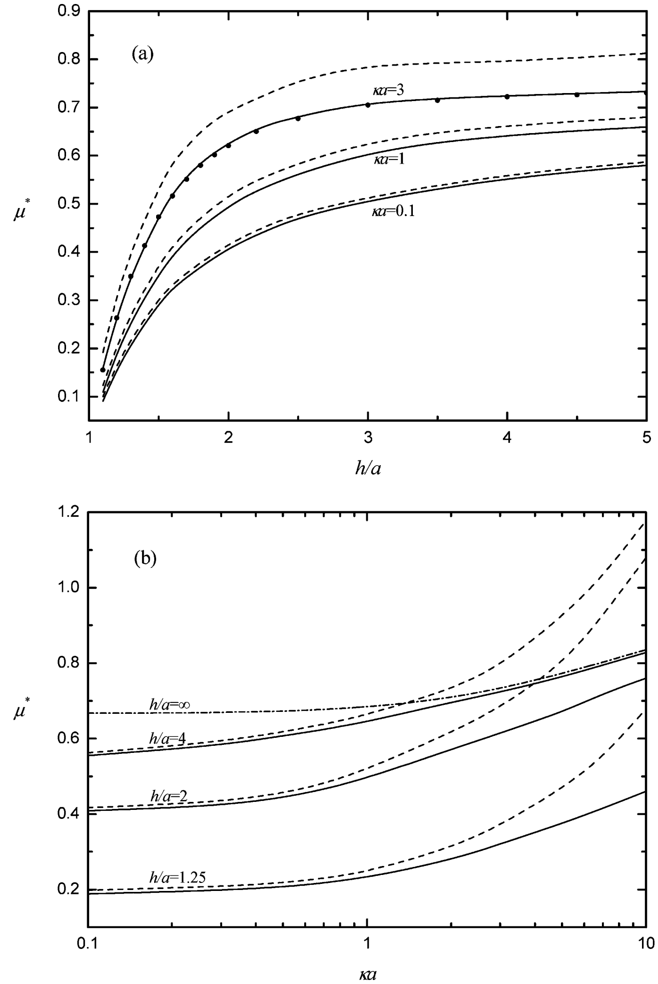


Figure 3. Variation of the scaled mobility μ^* as a function of (h/a) for various κa's (a) and as a function of κa for various (h/a)'s (b) for the case of a positively charged sphere and an uncharged disk. Solid curves present results for a Newtonian fluid; dashed curves present results for a Carreau fluid with $N = 0.8$ and $\alpha U/a = 0.8$; discrete symbols, numerical result based on Shugai and Carnie;[7] dashed-dotted curves, analytical result predicted by Henry's formula. Other parameters used are $\zeta_p^* = 1$ and $\zeta_w^* = 0$.

previous analyses in that the fluid phase is non-Newtonian and that a boundary is charged. The latter is of practical importance in applications such as EPD.[35,36] Also, a recently derived formula,[37,38] which is more accurate than those used in the literature, for the evaluation of the electric force acting on a particle is adopted. The influences of the charged conditions of the sphere and the disk surfaces, the thickness of the double layer, and the nature of a fluid on the electrophoretic behavior of a sphere are discussed.

2. Theory

Referring to Figure 1, we consider the electrophoresis of a rigid, nonconductive, sphere of radius a in an incompressible Carreau fluid[39,40] normal to a large, conducting disk. Suppose that the disk is sufficiently large so that it can be treated as an infinite plane.[4] The cylindrical coordinates (r, θ, z) with the origin at the center of the disk are adopted, and the disk lies on the plane $z = 0$. Since the present problem is θ-symmetric, only the (r, z) domain needs to be considered; that is, it is of a two-dimensional nature. Let h be the distance between the center of the sphere and the disk. A uniform electric field $\mathbf{E}_\infty$ in the z direction is applied, and its strength is E_∞. Both the surface of the sphere and that of the disk are maintained at constant potential, denoted as ζ_p and ζ_w, respectively.

The constitutive equation of a purely viscous non-Newtonian fluid can be expressed by

$$\boldsymbol{\tau} = -\eta(\dot{\gamma})\dot{\boldsymbol{\gamma}} \tag{1}$$

where $\boldsymbol{\tau}$, $\eta(\dot{\gamma})$, and $\dot{\boldsymbol{\gamma}}$ are the stress tensor, the fluid viscosity, and the rate of strain tensor, respectively; $\dot{\boldsymbol{\gamma}} = \nabla\mathbf{u} + (\nabla\mathbf{u})^T$, and $\dot{\gamma}$ is the magnitude of $\dot{\boldsymbol{\gamma}}$; ∇, $\mathbf{u}$, and $\mathbf{T}$ are the gradient operator, the fluid velocity, and the matrix transpose, respectively.[40] For the Carreau fluid considered in this study, we choose[39,40]

$$\eta(\dot{\gamma}) = \eta_\infty + (\eta_0 - \eta_\infty)[1 + (\alpha\dot{\gamma})^\beta]^{(N-1)/\beta} \tag{2}$$

where η_0 and η_∞ are the viscosities corresponding to the zero-shear rate and the infinite-shear rate, respectively; α is the

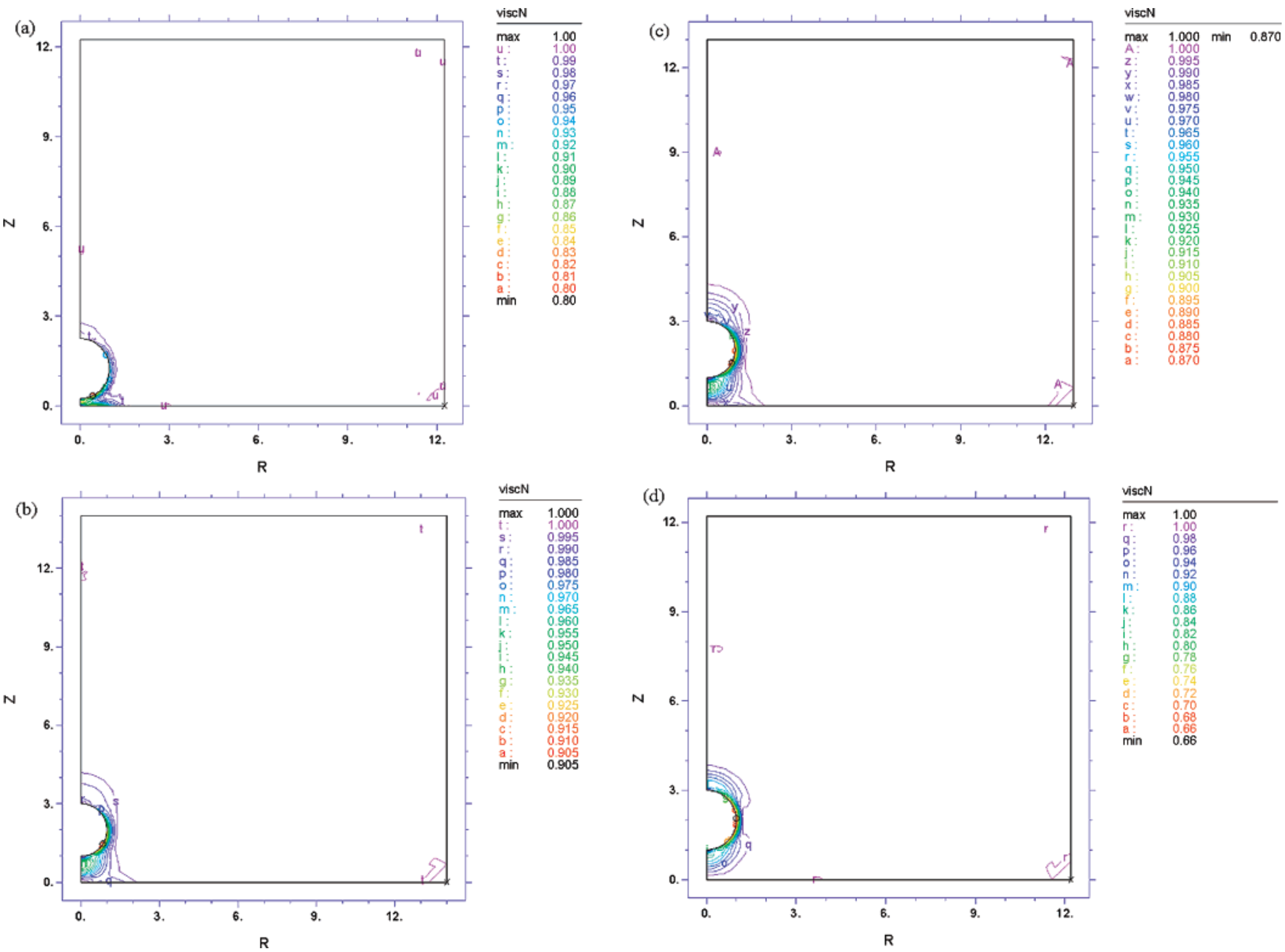


Figure 4. Contours of the scaled viscosity η^*, viscN, for a Carreau fluid with $N = 0.8$ and $\alpha U/a = 0.8$ for the case of Figure 3. (a) $h/a = 1.25$ and $\kappa a = 1$, (b) $h/a = 2$ and $\kappa a = 0.5$, (c) $h/a = 2$ and $\kappa a = 1$, and (d) $h/a = 2$ and $\kappa a = 5$.

relaxation time constant, N is the power-law index, and β is a dimensionless parameter describing the transition region between the zero-shear-rate region and the power-law region. In practice, β is close to 2 and η_∞ is small,[39,40] and therefore, eq 2 reduces to

$$\eta(\dot{\gamma}) = \eta_0[1 + (\alpha\dot{\gamma})^2]^{(N-1)/2} \tag{3}$$

Note that a Newtonian fluid can be recovered as a special case of the preset Carreau fluid by letting $N = 1$ and/or $\alpha = 0$, and a power-law fluid can be obtained by assuming a large α.

We assume that the surface potential of a sphere and/or a disk is sufficiently low and $\mathbf{E}_\infty$ is relatively weak compared with that established by the sphere and/or the disk. The former is appropriate if the surface potential is lower than about 25 mV, and the latter is adequate if E_∞ is lower than about 25 kV/m, which is usually satisfied for conditions of practical significance.[37] Also, the system under consideration is at a pseudo-steady state;[6,16−18] that is, all of the dependent variables are time-independent. Under these conditions, it can be shown that the electric potential of the system, Ψ, can be described by[41]

$$\nabla^2\Psi_1 = \kappa^2\Psi_1 \tag{4}$$

$$\nabla^2\Psi_2 = 0 \tag{5}$$

where ∇^2 is the Laplace operator, $\Psi = \Psi_1 + \Psi_2$, and Ψ_1 and Ψ_2 are the equilibrium potential in the absence of $\mathbf{E}_\infty$ and the perturbed potential outside the sphere arising from $\mathbf{E}_\infty$, respectively. $\kappa = [\sum_j n_j^0(ez_j)^2/\epsilon k_B T]^{1/2}$ is the reciprocal Debye length, ϵ is the permittivity of the liquid phase, n_j^0 and z_j are the bulk number concentration and the valence of ionic species j, respectively, and e, k_B, and T are the elementary charge, the Boltzmann constant, and the absolute temperature, respectively. We assume that both the sphere surface and the disk surface are maintained at constant potential, the sphere is nonconductive, and the disk is a perfect-conductor. Also, the equilibrium potential vanishes as $z \to \infty$ and/or at a point far away from the sphere, and the perturbed electric field arising from $\mathbf{E}_\infty$ is normal to the disk. On the basis of these assumptions and the symmetric nature of the problem, the boundary conditions associated with eqs 4 and 5 can be expressed as

$$\Psi_1 = \zeta_p \quad \text{and} \quad \mathbf{n}\cdot\nabla\Psi_2 = 0 \quad \text{on the sphere surface} \tag{6}$$

$$\Psi_1 = \zeta_w \quad \text{and} \quad \Psi_2 = \text{constant at } z = 0 \tag{7}$$

$$\Psi_1 = 0 \quad \text{and} \quad \nabla\Psi_2 = -E_\infty\mathbf{e}_z \text{ as } z \to \infty \tag{8}$$

$$\mathbf{n}\cdot\nabla\Psi_1 = 0 \quad \text{and} \quad \mathbf{n}\cdot\nabla\Psi_2 = 0 \quad \text{as } r \to \infty \text{ and } z > 0 \tag{9}$$

Here, $\mathbf{n}$ is the unit normal vector directed into the liquid phase, and $\mathbf{e}_z$ is the unit vector in the z direction.

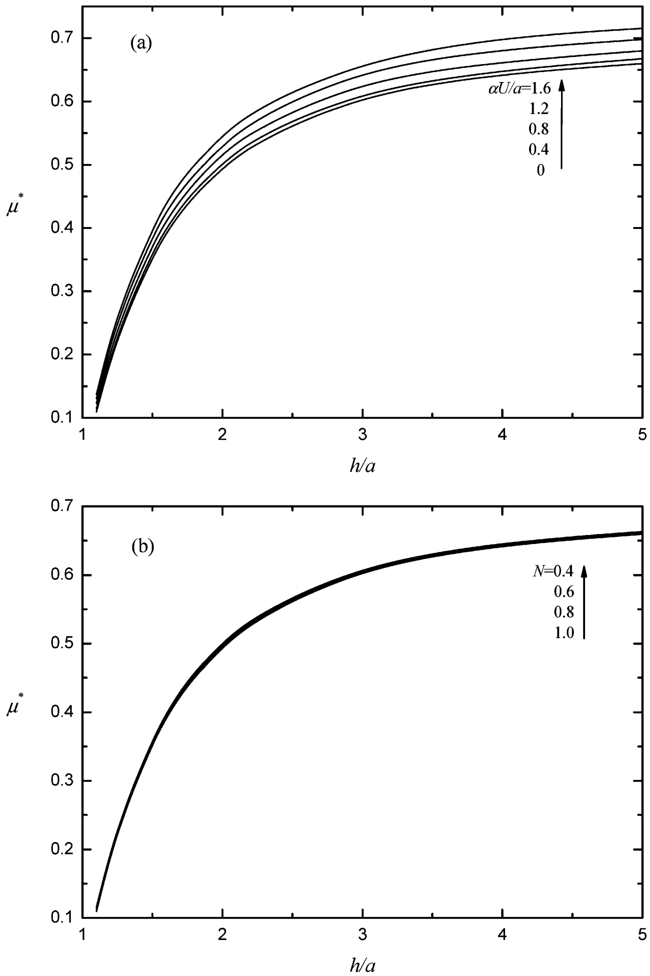


Figure 5. Variation of the scaled mobility μ^* as a function of (h/a) for various ($\alpha U/a$)'s at $N = 0.8$ (a) and that for various N's at $\alpha U/a = 0.2$ (b). Other parameters used are $\zeta_p^* = 1$, $\zeta_w{}^* = 0$, and $\kappa a = 1$.

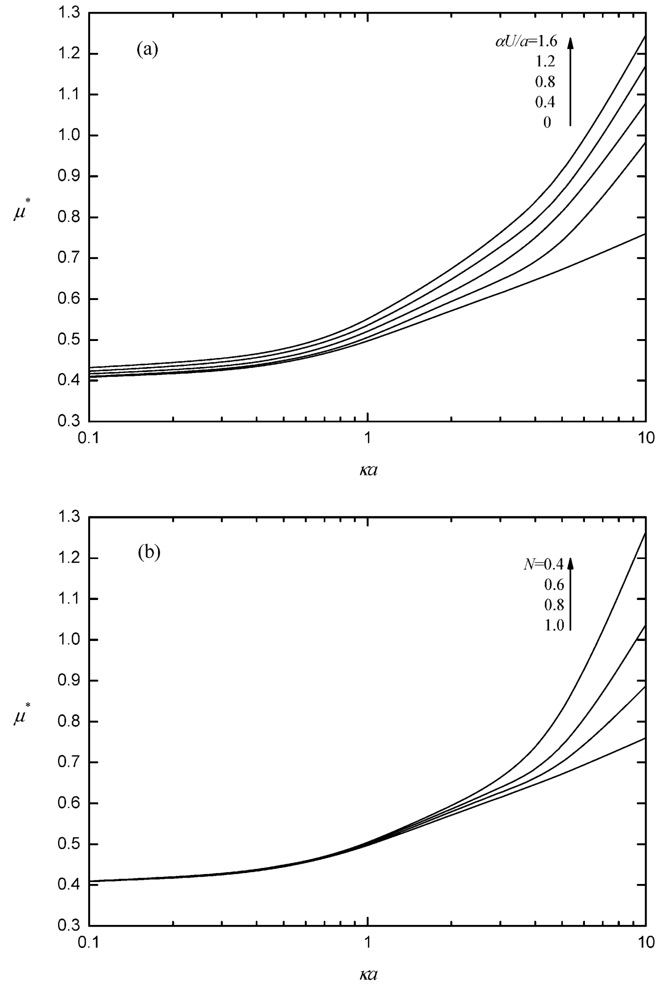


Figure 6. Variation of the scaled mobility μ^* as a function of κa for various ($\alpha U/a$)'s at $N = 0.8$ (a) and that for various N's at $\alpha U/a = 0.2$ (b). Other parameters used are $\zeta_p^* = 1$, $\zeta_w{}^* = 0$, and $h/a = 2$.

At steady state, the flow field of the present problem can be described by[40]

$$\nabla\cdot\mathbf{u} = 0 \tag{10}$$

$$-\nabla\cdot\boldsymbol{\tau} - \nabla p = \rho_e\nabla\Psi_2 \tag{11}$$

where p is the pressure. $\rho_e = \sum_j z_j e n_j^0 \exp(-z_j e\Psi/k_BT)$ is the space charge density, and $\rho_e\nabla\Psi_2$ is the electric body force acting on the fluid.[6,7,15,24,37,38,42] We assume that both the surface of a sphere and that of a disk are no-slip. Let U be the speed of the sphere in the z direction. Then the boundary conditions associated with the flow field under consideration can be expressed as

$$\mathbf{u} = U\mathbf{e}_z \qquad \text{on the sphere surface} \tag{12}$$

$$\mathbf{u} = 0 \text{ at } z = 0 \tag{13}$$

$$\mathbf{u} = 0 \text{ as } z \to \infty \text{ or } r \to \infty \tag{14}$$

The mobility of a sphere can be determined from the fact that the total force acting on a sphere in the z direction vanishes at steady state;[42] that is,

$$F_E + F_D = 0 \tag{15}$$

where F_E and F_D are the z component of the electric force acting on a sphere at steady state and that of the hydrodynamic force acting on a sphere at steady state, respectively. By letting S be the sphere surface, it can be shown that[37,38]

$$F_E = \int\int_S \sigma_p E_z \, dS \tag{16}$$

where $\sigma_p = -\epsilon\mathbf{n}\cdot\nabla\Psi_1$ is the charge density on S, and $E_z = -\partial\Psi_2/\partial z$ is the strength of the local external electric field in the z direction.

By letting $\mathbf{t}$ be the unit tangential vector on S, n be the magnitude of $\mathbf{n}$, and t_z and n_z be respectively the z component of $\mathbf{t}$ and that of $\mathbf{n}$, F_D can be evaluated by[37,43]

$$F_D = \int\int_S \eta \frac{\partial(\mathbf{u}\cdot\mathbf{t})}{\partial n} t_z \, dS + \int\int_S -pn_z \, dS \tag{17}$$

For convenience, we let $\mu^* = U/U_{ref}$ be the scaled mobility where $U_{ref} = \epsilon(k_BT/e)E_\infty/\eta$ is the electrophoretic velocity of a sphere with a constant surface potential (k_BT/e) when an electric field of strength E_∞ is applied.

The governing equations of the present problem and the associated boundary conditions are solved numerically by FlexPDE,[44] a differential equation solver based on a finite element method. Throughout the computation, double precision is used and grid independence is checked to ensure that the mesh used is fine enough. In our case, a total of 19 890 and 1615 nodes are sufficient for the resolution of the electric field and the flow field, respectively, and a convergent result can be

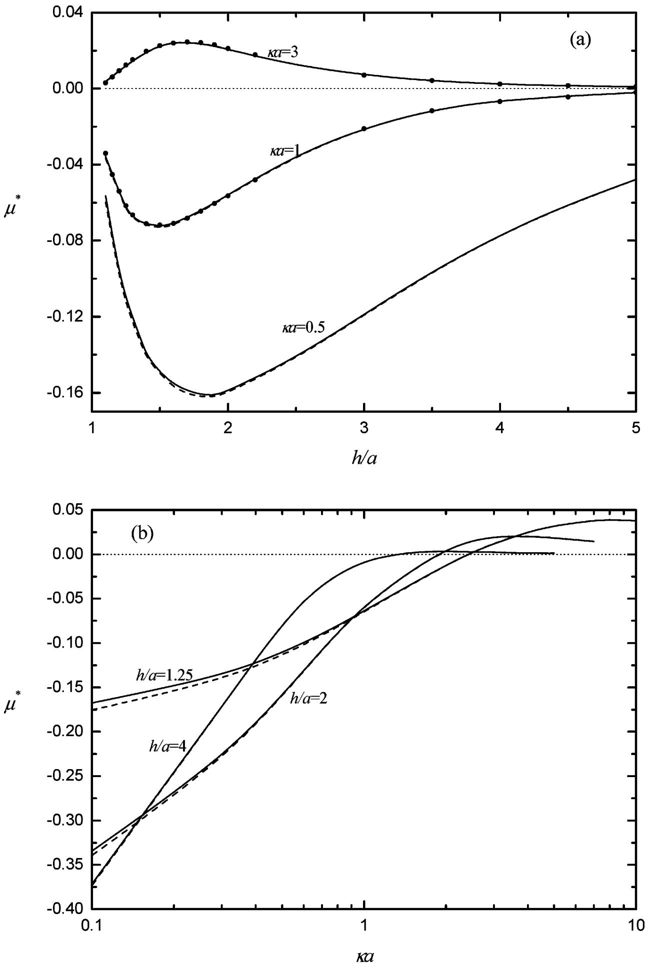


Figure 7. Variation of the scaled mobility μ^* as a function of (h/a) for various κa's (a) and as a function of κa for various (h/a)'s (b). Solid curves, present results for a Newtonian fluid; dashed curves, present results for a Carreau fluid with $N = 0.8$ and $\alpha U/a = 0.8$; discrete symbols, numerical result based on Shugai and Carnie.[7] Other parameters used are $\zeta_p^* = 0$ and $\zeta_w^* = 1$.

obtained by setting an error limit of 10^{-6} for the electric field and 10^{-3} for the flow field. Figure 2 shows the typical mesh used. Note that an unstructured triangular mesh is used with a finer mesh near the particle and a coarser mesh away from it. The applicability and the accuracy of the software adopted were justified previously[30,34] and will also be discussed later.

3. Results and Discussion

The electrophoretic behavior of a sphere is examined through numerical simulation. Three representative cases are considered, namely, a positively charged sphere and an uncharged disk, an uncharged sphere and a positively charged disk, and a positively charged sphere and positively charged disk. Note that, because of the nonlinear nature of the present problem, the results of the last case cannot be obtained directly from a linear combination of those of the first two cases.

3.1. Positively Charged Sphere and Uncharged Disk. Let us consider first the electrophoresis of a positively charged sphere normal to an uncharged disk. For a fixed sphere radius a, the influences of the scaled sphere-disk distance (h/a) and the thickness of double layer measured by κa on the scaled mobility of a sphere μ^* is illustrated in Figure 3. For comparison, the corresponding results for the case of a Newtonian fluid and the result of Shugai and Carnie[7] for the case a sphere moving in a Newtonian fluid normal to an infinite plane are also presented. Figure 3a reveals that the result based on the present numerical method for the case of a Newtonian fluid is consistent with that calculated by Shugai and Carnie,[7] which implies that the performance of the present numerical method is satisfactory. If κa is fixed, for both Newtonian and Carreau fluids, μ^* decreases monotonically with the decrease in (h/a) and $\mu^* \to 0$ as $h/a \to 1$, that is, as the sphere touches the disk. These are expected because the closer a sphere is to a disk, the more significant the hydrodynamic hindrance is on the movement of the sphere because of the presence of the disk. The same trend was observed by Shugai and Carnie.[7] Tang et al.,[8] Chih et al.,[9] and Lee et al.,[10] however, found that the closer the sphere–disk distance, the smaller the mobility of the sphere; but if a sphere is sufficiently close to a plane, its mobility may change the sign; that is, it moves in the opposite direction as that of the applied electric field. The latter arises if a problem is not of totally symmetric nature, such as the present one, an extraneous electric body force, $\rho_e \nabla \Psi_1$, will be included in eq 16 and an extraneous electrostatic force arising from the equilibrium potential, $\int\int_S \epsilon(\partial\Psi_1/\partial n)(\partial\Psi_1/\partial z)\,dS$, is included in eq 17. These terms become significant when a sphere is close to a plane.[15,37,38] Note that, because a nonslip boundary condition is assumed on the planar surface, the mobility of a sphere should vanish as it touches the plane. In general, the mobility of a sphere in a Carreau fluid is larger than that in the corresponding Newtonian fluid because of the shear-thinning nature of the former. We define[33,34]

$$PD = \frac{|\mu^*(\text{Carreau})| - |\mu^*(\text{Newtonian})|}{|\mu^*(\text{Newtonian})|} \times 100\% \quad (18)$$

where μ^*(Carreau) and μ^*(Newtonian) are the scaled mobilities in the present Carreau fluid and in the corresponding Newtonian fluid, respectively. The larger the PD, the more important the effect of shear thinning of a Carreau fluid is. In Figure 3a, if $\kappa a = 1$, PD are 6.44, 4.63, and 2.92% for $h/a = 1.25$, 2, and 4, respectively; that is, the closer a sphere is to a disk, the more significant is the effect of shear thinning, as is also justified by Figure 4. Similar behavior was also observed by Lee et al.[33] This can be explained by the presence of a disk having the effect of distorting the flow field below a sphere, implying that the spatial variation in the shear rate in the present case is greater than that in the case of an isolated sphere, and so is the spatial variation in the scaled viscosity $\eta^* = \eta/\eta_0$, as can be seen in Figure 4. Note that if $\eta^* = 1$, the fluid is Newtonian. For a fixed (h/a), Figure 3b indicates that μ^* increases monotonically with the increase in κa for both Newtonian and Carreau fluids, but μ^* is more sensitive to the variation in κa in the latter. Also, for a fixed κa, μ^*(Carreau) > μ^*(Newtonian), and [μ^*(Carreau) − μ^*(Newtonian)] increases with the increase in κa. Similar behavior was also observed in the electrophoresis of a rigid sphere at the center of a spherical cavity[29] and along the axis of a long cylindrical pore.[34] Note that the larger the κa, the thinner the double layer surrounding a sphere, the greater the absolute value of the gradient of the potential on its surface, the higher the surface charge density, and, therefore, the greater the electrical driving force acting on it. Also, because both the sphere surface and the disk are nonslip, a characteristic shear rate $(U - 0)/\kappa^{-1}$ can be defined,[29] which suggests that the larger the κa, the greater the shear rate and the more significant the shear-thinning effect of a fluid, as is justified in Figure 3. This explains why [μ^*(Carreau) − μ^*(Newtonian)] increases with the increase in κa. For $h/a = 2$, PD are 2.05, 4.63, 17.39, and 44.21% for $\kappa a = 0.1$, 1, 5, and 10, respectively, which suggests that the thinner the double layer surrounding a sphere is, the

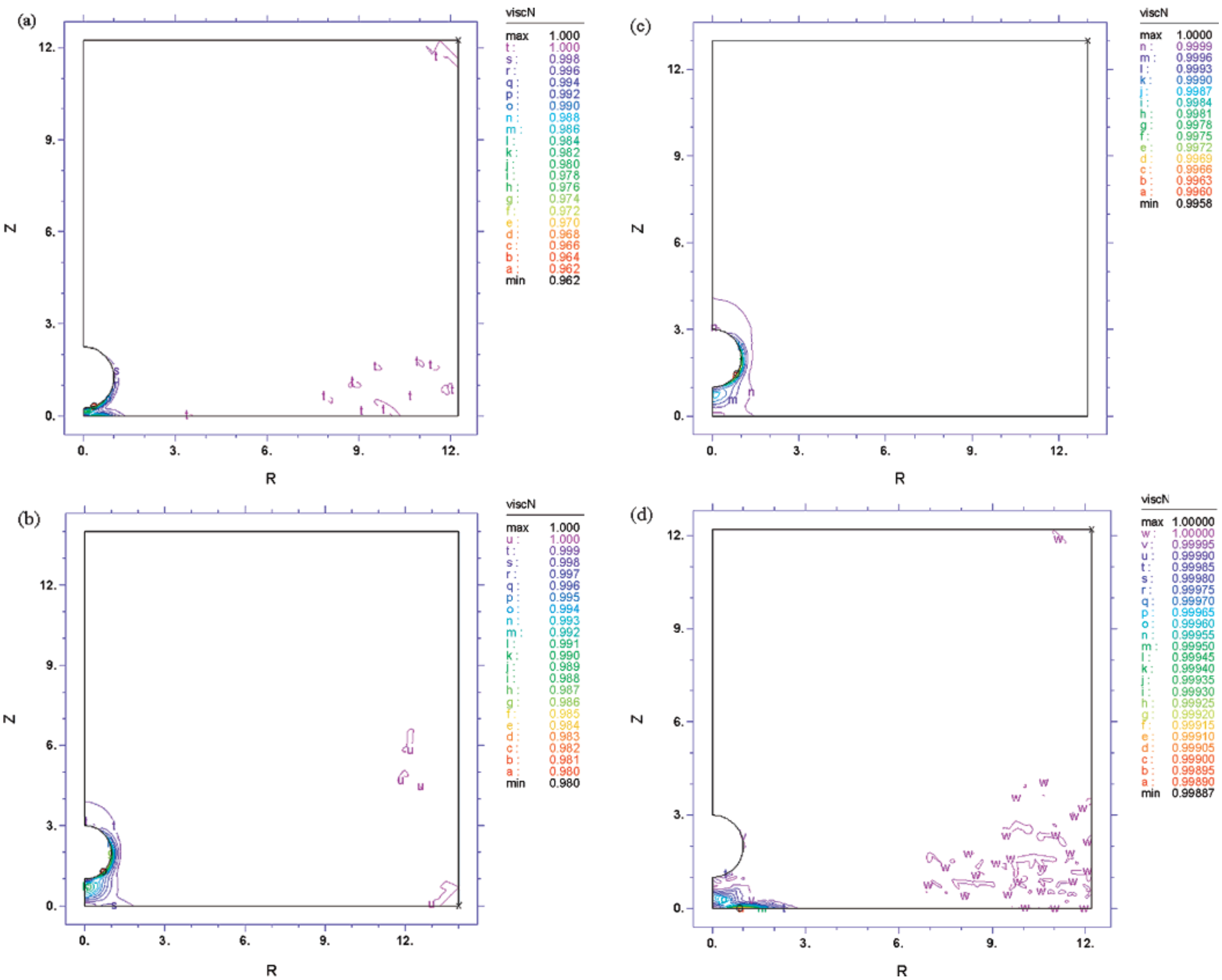
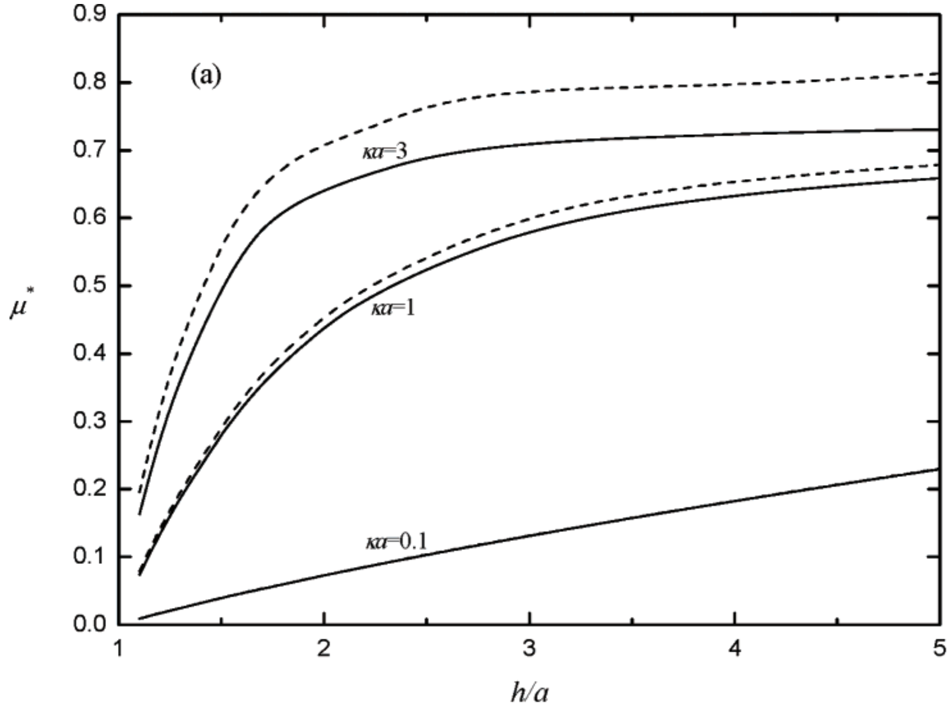


Figure 8. Contours of the scaled viscosity η^*, viscN, for a Carreau fluid with $N = 0.8$ and $\alpha U/a = 0.8$ for the case of Figure 7. (a) $h/a = 1.25$ and $\kappa a = 1$, (b) $h/a = 2$ and $\kappa a = 0.5$, (c) $h/a = 2$ and $\kappa a = 1$, and (d) $h/a = 2$ and $\kappa a = 5$.

more significant the effect of shear thinning is. This behavior is inconsistent with that reported by Lee et al.,[33] where the behavior of the shear-thinning effect as κa varies is unclear. This is because the thicker the double layer surrounding a sphere, the more significant the presence of the disk, and the more serious the deviation arising from the consideration of the extraneous terms, $\rho_e \nabla \Psi_1$ and $\int\int_S \epsilon(\partial\Psi_1/\partial n)(\partial\Psi_1/\partial z)dS$, in the calculation of the mobility. According to Figure 4, the viscosity of the fluid varies most significantly in the gap between the sphere and the disk, and the minimum viscosity occurs on the sphere surface. This minimum declines with the decrease in (h/a) and with the increase in κa. The latter was also observed in a study of the electrophoresis of a concentrated dispersion of spheres[31] and in the electrophoresis of a sphere along the axis of a cylindrical pore.[34]

Figures 5 and 6 illustrate the influences of the nature of a Carreau fluid on the scaled mobility of a sphere μ^*. The qualitative behaviors of μ^* as the scaled sphere−disk distance (h/a) or the double layer thickness κa varies are similar to those for the case of a Newtonian fluid, also, the larger the deviation of N from unity and/or the deviation of $\alpha U/a$ from 0, the more significant the effect of shear thinning, and therefore, the larger the μ^*, which is consistent with the relevant theoretical results in the literature.[29−34] Figure 6 shows that, if κa is small, μ^* is more sensitive to the variation in $\alpha U/a$ than to the variation in N. The same trend was also observed in the electrophoresis of a sphere at the center of a spherical cavity.[29]

3.2. Uncharged Sphere and Positively Charged Disk. The influences of the scaled sphere-disk distance (h/a) and the thickness of double layer κa on the scaled mobility of a sphere μ^* for the case in which the sphere is uncharged and the disk positively charged are presented in Figure 7. For comparison, the corresponding results for the case of a Newtonian fluid and the numerical results of Shugai and Carnie[7] are also shown. The present results agree well with those of Shugai and Carnie.[7] In the present case, negative charge is induced on the surface of a sphere, and it experiences an electric force in the $-z$ direction, and an osmotic pressure field is established.[12] As pointed out by Shugai and Carnie[7] and Hsu et al.,[12] for an isolated, charged disk, the spatial variation of mobile ions is $\rho_{e,\mathrm{iso}}(z) = -\kappa^2\epsilon\zeta_w \exp(-\kappa z)$, and an equilibrium osmotic or hydrostatic pressure field, $p_{\mathrm{iso}}(z) = \kappa\epsilon\zeta_w E_\infty \exp(-\kappa z)$, is established to balance the electric body force. If both ζ_w and E_∞ are positive, $-\mathrm{d}p_{\mathrm{iso}}/\mathrm{d}z = -\rho_{e,\mathrm{iso}}(z)E_\infty > 0$, and both the sphere and the fluid feel a pressure in the z direction when the sphere disturbs the equilibrium pressure field. If we neglect the influence on the equilibrium system due to the presence of the sphere, the z component of the hydrostatic pressure acting on the sphere is $F_p = 4\pi a\epsilon\zeta_w E_\infty \exp(-\kappa h)[\cos h(\kappa a) - \sin h(\kappa a)/\kappa a]$, which is the last term of eq 12 of Shugai and Carnie.[7]



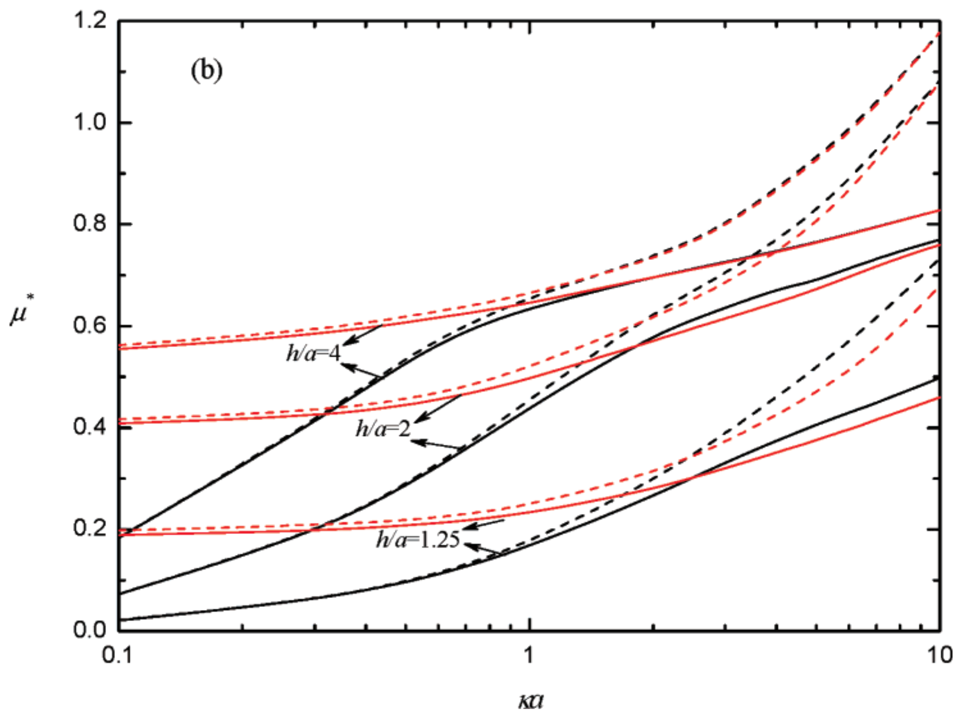


Figure 9. Variation of the scaled mobility μ^* as a function of (h/a) for various κa's (a) and as a function of κa for various (h/a)'s (b) for the case when $\zeta_p^* = 1$ and $\zeta_w^* = 1$. Red curves, results of Figure 3b; solid curves, present results for a Newtonian fluid; dashed curves, present results for a Carreau fluid with $N = 0.8$ and $\alpha U/a = 0.8$.

Therefore, although the body force acting on the fluid is in the $-z$ direction, its movement is influenced appreciably by a positive pressure drop, which is in the z direction. In the case of an uncharged sphere and a charged disk, the electrostatic force changes appreciably as the former approaches the latter because of a fast increase in the negative charge induced on the sphere surface. On the other hand, the hydrostatic pressure acting on a sphere increases with the decrease in h/a because both the sphere and the fluid are influenced by an appreciable increase in the positive pressure drop as stated previously. The competition of these two forces affect appreciably the behavior of a sphere and makes its electrophoretic behavior more complicated than that in the case of a charged sphere and an uncharged disk. As can be seen in Figure 7a, for both Newtonian and Carreau fluids, $|\mu^*|$ may have a local maximum as h/a varies, which was observed but not discussed in Shugai and Carnie,[7] where the fluid is Newtonian. The presence of the local maximum is the net results of the competition of the forces including the induced electric force in the $-z$ direction, the osmotic pressure in the z direction, and the drag as h/a varies. If $\kappa a = 1$, a negative electric force dominates, and an increase in its magnitude leads to a more negative μ^* as h/a decreases from infinite. A further decrease in h/a causes a large increase in the drag, yielding a negative maximum of μ^* at $h/a \cong 1.5$.

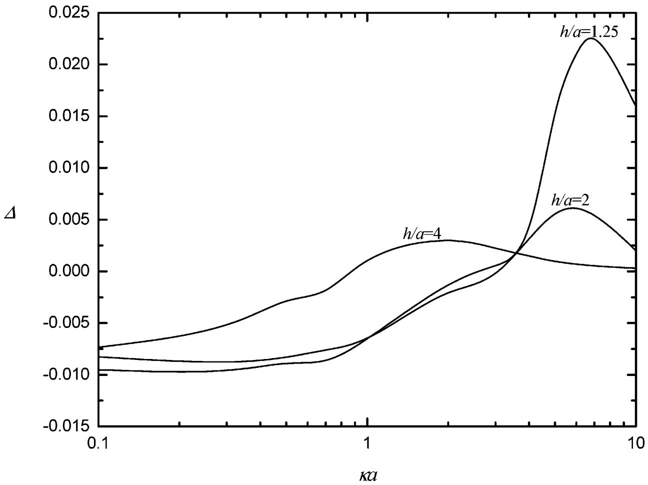


Figure 10. Variation of $\Delta = \{[\mu^*(\text{Carreau})-\mu^*(\text{Newtonian})]_{\text{sphere and disk positively charged}} - [\mu^*(\text{Carreau})-\mu^*(\text{Newtonian})]_{\text{sphere positively charged, disk uncharged}}\}$ for the cases of Figures 3b and 9b.

Similar reasoning is applicable to the curve corresponds to $\kappa a = 3$ in Figure 7a, except that a positive hydrostatic pressure due to the presence of a positive charged disk now dominates, leading to a positive maximum in μ^* at $h/a \cong 1.7$. Therefore, the existence of a local maximum in $|\mu^*|$ seen in Figure 7a is expected because $\mu^* \to 0$ as a sphere touches a disk ($h/a \to 1$), as is required by the non-slip condition assumed on the disk surface, or it is far away from the disk surface ($h/a \to \infty$). Figure 7b indicates that, for both Newtonian and Carreau fluids, μ^* may change its sign from negative to positive as κa increases, and it may have a local maximum; the later was not observed in Shugai and Carnie.[7] These phenomena arise from a sudden raise and fall in F_p for $\kappa(h-a) = \kappa a(h/a - 1)$ in the range [2, 4] and are the net result of the competition between the induced electric force in the $-z$ direction and the osmotic pressure in the z direction mentioned previously. First, because the amount of negative charge induced on the sphere surface increases with the decrease in κa, the corresponding electric force acting on the sphere increases accordingly. Second, for a fixed h/a, as κa increases, the pressure acting on a sphere increases first and then decreases. The pressure vanishes as $\kappa a \to \infty$. In short, the influence of the presence of a charged disk is twofold: it induces charge on the sphere surface, and it establishes a pressure field. The later is responsible for the presence of a local maximum in F_p as $\kappa(h-a)$ varies in the range [2, 4]. The former yields an electric force, which declines monotonically with the increase of $\kappa(h-a)$ in the range [2, 4]. Also, the electric force is much smaller than F_p. As is seen in Figure 7b, the net result is that μ^* has a local maximum and/or varies drastically with κa. The $|\mu^*|$ for a Carreau fluid is larger than that for the corresponding Newtonian fluid, which is consistent with the electroosmotic mobility of polymer solutions in a charged fused silica capillary[45] and can be explained by the shear-thinning effect of a Carreau fluid. In Figure 7, if $\kappa a = 1$, *PD* are 2.77, 1.25, and

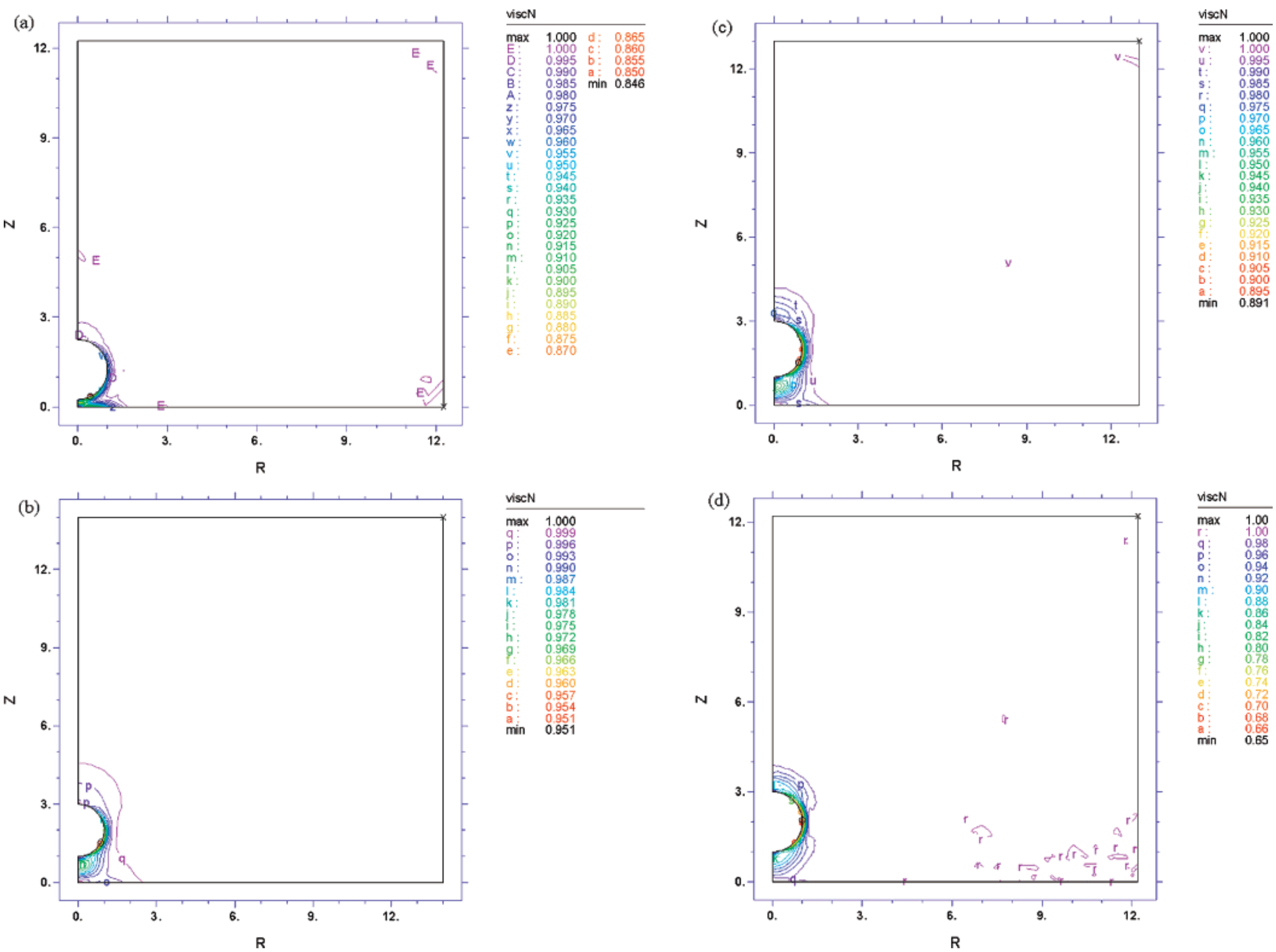


Figure 11. Contours of the scaled viscosity η^*, viscN, for a Carreau fluid with $N = 0.8$ and $\alpha U/a = 0.8$ for the case of Figure 9. (a) $h/a = 1.25$ and $\kappa a = 1$, (b) $h/a = 2$ and $\kappa a = 0.5$, (c) $h/a = 2$ and $\kappa a = 1$, and (d) $h/a = 2$ and $\kappa a = 5$.

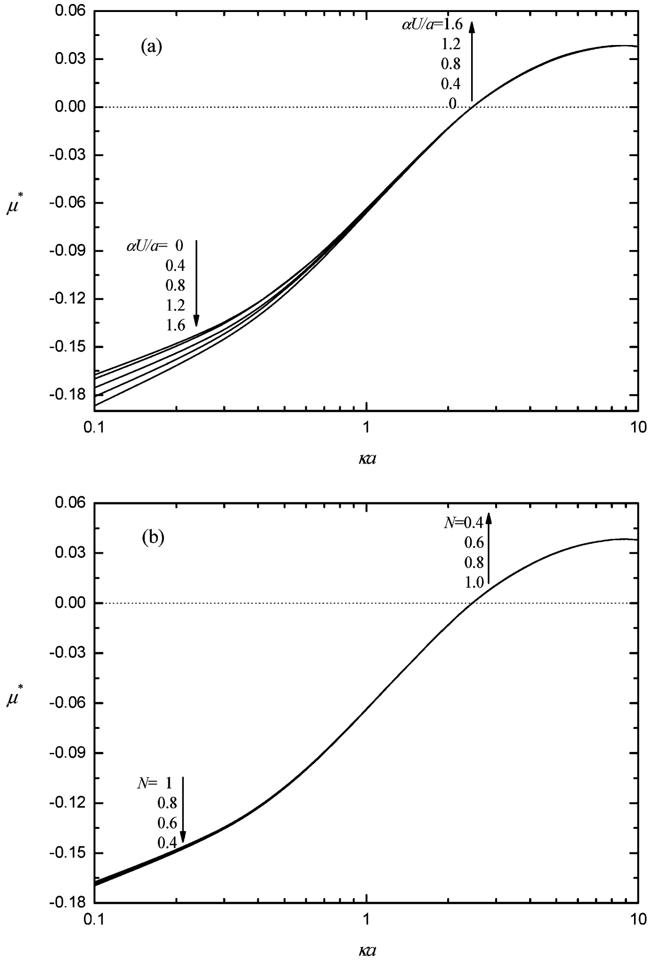


Figure 12. Variation of the scaled mobility μ^* as a function of κa for various ($\alpha U/a$)'s at $n = 0.8$ (a) and that for various N's at $\alpha U/a = 0.2$ (b). Other parameters used are $\zeta_p^* = 0$, $\zeta_w{}^* = 1$, and $h/a = 1.25$.

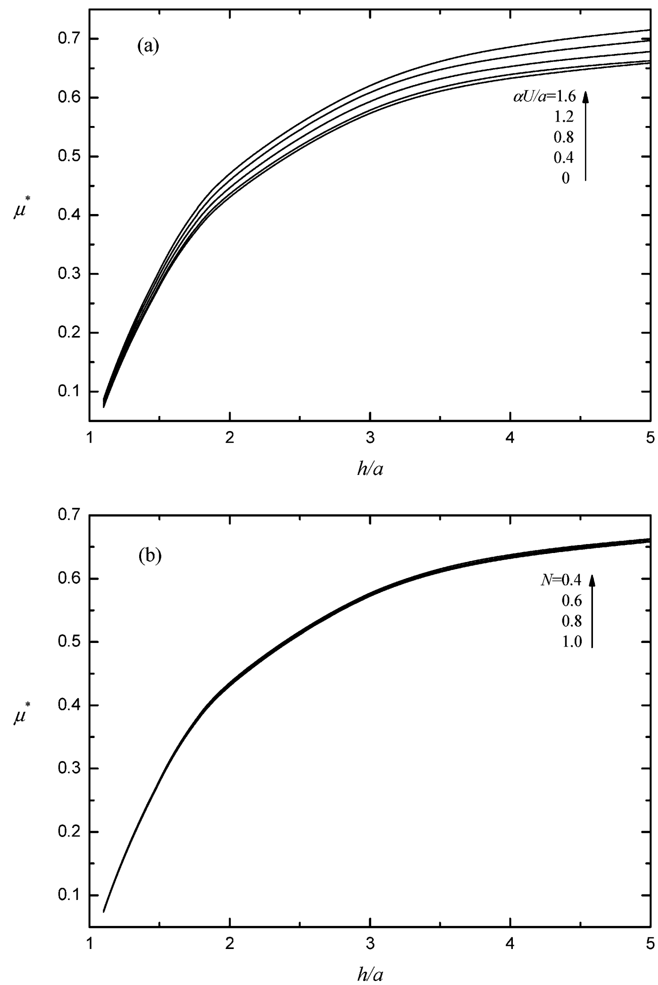


Figure 13. Variation of the scaled mobility μ^* as a function of (h/a) for various ($\alpha U/a$)'s at $N = 0.8$ (a) and that for various N's at $\alpha U/a = 0.2$ (b). Other parameters used are $\zeta_p^* = 1$, $\zeta_w{}^* = 1$, and $\kappa a = 1$.

0.29% for $h/a = 1.1$, 1.25, and 2, respectively, and if $h/a = 1.25$, *PD* are 4.78, 2.54, 1.25, and 0.81% for $\kappa a = 0.1$, 0.5, 1, and 3, respectively. That is, the effect of shear thinning is significant if the boundary effect is important and/or the double layer is sufficient thick. The former is consistent with the results shown in Figures 3 and 4, where a sphere is positively charged and a disk is uncharged, and can be explained by the same reasoning. The later is inconsistent with the results shown in Figures 3 and 4 and has not been reported previously. The effect of shear thinning can also be justified by examining the contours of η^* shown in Figure 8, where the fluid viscosity near a sphere is small when κa is small. This is because if the double layer is thick, the amount of negative charge induced on the sphere surface is large, so is the electric force acting on the sphere. Also, the pressure is small at the same time, yielding a large $|\mu^*|$ and a large variation in the velocity gradient. Figure 8b,c indicates that if κa is not too large, the minimal viscosity occurs on the sphere surface, which is consistent with the results reported in the literature.[31,33,34] However, if κa is sufficiently large, the minimal viscosity becomes large and occurs on the disk surface, as illustrated in Figure 8d. This is because if the double layer of a disk is sufficiently thin, the negative electric force and the positive pressure acting on a sphere is unimportant, leading to a small μ^* and a small variation in the velocity gradient near the sphere. Since only a limited electroosmotic flow is generated in the double layer of the disk, the minimal viscosity occurs on the disk surface.

3.3. Positively Charged Sphere and Positively Charged Disk. Let us consider next the case in which both a sphere and a disk are positively charged. The influences of the scaled separation distance between a sphere and a disk (h/a) and the thickness of double layer κa on the scaled mobility of a sphere μ^* are illustrated in Figure 9a,b, respectively. For comparison, the results for the corresponding Newtonian fluid are also presented in this figure. For the present case, μ^* is positive because the net electric force acting on a sphere is always positive. Figure 9a reveals that μ^* declines monotonically with the decrease in h/a, implying that, if the surface potential of a sphere is the same as that of a disk, the mobility of the former is dominated by the viscous effect arising from the nonslip condition on the latter, as is in the case in which a sphere is positively charged and a disk is uncharged. However, the influence of the disk on μ^* is significant only for $\kappa(h - a) < 3$, where double layer distortion is important. As can be seen in Figure 9b, for both Newtonian and Carreau fluids, μ^* increases monotonically with the increase in κa. However, μ^* is more sensitive to the variation of κa for a Carreau fluid than that for the corresponding Newtonian fluid. For fixed κa and h/a, μ^*(Carreau) > μ^*(Newtonian), and [μ^*(Carreau) − μ^*(Newtonian)] increases with the increase in κa, which is consistent with the result of Lee et al.[29] for the case of a charged sphere at the center of an uncharged spherical cavity. Let $\Delta = \{[\mu^*(\text{Carreau}) - \mu^*(\text{Newtonian})]_{\text{sphere and disk positively charged}} - [\mu^*(\text{Carreau}) - \mu^*(\text{Newtonian})]_{\text{sphere positively charged, disk uncharged}}\}$. Then, according to

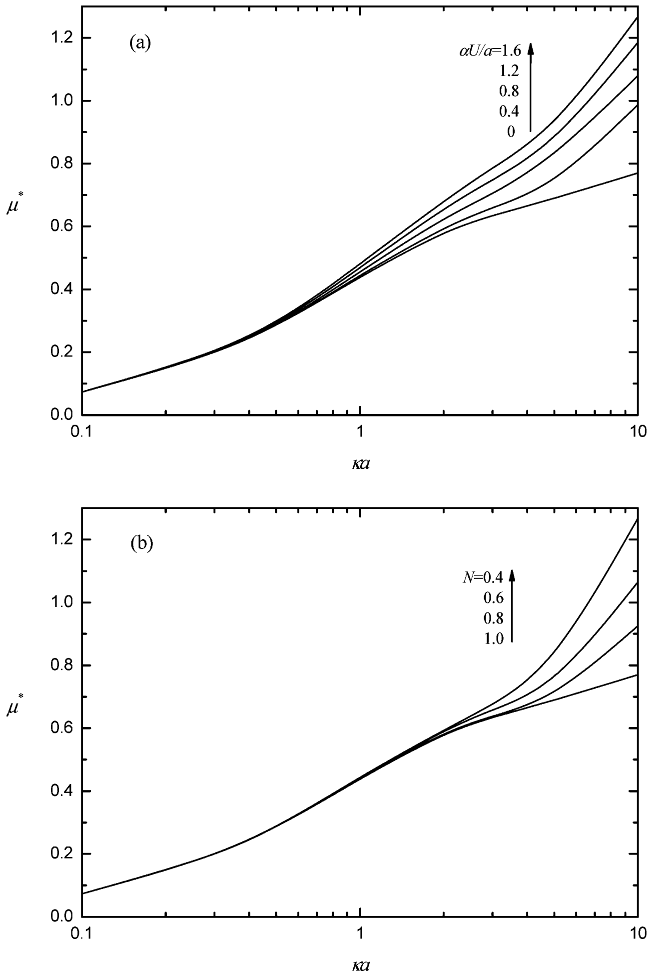


Figure 14. Variation of the scaled mobility μ^* as a function of κa for various ($\alpha U/a$)'s at $N = 0.8$ (a) and that for various N's at $\alpha U/a = 0.2$ (b). Other parameters used are $\zeta_p^* = 1$, $\zeta_w^* = 1$, and $h/a = 2$.

Figure 10, if κa is small, $\Delta < 0$. Also, Δ has a local maximum as κa varies for $\kappa(h - a)$ in the range [2, 4] and vanishes when κa is sufficiently large. These phenomena have not been reported in the literature and can be explained by comparing the behavior of μ^* as κa varies in a Carreau fluid with that in the corresponding Newtonian fluid or by examining the contours of η^* shown in Figure 11. Figure 9b also suggests that μ^* is more sensitive to the variation in κa when both a sphere and a disk are positively charged than when a sphere is positively charged and a disk is uncharged. We conclude that the presence of a charged disk can be advantageous to the movement of an identically charged sphere in that its mobility can have a local maximum and is larger than that for the case when a disk is uncharged. However, it becomes disadvantageous if κa is sufficiently small, that is, μ^*(charged disk) < μ^*(uncharged disk). Figure 9b suggests $\mu^* \to 0$ as $\kappa a \to 0$. A similar result was also obtained in previous studies[16,19,20] and was proven by Zydney[16] for the case of a charged sphere in a charged spherical cavity. The phenomenon are that (i) μ^* is more sensitive to the variation in κa when both a sphere and a disk are positively charged than when a sphere is positively charged and a disk is uncharged, (ii) the presence of a charged disk can be advantageous to the movement of an identically charged sphere in that its mobility can have a local maximum and is larger than that for the case when a disk is uncharged, and (iii) $\mu^* \to 0$ as $\kappa a \to 0$ arise from the behaviors of the driving forces acting on a sphere as $\kappa(h - a)$ varies. For a fixed h/a, the electric force acting on a sphere is positive (so is μ^*) and increases with the increase in κa. Also, the hydrostatic pressure acting on a sphere arising from the osmotic pressure,[12] which is in the same direction as that of the electric force, increases first and then decreases as κa increases for $\kappa(h - a)$ in the range [2, 4]. In the limit $\kappa a \to \infty$, a sphere feels no pressure. If κa is small, the amount of negative charge induced on the sphere surface due to the presence of the positively charged disk, which has the effect of decreasing the net electric force acting on the sphere, is large. Also, the pressure hydrostatic force is small at the same time. Therefore, μ^*(both sphere and disk positively charged) < μ^*-(sphere positively charged, disk uncharged). However, the situation becomes reversed if κa is large, that is, μ^*(both sphere and disk positively charged) > μ^*(sphere positively charged, disk uncharged). Since the larger the κa, the greater the net driving force (electric+pressure) acting on a sphere, the larger the μ^*, the greater the shear rate, and the more significant the effect of shear thinning, we conclude that, if κa is large, the effect of shear thinning for the case when both a sphere and a disk are positively charged is more important than that for the case when a sphere is positively charged and a disk is uncharged, and the reverse is true if κa is small. Note that a further increase in κa for $\kappa(h - a)$ in the range [2, 4] leads to a decline in the hydrostatic pressure, which vanishes as $\kappa a \to \infty$. That is, for both Newtonian and Carreau fluids, μ^*(both sphere and disk positively charged) is close to μ^*(sphere positively charged, disk uncharged), and they become the same when κa is sufficiently large. This explains that, why Δ has a local maximum as κa increases for $\kappa(h - a)$ in the range [2, 4], it vanishes when κa is sufficiently large. It is interesting to note that the value of κa at which Δ becomes positive increases with the decrease in *h/a*, as shown in Figure 10. This is because, if κa is small, the smaller the *h/a*, the more significant the influence of the negative charge induced on the sphere surface. Figure 11 reveals that the minimum viscosity of the system under consideration occurs on the sphere surface, and this minimum decreases with the decrease in *h/a* and/or the increase in κa. Similar behavior is also observed in Figure 4. A comparison between Figure 4b−d and Figure 11b−d reveals that, as mentioned previously, if κa is large, the effect of shear thinning in which both a sphere and a disk are positively charged is more important than that in which a sphere is positively charged and a disk uncharged, but the reverse is true if κa is small.

Figures 12 through 14 illustrate the influence of the nature of a Carreau fluid on the scaled mobility of a sphere μ^* for the case in which a disk is positively charged. In general, the larger the $\alpha U/a$ and/or the smaller the N, that is, the more significant the effect of shear thinning, the larger the $|\mu^*|$. These are consistent with those of the previous studies where a boundary is uncharged.[29−34] It is interesting to note that, if κa is small, μ^* is more sensitive to the variation in $\alpha U/a$ than that in N. This phenomenon is similar to the case in which a sphere is at the center of a spherical cavity,[29] even if the cavity is uncharged.

4. Conclusions

The boundary effect on the electrophoretic behavior of a particle in a non-Newtonian fluid is studied by considering the electrophoresis of a sphere in a Carreau fluid normal to a large disk under the conditions of low surface potential and weak applied electric field. We show that the mobility of a sphere is influenced by three key factors: the separation distance between the sphere and the disk, the thickness of double layer, and the nature of a Carreau fluid. Also, the shear-thinning effect of a Carreau fluid correlates positively with the boundary effect. If a sphere is positively charged and a disk is uncharged, the presence of the latter has the effect of retarding the movement of the former. Also, the shear-thinning effect becomes significant if a double layer is thin. On the other hand, although the presence of a charged disk does not cause an electroosmotic flow parallel to the applied field, it influences appreciably the movement of a sphere through inducing charge on its surface and establishing an osmotic pressure field. If a sphere is uncharged and a disk is positively charged, the charge induced on the sphere surface and the osmotic pressure field established by the disk can yield a local maximum in the mobility of the sphere or even change its sign. Also, the shear-thinning effect is pronounced if the double layer is thick, which has not been reported in the literature. If both a sphere and a disk are positively charged, the mobility of the sphere is always positive. For a fixed separation distance between a sphere and a disk, if the double layer is thick, the mobility in the present case is larger than that in the case in which a sphere is positively charged and a disk is uncharged, but the reverse is true if the double layer is thin. The mobility of a sphere is more sensitive to the variation in the thickness of the double layer in the case of a Carreau fluid than in the case of the corresponding Newtonian fluid. If the double layer is thick, the shear-thinning effect in the present case is less important than that in the case in which a sphere is positively charged and a disk is uncharged.

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