Electro-osmotic mobility of non-Newtonian fluids

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(Received 11 January 2011; accepted 8 March 2011; published online 23 March 2011)

Electrokinetically driven microfluidic devices are usually used to analyze and process biofluids which can be classified as non-Newtonian fluids. Conventional electrokinetic theories resulting from Newtonian hydrodynamics then fail to describe the behaviors of these fluids. In this study, a theoretical analysis of electro-osmotic mobility of non-Newtonian fluids is reported. The general Cauchy momentum equation is simplified by incorporation of the Gouy–Chapman solution to the Poisson–Boltzmann equation and the Carreau fluid constitutive model. Then a non-linear ordinary differential equation governing the electro-osmotic velocity of Carreau fluids is obtained and solved numerically. The effects of the Weissenberg number (Wi), the surface zeta potential ($\bar{\psi}_s$), the power-law exponent (n), and the transitional parameter (β) on electro-osmotic mobility are examined. It is shown that the results presented in this study for the electro-osmotic mobility of Carreau fluids are quite general so that the electro-osmotic mobility for the Newtonian fluids and the power-law fluids can be obtained as two limiting cases.

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I. INTRODUCTION

Microfluidics has the potential to enable biochemical analyses on single microchips. One of the most preferred ways for pumping samples in microfluidics is electro-osmosis.^{1–3} In electro-osmosis, an electric field generates a net force on the electric double layer (EDL) near a charged channel wall, where the excess counterions are localized within the EDL by Coulombic interactions. As the fluid in the EDL moves toward the oppositely charged electrode, it triggers the motion of bulk liquid in the channel. As a result, the velocity profile is essentially flat across the channel if the channel dimension is large. This type of flow is ideal for separations based on the charge-to-size ratio of the molecular species in a buffer solution,² since broadening of the separated bands of differing species occurs only by diffusion, not as a result of the differences in flow velocity across the channel when the flow is driven by pressure gradient.

Conventional electrokinetic phenomena, such as electro-osmosis or electrophoresis, result from the interaction between electrostatics and Newtonian hydrodynamics. However, biochemical analytical systems in reality are frequently used to process biofluids, such as saliva solutions, DNA solutions, and blood samples, which cannot be treated as Newtonian fluids. Thus, the Navier–Stokes equation fails to describe the behaviors of these biofluids, and the more general Cauchy momentum equation with a proper constitutive equation must be used instead. A number of constitutive equations have been established to relate the dynamic viscosity of non-Newtonian fluids to the shear rate, such as the power-law model,⁴ Carreau model,⁵ Moldflow first-order model,⁶ and Bingham model.⁷ Electrokinetic phenomena involving non-Newtonian fluids were already shown to behave differently from their Newtonian counterparts. Gur and Ravina⁸ and Lyklema⁹ reported rheological electrokinetic phenomena due to the viscoelectric effect. In their models, the viscosity of fluids is a quadratic function of the local electric field strength. In Refs.

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10–15, Hsu's group investigated the electrophoresis of particles with different shapes in non-Newtonian Carreau fluids confined by different geometries. Their results showed that the electro-phoretic mobility in shear thinning fluids is higher than that in Newtonian fluids. Electro-osmotic flows of non-Newtonian fluids in the microchannels were also addressed theoretically.^{16–23} Recently, experimental investigations^{24,25} were performed for the electro-osmotic flow of a typical polymer solution in microchannels. In addition, the generalized Helmholtz–Smoluchowski velocity derived in Ref. 20 was confirmed.

For electro-osmotic flows in a microchannel, the velocity in the bulk (outside the EDL) is uniform and the corresponding velocity profile exhibits a plug-like shape. Electro-osmotic mobility denotes the bulk velocity of the liquid driven by unit electric field strength. For the Newtonian fluids, it is well known that the electro-osmotic mobility is expressed as $\mu = -\varepsilon\psi_s/\eta_0$ (ε and η_0 are electric permittivity and dynamic viscosity of the fluid, ψ_s is the surface zeta potential of the charged wall). The effects of rheological properties of the fluids on the Helmholtz–Smoluchowski velocity were reported.^{20,22} However, the constitutive model for non-Newtonian fluids adopted in these studies is only an extreme case of the more general non-Newtonian Carreau fluid model. In view of no general treatment of electro-osmotic mobility of non-Newtonian fluids, the present study focuses on the electro-osmotic motion near an arbitrarily charged surface with a non-Newtonian fluid whose rheological behavior can be described by the Carreau model. In comparison with the Newtonian case, the Carreau constitutive model involves five additional parameters, and thus can describe the rheology of a wide range of non-Newtonian fluids. At first, the equation governing the electro-osmotic mobility of Carreau fluids is formulated by using the general Cauchy momentum equation. Subsequently, the numerical solution for the formulated problem is presented and validated. Finally, the effects of rheological properties of Carreau fluids on electro-osmotic mobility are examined.

II. PROBLEM FORMULATION

We consider the electro-osmosis over a charged surface with a zeta potential, ψ_s . The y axis is perpendicular to the charged surface and the x axis is parallel with the surface. The semi-infinite space above the surface is occupied by a liquid solution with an electric permittivity, ε . It is assumed that the liquid solution is a typical non-Newtonian fluid whose behavior can be described by the well-known Carreau model. When an external electric field E_0 is imposed along the x -axis direction, the fluid inside the EDL sets in motion due to electro-osmosis. The no slip boundary condition causes the velocity to increase from zero on the charged surface to a maximum value (termed the Helmholtz–Smoluchowski velocity) at the outer edge of the EDL, beyond which there is no variation in the velocity. The ratio of the Helmholtz–Smoluchowski velocity to the external electric field strength is defined as the electro-osmotic mobility of fluids.

A. Electric field in the EDL

Introducing the dimensionless groups, $\bar{y} = y/\lambda_D$ and $\bar{\psi} = z_v e \psi / (k_B T)$, one can write the Poisson–Boltzmann equation governing the electric potential distribution in the EDL region in nondimensionalized form as [assuming a symmetric electrolyte ($z_v : z_v$)]

$$\frac{d^2 \bar{\psi}}{d\bar{y}^2} = \sinh(\bar{\psi}). \quad (1)$$

Equation (1) is subject to the following boundary conditions:

$$\bar{\psi}|_{\bar{y}=0} = \bar{\psi}_s \quad \left. \frac{d\bar{\psi}}{d\bar{y}} \right|_{\bar{y} \rightarrow \infty} = 0, \quad (2)$$

where the dimensionless wall zeta potential is given by $\bar{\psi}_s = z_v e \psi_s / (k_B T)$, the Debye length λ_D is defined as $\lambda_D = [\varepsilon k_B T / (2 z_v^2 e^2 n_\infty)]^{1/2}$ with e being the fundamental charge, k_B being the Boltzmann

constant, T being the absolute temperature, and n_∞ and z_v being the bulk number concentration and the valence of ions, respectively.

The solution to Eq. (1) is the famous Gouy–Chapman solution^{26,27}

$$\bar{\psi}(\bar{y}) = 2 \ln \left[\frac{1 + e^{-\bar{y}} \tanh(\bar{\psi}_s/4)}{1 - e^{-\bar{y}} \tanh(\bar{\psi}_s/4)} \right]. \quad (3)$$

The net charge density in the EDL region is related to the EDL potential by using the widely assumed Boltzmann distribution,

$$\rho_e = -2z_v e n_\infty \sinh(\bar{\psi}). \quad (4)$$

B. Electro-osmotic flow of Carreau fluids

In the present work, non-Newtonian effects are considered for fluids obeying the Carreau constitutive relationship proposed by Carreau and others.^{10,28–30} For the Carreau fluid model, the dynamic viscosity η is given by

$$\eta(\Gamma) = \eta_0 [1 + (2\lambda\Gamma)^\beta]^{(n-1)/\beta}, \quad (5)$$

where η_0 is the zero-shear-rate viscosity, λ is the relaxation time constant, n is the power-law exponent, Γ is the magnitude of the rate of strain tensor, and β is a dimensionless transitional parameter that adjusts the breadth of the transition region between the zero-shear-rate viscosity and the power-law regions. When $n \rightarrow 1$ or $\lambda \rightarrow 0$, it reduces to a Newtonian fluid, and it becomes a power-law fluid if λ is sufficiently large. For $n < 1$, the fluid shows shear thinning behavior and the viscosity decreases with the increase of shear rate; for $n > 1$, the fluid shows shear thickening behavior and the viscosity decreases with the decrease of shear rate. Contrary to the commonly used power-law model which predicts an infinite viscosity at the limit of zero shear rate when $n < 1$, the Carreau model leads to a smooth transition to a constant viscosity at the limit of zero shear rate. Carreau fluid model can well characterize the rheology of various polymeric solutions, such as 1% methylcellulose Tylose in glycerol solution and 0.3% hydroxyethyl-cellulose Natrosol HXH in glycerol solution,³¹ and pure poly(ethylene oxide).³² These polymers are widely used in capillary electrophoresis to improve the selectivity and resolution in the separation of proteins³³ and DNAs.³⁴

Electro-osmotic flow over the charged solid surface is unidirectional, and thus the corresponding velocity distribution is only y dependent, i.e., $u(y)$. Taking into account the dynamic viscosity given by Eq. (5), the simplified steady Cauchy momentum equation with an electrostatic body force reads

$$\frac{d}{dy} \left[\eta_0 \left[1 + \left(\lambda \frac{du}{dy} \right)^\beta \right]^{(n-1)/\beta} \frac{du}{dy} \right] + \rho_e E_0 = 0. \quad (6)$$

Substituting Eqs. (1) and (4) into Eq. (6), one can obtain

$$\frac{d}{dy} \left[\eta_0 \left[1 + \left(\lambda \frac{du}{dy} \right)^\beta \right]^{(n-1)/\beta} \frac{du}{dy} \right] - \varepsilon \frac{d^2 \psi}{dy^2} E_0 = 0. \quad (7)$$

Two additional nondimensional parameters are introduced as follows:

$$\bar{u} = \frac{u}{u_{s0}} \quad \text{Wi} = \frac{\varepsilon E_0 k_B T \lambda}{\eta_0 z_v e \lambda_D}, \quad (8)$$

where u_{s0} denotes the Helmholtz–Smoluchowski velocity for the Newtonian fluids with dynamic viscosity of η_0 , and is given by

$$u_{s0} = -\frac{\varepsilon \psi_s E_0}{\eta_0}. \quad (9)$$

The electro-osmotic mobility is defined as the ratio of the Helmholtz–Smoluchowski velocity to the applied electric field strength, namely, $\mu = u_s/E_0$ ($u_s = u|_{y \rightarrow \infty}$). Then the dimensionless velocity $\bar{u}_s$ ($\bar{u}_s = \bar{u}|_{\bar{y} \rightarrow \infty}$) can be treated as the ratio of the electro-osmotic mobility of non-Newtonian Carreau fluids to that of Newtonian fluids. Wi stands for the dimensionless relaxation time constant and is called the Weissenberg number.⁵

With dimensionless parameters given in Eq. (8), Eq. (7) can be rewritten in dimensionless form as

$$\frac{d}{d\bar{y}} \left\{ \left[1 + \left(-Wi \bar{\psi}_s \frac{d\bar{u}}{d\bar{y}} \right)^\beta \right]^{(n-1)/\beta} \frac{d\bar{u}}{d\bar{y}} \right\} + \frac{1}{\bar{\psi}_s} \frac{d^2 \bar{\psi}}{d\bar{y}^2} = 0, \quad (10)$$

which is subject to the following boundary conditions:

$$\bar{u}|_{\bar{y}=0} = 0 \quad \left. \frac{d\bar{u}}{d\bar{y}} \right|_{\bar{y} \rightarrow \infty} = 0. \quad (11)$$

One should note that the far-field boundary conditions expressed in Eqs. (2) and (11) for electric potential and velocity fields make the results presented in this study only applicable to electro-osmosis in microchannels with thin EDLs.

Integrating both sides of Eq. (10) from $\bar{y}$ to infinity and taking into account the boundary conditions at the infinity for both velocity and electric potential distributions, one can obtain

$$\left[1 + \left(-Wi \bar{\psi}_s \frac{d\bar{u}}{d\bar{y}} \right)^\beta \right]^{(n-1)/\beta} \frac{d\bar{u}}{d\bar{y}} - \frac{1}{\bar{\psi}_s} \frac{2 \sinh\left(\frac{\bar{\psi}}{2}\right)}{e^{\bar{y}} \cosh^2\left(\frac{\bar{\psi}_s}{4}\right) - e^{-\bar{y}} \sinh^2\left(\frac{\bar{\psi}_s}{4}\right)} = 0, \quad (12)$$

and the boundary condition for Eq. (12) is simply reduced to the first equation in Eq. (11).

An examination of Eq. (12) indicates that four dimensionless parameters including Wi , $\bar{\psi}_s$, β , and n are main influencing factors for the electro-osmotic velocity distribution. Equation (12) is a nonlinear ordinary differential equation which is to be solved numerically.

However, for two limiting cases, exact solutions can be obtained for Eq. (12). For the Newtonian fluids ($n=1$), it is straightforward to obtain an exact solution for Eq. (12) as

$$\bar{u} = 1 - \frac{\bar{\psi}(\bar{y})}{\bar{\psi}_s}, \quad (13)$$

and it is quite obvious that $\bar{u}_s = \bar{u}|_{\bar{y} \rightarrow \infty} = 1$. When Wi is sufficiently large, i.e., $(-Wi \bar{\psi}_s d\bar{u}/d\bar{y})^\beta \gg 1$, the Carreau fluid model reduces to the power-law model, and Eq. (12) can be approximated as

$$(-Wi \bar{\psi}_s)^{n-1} \left(\frac{d\bar{u}}{d\bar{y}} \right)^n - \frac{1}{\bar{\psi}_s} \frac{2 \sinh\left(\frac{\bar{\psi}}{2}\right)}{e^{\bar{y}} \cosh^2\left(\frac{\bar{\psi}_s}{4}\right) - e^{-\bar{y}} \sinh^2\left(\frac{\bar{\psi}_s}{4}\right)} = 0. \quad (14)$$

The exact solution for Eq. (14) can be found as

$$\bar{u} = \frac{n \left[-2 \sinh\left(\frac{\bar{\psi}_s}{2}\right) \right]^{1/n}}{\text{Wi}^{(n-1)/n}(-\bar{\psi}_s)} \left[H\left(\bar{y}, \frac{\bar{\psi}_s}{4}, n\right) - H\left(0, \frac{\bar{\psi}_s}{4}, n\right) \right], \quad (15)$$

where the function $H(\alpha, \beta, \gamma)$ is defined by the following integral:

$$\begin{aligned} H(\alpha, \beta, \gamma) &= \frac{1}{\gamma} \int \left[\frac{1}{\cosh^2(\beta) e^\alpha - e^{-\alpha} \sinh^2(\beta)} \right]^{1/\gamma} d\alpha \\ &= \left[-\frac{e^\alpha}{\sinh^2(\beta)} \right]^{1/\gamma} {}_2F_1 \left[\frac{1}{2\gamma}, \frac{1}{\gamma}; \frac{1+2\gamma}{2\gamma}; e^{2\alpha} \coth^2(\beta) \right]. \end{aligned} \quad (16)$$

In Eq. (16), ${}_2F_1[a, b; c; z]$ is the Gauss' hypergeometric function and can be defined by the following infinite series:³⁵

$${}_2F_1[a, b; c; z] = \sum_{k=0}^{\infty} \frac{(a)_k (b)_k}{(c)_k k!} z^k, \quad (17)$$

where $(a)_k$ represents the Pochhammer symbol, and can be expanded as

$$(a)_k = a(a+1) \cdots (a+k-1) \quad \text{and} \quad (a)_0 = 1. \quad (18)$$

The dimensionless electro-osmotic mobility for the power-law fluids can be obtained as

$$\bar{u}_s = \bar{u}|_{\bar{y} \rightarrow \infty} = \frac{n \left[-2 \sinh\left(\frac{\bar{\psi}_s}{2}\right) \right]^{1/n}}{\text{Wi}^{(n-1)/n}(-\bar{\psi}_s)} {}_2F_1 \left[\frac{1}{2n}, \frac{1}{n}; \frac{1+2n}{2n}; \tanh^2\left(\frac{\bar{\psi}_s}{4}\right) \right] \left[\text{sech}^2\left(\frac{\bar{\psi}_s}{4}\right) \right]^{1/n}. \quad (19)$$

III. RESULTS AND DISCUSSION

In the present analysis, the electro-osmotic flow field of the Carreau fluids is obtained by numerically solving Eq. (12) in the finite element analysis package COMSOL MULTIPHYSICS 3.4. Then the electro-osmotic mobility $\bar{u}_s$ is sought from the electro-osmotic velocity at the outer edge of EDL where the velocity does not vary with the vertical position anymore and reaches the so-called Helmholtz–Smoluchowski velocity. In order to check the validity and accuracy of the present numerical model, we compared the numerical solutions with the exact solutions for two limiting cases (Newtonian fluids and power-law fluids) and excellent agreements can be seen in Figs. 1–3.

The effects of four parameters including Wi , $\bar{\psi}_s$, n , and β on the electro-osmotic mobility of Carreau fluids are shown in Figs. 1–3. The two limiting cases, (i) the electro-osmotic mobility of Newtonian fluids and (ii) the electro-osmotic mobility of power-law fluids, are also included for comparisons. Figure 1 presents the variation of dimensionless electro-osmotic mobility with the surface zeta potential for different values of Wi . It is clearly seen that as long as Wi is not zero, the electro-osmotic mobility increases with the increase of the magnitude of surface zeta potential. This is expected because a higher zeta potential leads to a stronger electrostatic body force in the EDL and thus a higher electro-osmotic mobility. In addition, for a larger Wi , the electro-osmotic mobility more obviously depends on the surface zeta potential. While as $\text{Wi} \rightarrow 0$ (i.e., Newtonian fluids), the dimensionless electro-osmotic mobility becomes independent of surface zeta potential and acquires a constant value of unity. For a large value of Wi , for example, $\text{Wi}=10$, the electro-osmotic mobility predicted by the Carreau fluid model becomes very close to that predicted by the power-law fluid model, which is also featured in the following figures. The effect of the power-law exponent on electro-osmotic mobility is shown in Fig. 2. A higher value of power-law exponent indicates a more viscous fluid, thus giving rise to a lower electro-osmotic mobility. For Newtonian

FIG. 1. Variation of electro-osmotic mobility with the surface zeta potential for three different values of Wi ($Wi=0.1, 1.0, 10.0$) when $n=0.8$ and $\beta=2$. There are also two limiting cases included in the figure: (i) the Newtonian fluid model with the electro-osmotic mobility being unity and (ii) the power-law fluid model with the electro-osmotic mobility computed from Eq. (19).

fluids ($n=1$), the electro-osmotic mobility is independent of Wi . However, for fluids with nonunity power-law exponent, the dependence of electro-osmotic mobility on Wi shows opposite trends for shear thinning fluids and shear thickening fluids. For shear thinning fluids ($n < 1$), the electro-osmotic mobility increases with the increase of Wi , and its magnitude is larger than that for

FIG. 2. Variation of electro-osmotic mobility with fluid power-law exponent n for three different values of Wi ($Wi=0.1, 1.0, 10.0$) when $\beta=2$ and $\bar{\psi}_s=-5$. There are also two limiting cases included in the figure: (i) the Newtonian fluid model with the electro-osmotic mobility being unity and (ii) the power-law fluid model with the electro-osmotic mobility computed from Eq. (19).

FIG. 3. Variation of electro-osmotic mobility with β for three different values of Wi ($Wi=0.1, 1.0, 10.0$) when $n=0.8$ and $\bar{\psi}_s=-5$. There are also two limiting cases included in the figure: (i) the Newtonian fluid model with the electro-osmotic mobility being unity and (ii) the power-law fluid model with the electro-osmotic mobility computed from Eq. (19).

Newtonian fluids. While for shear thickening fluids ($n > 1$) the electro-osmotic mobility increases with the decrease of Wi , its magnitude is smaller than that for Newtonian fluids. Apparently, adjusting Wi is a more effective measure for changing electro-osmotic mobility of shear thinning fluids as compared with shear thickening fluids. It is also worth noting that for shear thinning fluids with large values of Wi (e.g., $Wi=10$), the electro-osmotic mobility can be more than ten times larger than that of Newtonian fluids, which is advantageous in microfluidic applications when large pumping capacity is required.

Figure 3 illustrates the effect of the transitional parameter β on electro-osmotic mobility. Generally, the electro-osmotic mobility decreases with the increase of β . Furthermore, the electro-osmotic mobility becomes less dependent on β as Wi increases. For the Newtonian fluid model ($Wi \rightarrow 0$) and the power-law fluid model ($Wi \rightarrow \infty$), the electro-osmotic mobility is both constant and independent of β . In addition, it can also be concluded from these three figures that the electro-osmotic mobility of Carreau fluids transits from one extreme case of Newtonian fluids to another extreme case of power-law fluids. This makes the present predictions more general and applicable to various kinds of non-Newtonian fluids.

It should be noted that the problem formulated in Sec. II B for the electro-osmotic flow of non-Newtonian fluids can also characterize the electrophoresis of a particle with thin EDL in an unbounded non-Newtonian fluid domain.²² For the latter case, we can change the reference system and imagine that the fluid velocity asymptotically approaches zero far from the charged surface. Then we can obtain a velocity $-\bar{u}_s$ for the charged surface. $-\bar{u}_s$ actually represents the electrophoretic mobility of a particle with thin EDL in an unbounded non-Newtonian fluid domain, and the only change is to replace the zeta potential of the charged wall surface with the zeta potential of the charged particle. Apparently, the above-discussed characteristics of electro-osmotic mobility are also applicable to the electrophoretic mobility of a particle with thin EDL in an unbounded non-Newtonian fluid domain.

IV. CONCLUSIONS

The coupling between the electrodynamics and the non-Newtonian hydrodynamics complicates the electrokinetics of non-Newtonian fluids. The combination of the Cauchy momentum

equation and the Gouy–Chapman solution to the Poisson–Boltzmann equation leads to a nonlinear ordinary differential equation [i.e., Eq. (12)] that governs the electro-osmotic velocity of Carreau fluids. Numerical solutions to Eq. (12) are sought to examine the effects of the Weissenberg number (Wi), the surface zeta potential ($\bar{\psi}_s$), the power-law exponent (n), and the transitional parameter (β) on electro-osmotic mobility of Carreau fluids. The numerical results are validated by the exact solutions for Newtonian fluids and power-law fluids. It is found that the electro-osmotic mobility of Carreau fluids increases with the increase of the magnitude of $\bar{\psi}_s$ and the decreases of n and β . As Wi increases, the electro-osmotic mobility increases for shear thinning fluids but decreases for shear thickening fluids. These characteristics differ from those of the Newtonian electro-osmotic mobility which is independent of aforementioned four parameters. These characteristics for electro-osmotic mobility are of potential importance to the design and control of electrokinetically driven microfluidic devices dealing with non-Newtonian Carreau fluids. For situations where we have Carreau fluids with small n ($n < 1$) and large Wi over a highly charged surface (large $\bar{\psi}_s$), the electro-osmotic mobility can be much larger than its Newtonian counterpart, this feature provides the potential for enhancing electro-osmotic pumping capacity. Another fundamental aspect is that the predicted characteristics for the electro-osmotic mobility of Carreau fluids are also applicable to the electrophoretic mobility of a particle with thin EDL in an unbounded Carreau fluid domain.

ACKNOWLEDGMENTS

Z. C. L. would like to express his gratitude to Nanyang Technological University for the research assistantship.

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