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Force acting on a dielectric particle in a concentration gradient by ionic concentration polarization under an externally applied DC electric field

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Abstract

There is a concentration-polarization (CP) force acting on a particle submerged in an electrolyte solution with a concentration (conductivity) gradient under an externally applied DC electric field. This force originates from the two mechanisms: (i) gradient of electrohydrodynamic pressure around the particle developed by the Coulombic force acting on induced free charges by the concentration polarization, and (ii) dielectric force due to nonuniform electric field induced by the conductivity gradient. A perturbation analysis is performed for the electric field, the concentration field, and the hydrodynamic field, under the assumptions of creeping flow and small concentration gradient. The leading order component of this force acting on a dielectric spherical particle is obtained by integrating the Maxwell and the hydrodynamic stress tensors. The analytical results are validated by comparing the surface pressure and the skin friction to those of a numerical analysis. The CP force is proportional to square of the applied electric field, effective for electrically neutral particles, and always directs towards the region of higher ionic concentration. The magnitude of the CP force is compared to that of the electrophoretic and the conventional dielectrophoretic forces.

Keywords: Concentration-polarization force; Electric field; Concentration gradient; Particle; Electrohydrodynamics; EHD; Dielectrophoresis; Microfluidics; Microchannel

1. Introduction

Many sorts of forces are involved in the dynamics of particles in micro- and nanofluidic environments [1], such as the electrophoretic force, dielectrophoretic force, gravity force, and Stokes force. Utilization of these forces for manipulation and assembly of micro- and nanoparticles leads to evolution of new nanoscale manufacturing technology and new generation of microfluidic devices.

It is highly desirable to understand the effects of electric field on a particle placed in an environment with an electrical-conductivity gradient for many practically important applications. These include the particle assemblies near an (electrochemically reacting) electrode in the electrophoretic deposition process [2], particle–particle and particle–wall interactions via electrical double layers, and chemotaxis of biological cells under a natural [3] or an artificially-generated concentration gradient [4,5].

In this work, we suggest that a particle submerged in an electrolyte solution with a conductivity gradient under an applied electric field will experience a new type of force, as we call it, the concentration-polarization force (CP force), which has never been considered previously. We focus on the case in which the conductivity gradient is generated by the concentration gradient of the electrolyte, rather than by the Joule heating and subsequent temperature gradient [1].

The CP force originates from the two different mechanisms: (i) a gradient of electrohydrodynamic (EHD) pressure around the particle developed by the Coulombic force acting on "induced" free charge by "externally" applied electric field, and (ii) a dielectric force due to nonuniform electric

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field induced by the conductivity gradient. It is known that the concentration gradient may generate the diffusiophoresis of a particle [3,6]. The diffusiophoresis represents the motion of a particle driven by (i) the electrophoretic force exerted by the naturally-developed electric field (only for electrolyte solutions), and/or (ii) the hydrodynamic flow which is originated from the interaction of solute molecules and the particle (e.g., adsorption and osmotic penetration of solute into the particle). Here, as will be discussed in Section 3.2, the "naturally-developed" electric field represents the field induced by the mobility difference of ionic species [7]. That is, in the presence of a concentration gradient, the difference in diffusion speeds between the ionic species develops a local imbalance of densities of cationic and anionic species and hence a corresponding electric field. The CP force is completely different from the diffusiophoretic force since it is not related with any of mobility difference of ionic species and solute–particle interaction.

The present article is organized as follows. Section 2 provides an explanation on how the concentration polarization occurs and the CP force is generated. In Section 3, the problem is formulated by introducing the governing equations and the boundary conditions for the electrostatic, concentration, and hydrodynamic fields. In Section 4, a linear analysis is performed for some limiting conditions, and the analytical results are validated by a numerical analysis. Then, the CP force is obtained by integrating the stress tensors. Finally, in Section 5, the magnitude of the CP force is compared to the electrophoretic and the dielectrophoretic forces, and some characteristics and consequences of the CP force are discussed.

2. Overview

In what follows, we wish to provide a brief overview on how the free charge is generated inside the liquid domain and the CP force is developed, before performing a detailed analysis for quantitative prediction of the force.

2.1. Concentration polarization

Consider a medium filled with a binary symmetric electrolyte liquid (e.g., aqueous NaCl solution) that has a one-dimensional concentration gradient (see Fig. 1). An electric field is applied in parallel to the direction of the concentration gradient. The domain extends to infinity in the direction perpendicular to the electric field (vertical direction in Fig. 1). For the case of Fig. 1, the concentration decreases in the positive x direction, and the electric field directs to the positive x-direction. Hereinafter, for the sake of simplicity, we limit our attention to the 1:1 symmetric electrolyte, and the subscripts "+" and "−" represent the variables associated with the cationic and the anionic species, respectively.

Suppose, all the ions inside the liquid domain are stationary initially, and there is no free charge in the space, i.e., $\rho^f = F(c_+ - c_-) = 0$ and $c_+ = c_- = c$, in which ρ^f denotes the free charge density, F the Faraday constant, and $c_\pm$ the molar concentration for each type of ions. In this imaginary state, the domain can be regarded as a parallel plate capacitor filled with a nonconducting dielectric material, so that the electrical potential (ϕ) has a linear profile and the associated electric field ($\mathbf{E} = -\nabla\phi$) is constant across the layer, i.e., $\phi(x) = \phi_0 - mx$ and $\mathbf{E} = m\mathbf{e}_x$ where m is a constant and $\mathbf{e}_x$ is the unit vector parallel to the x direction.

For the time being, we assume that initially there are no molecular diffusion of species and no bulk hydrodynamic flow. All the ions are suddenly set free to move. We consider the species conservation, immediately after the ions are released, for a control volume taken as a slab region shown in Fig. 1. The control volume is centered at x_0 and has a width of δx. Due to the action of the Coulombic force on each ion, the cationic and anionic species migrate following and against the direction of the electric field, respectively. For a medium obeying the Ohm's law, the molar flux of each species $\mathbf{N}_\pm$ is proportional to the local concentration times the electric-field strength, and can be expressed specifically as $\mathbf{N}_\pm = z_\pm \omega_\pm F c_\pm \mathbf{E}$. Then, the current density (ionic flux) becomes $\mathbf{i} = F(\mathbf{N}_+ - \mathbf{N}_-)$. Here, for a monovalent elec-

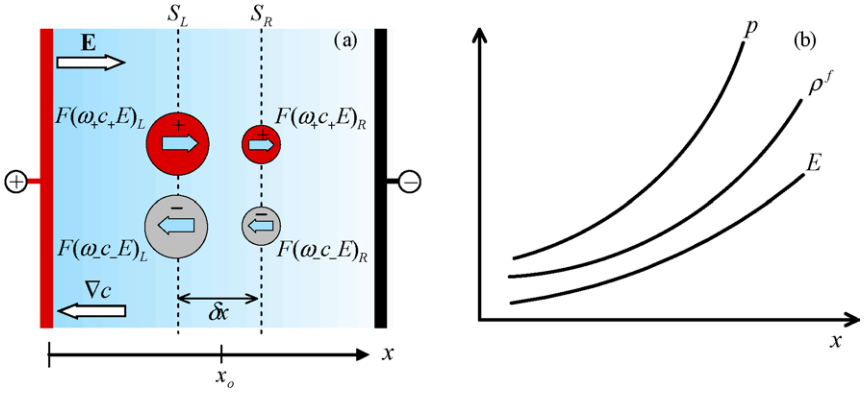

Fig. 1. (a) An electrolyte solution with a concentration gradient under a DC electric field. (b) Schematic diagram for the distribution of the free charge density (ρ^f), electric-field strength (E) and pressure (p).

trolyte, $z_+ = 1$ and $z_- = -1$, and $\omega_\pm$ represents the mobility of each ionic species.

The net ionic flux into the control volume (per unit length in perpendicular to x direction) becomes $q^f = -L^{-1}\oint_S \mathbf{i}\cdot\mathbf{n}\,\mathrm{d}S$, in which $\mathbf{n}$ is the outward unit normal vector at the control surface S, and L is the height of the control surface. Then, it follows

$$q^f = -\omega_+ F^2\big[(c_+E)_R - (c_+E)_L\big] - \omega_- F^2\big[(c_-E)_R - (c_-E)_L\big],$$

where $F^2(\omega_\pm c_\pm E)_R$ and $F^2(\omega_\pm c_\pm E)_L$ represent the ionic fluxes at right-hand side (RHS) and left-hand side (LHS) boundaries of the control volume, respectively (see Fig. 1). By using the Taylor series expansion, we get

$$q^f \cong -F^2\left[\frac{\mathrm{d}\omega_+ c_+ E}{\mathrm{d}x} + \frac{\mathrm{d}\omega_- c_- E}{\mathrm{d}x}\right]_{x=x_0}\delta x.$$

At the beginning, there is no free charge, so that $c_+ = c_- = c$, and we define the electrical conductivity of liquid as

$$\sigma_f = F^2(\omega_+ + \omega_-)c. \tag{1}$$

Thus, the net ionic flux into the control volume q^f can be rewritten as

$$q^f \cong -\frac{\mathrm{d}\sigma_f E}{\mathrm{d}x}\bigg|_{x=x_0}\delta x.$$

Initially, q^f is positive because it is assumed $E = m > 0$ and $\partial\sigma_f/\partial x < 0$ (due to the concentration gradient). This means that the positive ions begin to accumulate at the control volume and the free charges are induced; that is, the "concentration polarization" occurs.

The induced free charges in turn modify the electric-field distribution. According to the Gauss law, $\oint_S \varepsilon_f \mathbf{E}\cdot\mathbf{n}\,\mathrm{d}S = Q^f L$, where ε_f is the electrical permittivity of liquid, and Q^f is the total charge per unit length of the control surface in vertical direction. The Gauss law can be written for the one-dimensional slab region as

$$E_R - E_L = \frac{Q^f}{\varepsilon}.$$

Since $q^f > 0$ and $Q^f > 0$, the induced free charge makes E_R greater than E_L (they were identical without the concentration polarization). This limits the continuous accumulation of the positive ions, by balancing the fluxes of ionic species through the boundaries. The accumulation of positive ions stops when there is no net flux of ionic species across the boundary, i.e.,

$$\frac{\mathrm{d}(\sigma_f E)}{\mathrm{d}x} = 0. \tag{2}$$

Equation (2) is just the one-dimensional version of the electric-field equation for a conducting medium obeying the Ohm's law. This also corresponds to the current conservation equation $\nabla\cdot\mathbf{i} = 0$ where $\mathbf{i} = \sigma_f\mathbf{E}$ is the "approximated" current density under the assumption $c_+ = c_-$. The equilibrium condition for the ion transport is established only when the local electric field, which was initially uniform throughout the medium, is changed to satisfy $E \propto 1/\sigma_f \propto 1/c$.

In fact, the polarization of the liquid medium means $c_+ \neq c_-$. In the Poisson equation written as

$$\nabla^2\phi = -\frac{F}{\varepsilon_f}(c_+ - c_-), \tag{3}$$

the value of F/ε is as great as about 10^{14}. In practice, therefore, only a very small difference between c_+ and c_- can induce an enormous change of electric field. This "induced" electric field counteracts against the further deviation from the electroneutrality. Thus, only a small fraction of the ions are involved in the polarization of the medium, and the solution remains "effectively" neutral in considering the species transport. This is the basis of the so-called quasi-electroneutrality assumption [7–10]. On the other hand, we have two equations for the electric field of Eqs. (2) and (3). Equation (2) is in fact an approximation that is derived on the basis of the quasi-electroneutrality assumption of $c_+ \cong c_- = c$. It is a common practice to solve the current conservation equation, such as Eq. (2), to obtain the electric field, under the electroneutrality assumption. The Poisson equation, Eq. (3), is used only for "after-the-fact" evaluation of the free charge density [7–10].

In the above, it is shown that the "adjustment" process of electric field in order to satisfy both the current conservation and the Ohm's law should accompany the generation of free charges. The magnitude of the free charge is obtained by using the Poisson equation of $\nabla^2\phi = -\rho^f/\varepsilon_f$. That is, by combining the generalized version of Eq. (2), $\nabla\cdot(\sigma_f\nabla\phi) = 0$, and the Poisson equation, the free charge density can be obtained approximately as

$$\rho^f = -\varepsilon_f\nabla^2\phi \cong -\frac{\varepsilon_f\nabla\sigma_f\cdot\mathbf{E}}{\sigma_f}. \tag{4}$$

In Fig. 1, the directions of electric field and the concentration gradient are opposed, i.e., $\nabla\sigma_f\cdot\mathbf{E} < 0$. Thus, Eq. (4) reconfirms that positive free charges ($\rho^f < 0$) should be generated. Alternatively, if the concentration gradient and the electric field are in the same direction, negative charges should be induced.

Basically, the free charge is generated during the electromigration of ions, which is dependent on local ionic concentration, to satisfy the constant current condition. That is, a slight deviation from the electroneutrality effectively changes and adjusts the electric field so as to keep the current conservation condition. We call this phenomenon, in which the free charges are generated in a medium being associated with the concentration gradient, the "concentration polarization" [10–14].

2.2. Concentration-polarization force

Let us consider the effect of the concentration polarization on the pressure field. All the free charges are subject

to the Coulombic force, which develops a pressure field inside the fluid domain, in analogy to the hydrostatic pressure generated by the gravity force. We call this pressure the electrohydrodynamic (EHD) pressure. If any fluid motion inside the domain is neglected, the Coulombic force is balanced by the EHD pressure (p_s). That is,

$$\nabla p_s = \rho^f \mathbf{E}. \tag{5}$$

For an unbounded one-dimensional domain, such as shown in Fig. 1, the pressure gradient becomes, from Eqs. (4) and (5),

$$\frac{\mathrm{d}p}{\mathrm{d}x} = -\left[\frac{\varepsilon_f(\mathrm{d}\sigma_f/\mathrm{d}x)E}{\sigma_f}\right]E = -\frac{\varepsilon_f i^2}{\sigma_f^3}\frac{\mathrm{d}\sigma_f}{\mathrm{d}x},$$

where $i = \sigma_f E$ is constant over the one-dimensional domain and $E = |\mathbf{E}|$. By integrating both sides of the preceding equation, we obtain the pressure distribution of

$$p_s = p_{\mathrm{ref}} + \frac{\varepsilon_f}{2}\frac{i^2}{\sigma_f^2} = p_{\mathrm{ref}} + \frac{\varepsilon_f E^2}{2}, \tag{6}$$

where p_{ref} is a constant and the electric field is a function of local concentration.

Such kind of mechanism for free-charge generation and the subsequent development of the EHD pressure has been well perceived in the electrochemistry and the electrohydrodynamics areas. For example, it can generate the troublesome hydrodynamic flow in isoelectric focusing devices [13], and the associated EHD flow can significantly influence the current–voltage relationship of an electrode [11]. It is also suggested that the long-range interaction and the subsequent aggregation of particles on the electrode surface is a direct consequence of the EHD flow [14,15].

Now, we place a dielectric particle in the liquid as considered in Fig. 1. According to Eq. (6), the magnitude of the EHD pressure is inversely proportional to the concentration (conductivity). For the case in Fig. 1, the concentration is lower at RHS of the particle, and therefore, the pressure will be higher at RHS, as depicted in Fig. 2a. This will generate a finite magnitude of force acting on the particle, which constitutes a part of the CP force.

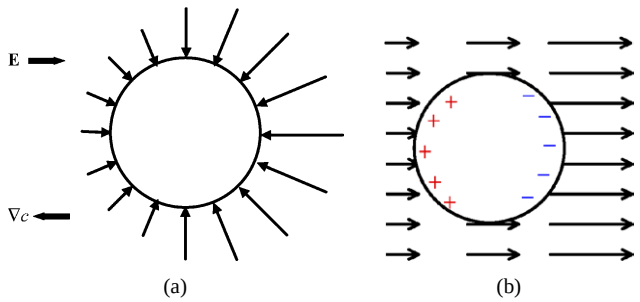

Fig. 2. Schematic diagram representing the two contributions to the CP force: (a) by electrohydrodynamic pressure; (b) by nonuniform electric field. The arrows in (a) and (b) represent the pressure force and the electric field, respectively.

On the other hand, when a dielectric particle is placed in a nonuniform electric field, the dielectrophoretic (DEP) force arises (see Fig. 2b) [16,17]. The DEP force has often been interpreted, for the case of a nonconducting medium, as the net force acting on the polarized charge inside a dielectric particle by the surrounding "nonuniform" electric field. That is, if the unperturbed electric field by the particle is nonuniform and nonsymmetric, alike those depicted in Fig. 2b, the force acting on RHS and LHS of the particle is different. This difference develops the DEP force. In the case we have been considering, the electric field will be stronger at RHS than at LHS of the particle (see Fig. 2b). We will show later that this will generate a DEP force with a finite magnitude.

These two kinds of forces primarily constitute the CP force. In addition, the nonuniform pressure field around the particle generates a hydrodynamic flow, and the flow also exerts a certain amount of hydrodynamic force on the particle. The practical magnitude of the CP force should be determined by considering the actual concentration, electric field, and hydrodynamic field around the particle. The subsequent two sections are devoted to a linear analysis of the governing equations for quantitative prediction of the CP force.

3. Formulation

In Section 3.1, we will derive the basic equations for the electric, concentration, and hydrodynamic fields. Then, the expression for the hydrodynamic and the electrical contribution to the CP force will be given. In Section 3.2, some simplifying assumptions are introduced to obtain a closed form solution for the CP force. These include the electroneutrality and quasi-steady process assumptions, and a simple linear profile for the imposed concentration distribution. The boundary conditions are specified after derivation of each approximated form of the equation. In Section 3.3, all the equations and boundary conditions are summarized in a nondimensional form. Finally, a perturbation analysis will be conducted in Section 4, with respect to the modeled system under a limiting condition.

3.1. Basic equations

Consider a dielectric solid particle of a nonconducting material with a radius a submerged in an electrolyte stream (see Fig. 3) and moving with the flow. The electrolyte has a concentration gradient in the direction perpendicular to the flow, and is free from any chemical reaction. There exists an externally applied DC electric field parallel to the concentration gradient, and the electric field is nonuniform due to the conductivity gradient. All the material properties including the dielectric constant are assumed uniform in the liquid and the solid regions, respectively, unless specified otherwise. The activity coefficient of the ionic species is assumed to be unity. This condition is justified for the case of infinite dilution [18]. All electrokinetic effects are neglected.

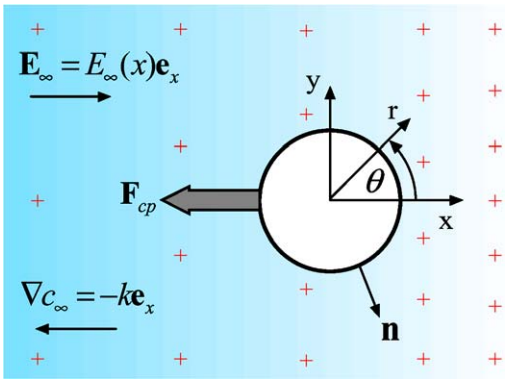

Fig. 3. Domain of analysis and coordinate systems. The symbols "+" represents that the domain is positively charged due to the concentration polarization, under the conditions given in this figure.

The CP force acting on the particle generates a particle motion with a velocity $\mathbf{U}_{\rm cp}$ relative to the liquid. We introduce the (r,θ,φ) spherical coordinate system and the (x,y,z) Cartesian coordinate system with their origins fixed at the center of the particle moving with the velocity $\mathbf{U}_{\rm cp}$. Thus, the particle motion can be translated into the relative liquid flow. We assume the axial symmetry of the system in φ direction.

The electromagnetic field inside the region is governed by the Maxwell equations. In the quasi-electrostatic limit, the electric field is separable from the magnetic field, and the Maxwell equations are simplified to [1,9]

$$\nabla\cdot(\varepsilon\mathbf{E}) = \rho^f, \tag{7}$$

$$\nabla\times\mathbf{E} = 0, \tag{8}$$

$$\frac{\partial\rho^f}{\partial t} + \nabla\cdot\mathbf{i} = 0, \tag{9}$$

where t is the time, and $\varepsilon = \varepsilon_f$ in the liquid domain and $\varepsilon = \varepsilon_p$ inside the particle. The electric field is coupled with other transport equations by the free charge that is represented by

$$\rho^f = F(c_+ - c_-), \tag{10}$$

where $c_\pm$ is the molar concentration (mol/m^3). To complete the description of the system, the transport equation for the concentration is necessary.

For dilute solutions, the molar flux density of ionic species is written as the sum of diffusional, electromigrational, and convectional components as follows [8]:

$$\mathbf{N}_\pm = -z_\pm\omega_\pm F c_\pm\nabla\phi - D_\pm\nabla c_\pm + c_\pm\mathbf{u},$$

where $D_\pm$ is the diffusivity of the ionic species (m^2/s), $z_+ = 1$, $z_- = -1$, $F = N_A e$ the Faraday constant, N_A the Avogadro number, e the electronic charge, $\omega_\pm = D_\pm/N_A kT$ the mobility of the ionic species (mol mN^{-1} s^{-1}), k the Boltzmann constant, T the absolute temperature, and $\mathbf{u}$ the convection velocity. If there is no chemical reaction and no generation of ionic species, the conservation of species at a point requires $\partial c_\pm/\partial t + \nabla\cdot\mathbf{N}_\pm = 0$. If we substitute $\mathbf{N}_\pm$ to the preceding equation, we obtain the transport equations for each sign of ionic species as follows:

$$\frac{\partial c_+}{\partial t} + \mathbf{u}\cdot\nabla c_+ = D_+\nabla^2 c_+ + \beta D_+\nabla\cdot(c_+\nabla\phi), \tag{11}$$

$$\frac{\partial c_-}{\partial t} + \mathbf{u}\cdot\nabla c_- = D_-\nabla^2 c_- - \beta D_-\nabla\cdot(c_-\nabla\phi), \tag{12}$$

in which $\beta = e/(kT)$.

The incompressible hydrodynamic flow satisfies the following mass conservation and the momentum equations:

$$\nabla\cdot\mathbf{u} = 0, \tag{13}$$

$$\rho\left(\frac{\partial\mathbf{u}}{\partial t} + \mathbf{u}\cdot\nabla\mathbf{u}\right) = \nabla\cdot(\boldsymbol{\tau} + \mathbf{T}). \tag{14}$$

Here, ρ is the fluid density, and $\boldsymbol{\tau}$ and $\mathbf{T}$ are the hydrodynamic and the Maxwell stresses, respectively, which are defined as

$$\boldsymbol{\tau} = -p\mathbf{I} + \mu(\nabla\mathbf{u} + \nabla\mathbf{u}^T), \tag{15}$$

$$\mathbf{T} = -\frac{\varepsilon}{2}E^2\mathbf{I} + \varepsilon\mathbf{E}\mathbf{E}, \tag{16}$$

where $E^2 = |\mathbf{E}|^2$, $\mathbf{I}$ the second-order isotropic tensor, p the pressure, and μ the fluid viscosity.

The force acting on a particle is obtained as $\mathbf{F}_{\rm CP} = \int_{S_p}\|\boldsymbol{\tau}+\mathbf{T}\|\cdot\mathbf{n}\,\mathrm{d}S$, in which S_p denotes the particle surface, and $\|\cdot\|$ denotes the difference between the value at the exterior side and the value at the interior side of the particle surface. The x-directional force, $F_{\rm CP} = \mathbf{F}_{\rm CP}\cdot\mathbf{e}_x = F^M + F^h$, is a sum of the contributions from the Maxwell stress (F^M) and the hydrodynamic stress (F^h), in which [10,19]

$$F^M = \int_0^{2\pi}\int_0^{\pi}\|\cos\theta T_{rr} - \sin\theta T_{r\theta}\|\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\varphi, \tag{17}$$

$$F^h = \int_0^{2\pi}\int_0^{\pi}(\cos\theta\tau_{rr} - \sin\theta\tau_{r\theta})_{r=a}\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\varphi. \tag{18}$$

Here,

$$T_{rr} = \frac{\varepsilon}{2}\left(E_r^2 - E_\theta^2\right),$$

$$T_{r\theta} = \varepsilon E_r E_\theta,$$

$$\tau_{rr} = -p + 2\mu\frac{\partial u_r}{\partial r},$$

$$\tau_{r\theta} = \mu\left[r\frac{\partial}{\partial r}\left(\frac{u_\theta}{r}\right) + \frac{1}{r}\frac{\partial u_r}{\partial\theta}\right],$$

where $E_r = \mathbf{E}\cdot\mathbf{e}_r$, $E_\theta = \mathbf{E}\cdot\mathbf{e}_\theta$, and $\mathbf{e}_r$ and $\mathbf{e}_\theta$ represent the unit vectors in r and θ direction, respectively.

3.2. Simplifications

Equations (7) through (16) complete the description for the electric, concentration and hydrodynamic fields. For the analysis of the system, two important assumptions are introduced. One is the quasi-steady approximation for the electric, the concentration, and the hydrodynamic fields. The

Table 1
Comparison of the characteristic time scales

Classification	Characteristic time scale	Sample value (s)
Electric field	$\tau_c = \varepsilon_f/\sigma_f$	1.2×10^{-7}
Concentration	$\tau_D^a = a^2/D$	6.2×10^{-2}
	$\tau_D^l = l^2/D$	6.2
Momentum	$\tau_v = a^2/\nu$	9.3×10^{-8}

other is the quasi-electroneutrality assumption, which has been discussed earlier, under which we let $c_+ \cong c_- = c$.

The quasi-steady approximation is based on the slow molecular diffusion process and subsequent slow change of bulk concentration distribution. The characteristic time scales, which measure the time required to establish a steady condition for each process, are summarized and sample values are computed in Table 1. For the electric field, the so-called electrical relaxation time ($\tau_c = \varepsilon_f/\sigma_f$) is chosen [10]. For the concentration field, the characteristic time scale for the bulk concentration is represented by $\tau_D^l = l^2/D$, while that for the localized change of concentration around the particle is represented by $\tau_D^a = a^2/D$. Here, l represents the length scale of the bulk liquid and $D = 2D_+D_-/(D_+ + D_-)$ is the equivalent diffusivity of electrolyte. In the sample calculation, the case of particle having $a = 10^{-5}$ m in an aqueous NaCl solution, in which $D_+ = 1.33 \times 10^{-9}$ m^2/s and $D_- = 2.03 \times 10^{-9}$ m^2/s, $D = 1.607 \times 10^{-9}$ m^2/s, $c = 1$ mol/m^3, and $\sigma_f = 5.6 \times 10^{-3}$ S/m, is considered. The macroscopic length scale l is chosen as $l = 10^{-4}$ m, considering the width of a conventional microchannel. Compared to other transport phenomena, the bulk-concentration-change process is sufficiently slow. Thus, for a given bulk concentration distribution, all the other transport processes can be approximately considered steady.

For an electrolyte, the current is due to the motion of ionic species. The current density can be expressed as $\mathbf{i} = F(\mathbf{N}_+ - \mathbf{N}_-)$ which becomes, under the quasi-electroneutrality assumption,

$$\mathbf{i} = -\sigma_f \nabla\phi - F(D_+ - D_-)\nabla c,$$

where the conductivity in Eq. (1) is rewritten as $\sigma_f = F\beta(D_+ + D_-)c$. When there exists a macroscopic concentration gradient, the diffusivity difference autonomously develops the so-called diffusion potential without any external electric field. For an 1:1 electrolyte, the associated electric field is expressed as $\mathbf{E}_D = \beta^{-1}(\alpha_+ - \alpha_-)\nabla \ln c$ [3], in which $\alpha_\pm = D_\pm/(D_+ + D_-)$. For the case of NaCl, $\alpha_+ - \alpha_- = -0.21$, and we let $\nabla c = 10^4$ mol/m^3 and $c = 1$ mol/m^3. Then, the estimated electric field is about 54 V/m. Here, we are interested in moderate strength of electric fields which is in the order of 10^4 V/m (or higher). We neglect, therefore, the diffusional contribution to current and the associated diffusion potential. Then, the current density satisfies the following Ohm's law:

$$\mathbf{i} = \sigma_f \mathbf{E}.$$

In Eq. (9), under a DC field, the displacement current, $\partial\rho^f/\partial t$, can be neglected for microscale systems [1,9,10], due to the quasi-steady approximation. Then, the current conservation equation, $\nabla \cdot \mathbf{i} = 0$, reduces to the following equation for the electrical potential at the exterior (ϕ_e) and the interior of the particle (ϕ_i):

$$\nabla \cdot (\sigma_f \nabla\phi_e) = 0 \quad \text{at } r > a, \tag{19}$$

$$\nabla^2\phi_i = 0 \quad \text{at } r < a. \tag{20}$$

In general, the current should be conserved when it passes through the particle surface, which is expressed as $\sigma_f \mathbf{n} \cdot \nabla\phi_e|_{r=a} = \sigma_p \mathbf{n} \cdot \nabla\phi_i|_{r=a}$, where σ_p represents the particle conductivity. However, we assumed that the particle has zero conductivity, i.e., $\sigma_p = 0$. Accordingly, there is no electric current penetrating the particle surface, and the preceding current conservation relation becomes

$$\nabla\phi_e \cdot \mathbf{n} = 0 \quad \text{at } r = a. \tag{21}$$

An electric field of $\mathbf{E}_\infty = E_\infty(x)\mathbf{e}_x$ is formed far from the particle. That is,

$$-\nabla\phi_e \to \mathbf{E}_\infty(x) \quad \text{as } r \to \infty. \tag{22}$$

If the equation $\nabla \times \mathbf{E} = 0$ in Eq. (8) is applied to an infinitesimal surface element at the particle–fluid interface, we obtain the following condition stating that the tangential electric field is continuous at the particle surface [9,10]:

$$\frac{\partial\phi_e}{\partial\theta} = \frac{\partial\phi_i}{\partial\theta} \quad \text{at } r = a, \tag{23}$$

which can be further reduced to

$$\phi_e = \phi_i \quad \text{at } r = a. \tag{24}$$

We multiply D_- and D_+ to Eqs. (11) and (12), respectively, and then add the resulting two equations to eliminate the electromigration term. This results in the following equation for the species conservation: $\partial c/\partial t + \mathbf{u} \cdot \nabla c = D\nabla^2 c$. However, due to the quasi-steady approximation, the unsteady term in the species conservation equation is dropped, and the equation is reduced to

$$\mathbf{u} \cdot \nabla c = D\nabla^2 c. \tag{25}$$

The particle is assumed nonreacting with the electrolyte; and therefore, there is no penetration of the solute into the particle surface, i.e.,

$$\mathbf{n} \cdot \nabla c = 0 \quad \text{at } r = a. \tag{26}$$

We consider the case of a linear concentration profile. Therefore, at a position far from the particle, the concentration can be expressed as

$$c \to c_\infty(x) = c_0 - kx = c_0\left(1 - \frac{\lambda x}{a}\right) \quad \text{as } r \to \infty, \tag{27}$$

where $k = -\nabla c_\infty$ and $c_0 = c_\infty(x = 0)$. The solution of the species conservation equation, Eq. (25), for the linear concentration profile may provide a leading order solution for an

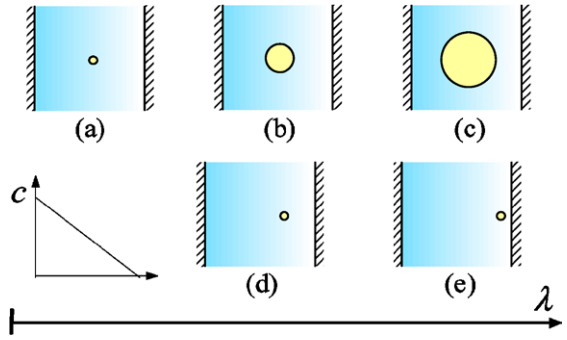

Fig. 4. Effect of particle size and the location on λ for an identical concentration profile.

arbitrary concentration distribution. In the preceding equation, λ is the so-called effective concentration gradient which is defined by

$$\lambda \equiv \frac{ka}{c_0}.$$

This represents the concentration difference between the ends of a particle normalized by the concentration at the particle center. Since the electric-field strength is proportional to the inverse of concentration, λ also can be considered as the difference in electric-field strength between the ends of the particle normalized by the electric-field strength at the center position of the particle.

Fig. 4 illustrates the dependence of λ on particle size and its location in a channel with respect to a linear concentration distribution. The value of λ may increase proportionally to particle size and the inverse of local concentration. For a large value of λ, the influence of the sidewalls may be significant. Thus, the present analysis is focused on a rather limiting condition of small λ, so that the wall effect can be neglected, such as the case of Fig. 4a. For the sample calculation of λ, let us consider two streams of different electrolyte concentration of 50 mol/m^3 (=50 mM) and 1 mol/m^3 (=1 mM) flowing in parallel in a microchannel having a width of 100 μm. The diffusional mixing at the interface will generate a concentration gradient. Then, k may be in the order of 5×10^5 mol/m^4. When $a = 10$ μm, it becomes $\lambda = 0.1$.

For the momentum equation in Eq. (14), we neglect the convection term under the creeping flow assumption, and the unsteady term is also dropped off due to the quasi-steady approximation. Then, it becomes

$$-\nabla p - \mu \nabla \times \nabla \times \mathbf{u} + \rho^f \mathbf{E} = 0. \tag{28}$$

The no-slip condition is applied at the particle surface. Far from the particle, there is a relative fluid motion arising from the migration of particle, $\mathbf{U}_{\mathrm{CP}}$. Then, the hydrodynamic problem will be decomposed into two problems: (i) the flow with $\mathbf{u}|_{r\to\infty} = -\mathbf{U}_{\mathrm{CP}}$ but without the body force, and (ii) the flow with $\mathbf{u}|_{r\to\infty} = 0$ and with the Coulombic body force. Obviously, the problem (i) produces the Stokes force. Therefore, we analyze only the second problem, in which the boundary conditions become

$$\mathbf{u} = \mathbf{0} \quad \text{at } r = a, \tag{29}$$

$$\mathbf{u} \to \mathbf{0} \quad \text{as } r \to \infty. \tag{30}$$

3.3. Nondimensionalization

The variables are nondimensionalized as $\tilde{x} = x/a$, $\tilde{p} = p/(\varepsilon_f E_0^2)$, $\tilde{\mathbf{u}} = \mathbf{u}/u_c$, $\tilde{c} = (c - c_0)/c_0$, $\tilde{\phi} = \phi/(aE_0)$, and $\tilde{\rho}^f = \rho^f/(\varepsilon_f E_0/a)$, where $E_0 = |\mathbf{E}_\infty(x = 0)|$. The particle radius a is chosen as the characteristic length. The characteristic velocity u_c is determined, through the balance of the Coulombic force and the viscous force, as $u_c = \varepsilon_f E_0^2 a/\mu$. Substituting the dimensionless variables into the governing equations, we obtain the following set of nondimensional equations and boundary conditions.

Governing equations:

$$-\tilde{\nabla}\tilde{p} - \tilde{\nabla} \times \tilde{\nabla} \times \tilde{\mathbf{u}} + \tilde{\rho}^f \tilde{\mathbf{E}} = 0, \tag{31}$$

$$\tilde{\nabla} \cdot \left[(\tilde{c} + 1)\tilde{\nabla}\tilde{\phi}_e\right] = 0, \tag{32}$$

$$\tilde{\nabla}^2 \tilde{\phi}_i = 0, \tag{33}$$

$$\tilde{\nabla}^2 \tilde{c} = Pe_{\mathrm{el}} \tilde{\mathbf{u}} \cdot \tilde{\nabla}\tilde{c}. \tag{34}$$

Boundary conditions:

$$\tilde{\mathbf{u}} = \mathbf{0} \quad \text{at } \tilde{r} = 1, \tag{35}$$

$$\tilde{\mathbf{u}} \to \mathbf{0} \quad \text{at } \tilde{r} \to \infty, \tag{36}$$

$$\mathbf{n} \cdot \tilde{\nabla}\tilde{c} = 0 \quad \text{at } \tilde{r} = 1, \tag{37}$$

$$\tilde{c} \to \tilde{c}_\infty = 1 - \lambda\tilde{x} \quad \text{as } \tilde{r} \to \infty, \tag{38}$$

$$\mathbf{n} \cdot \tilde{\nabla}\tilde{\phi}_e = 0 \quad \text{at } \tilde{r} = 1, \tag{39}$$

$$\tilde{\phi}_e = \tilde{\phi}_i \quad \text{at } \tilde{r} = 1, \tag{40}$$

$$-\tilde{\nabla}\tilde{\phi}_e \to \tilde{E}_\infty(\tilde{x})\mathbf{e}_x = \frac{1}{1 - \lambda\tilde{x}} \quad \text{as } \tilde{r} \to \infty. \tag{41}$$

Here, $Pe_{\mathrm{el}} = \varepsilon_f E_0^2 a^2/(\mu D)$ is the electrical Peclet number that represents the ratio of the convective transport due to the EHD flow to the diffusive transport. Hereinafter, the tilde is dropped for the sake of convenience.

4. Analysis

In this section, we are going to obtain a leading order term of the CP force after performing a perturbation analysis for the equations summarized at the end of the previous section. As stated in Section 2.2, the CP force is composed of the EHD and DEP contributions. The leading order term of the DEP contribution can be obtained without considering the hydrodynamic field. What is necessary for the evaluation of the DEP contribution is just the electric field in and outside of the particle. To evaluate the EHD contribution, the flow field should be analyzed to obtain the pressure and the skin friction distributions on a particle surface.

The analysis will be done as follows: In Section 4.1, an accurate form of the solution for the electric field for a given

concentration distribution will be given first. Then, the solution for the concentration field will be obtained neglecting the convective contribution. The leading order solution for the electric field at exterior and interior of the particle will be obtained for the given concentration distribution. In Section 4.2, the creeping flow will be analyzed. In the analysis, the solution for the electric field will be used to determine the leading order term of the Coulombic body force. In Section 4.3, each term appearing in the integrals of the Maxwell stress and the hydrodynamic stresses will be evaluated, separately, and then the CP force will be obtained by summing all the terms. In Section 4.4, a numerical validation of the theoretical prediction will be made with respect to the surface pressure and the skin friction distribution.

4.1. Electric field and concentration field

We performed a perturbation analysis on a set of equations shown in Eqs. (31)–(41) under the assumptions of $0 < \lambda \ll 1$ and $O(Pe_{\mathrm{el}}) < 1$. The latter assumption is used to eliminate the convection term in the concentration equation, Eq. (34), in order to decouple the concentration field from the hydrodynamic field. The validity of the latter condition, for a particle of $a = 10$ μm in an aqueous NaCl electrolyte solution, may be assured up to an E_0 value of approximately 10^4 V/m or 100 V/cm. This value of electric field is quite ordinary in many electrokinetic problems.

The variables $\mathbf{u}$, P, c, and $\mathbf{E}$ are expanded by perturbation series, with respect to a small parameter λ, in the form of

$$\phi_e = \phi_{e0} + \lambda\phi_{e1} + \lambda^2\phi_{e2} + \cdots,$$
$$\phi_i = \phi_{i0} + \lambda\phi_{i1} + \lambda^2\phi_{i2} + \cdots,$$
$$c = \lambda c_1 + \lambda^2 c_2 + \lambda^3 c_3 + \cdots,$$
$$\mathbf{u} = \lambda\mathbf{u}_1 + \lambda^2\mathbf{u}_2 + \lambda^3\mathbf{u}_3 + \cdots,$$
$$p = \lambda p_1 + \lambda^2 p_2 + \lambda^3 p_3 + \cdots.$$

In the above expressions, ϕ_{e0} and ϕ_{i0} represent the zeroth order solution for the electrostatic potential at exterior and interior of the particle, respectively. Therefore, ϕ_{e0} and ϕ_{i0} represent the electrostatic field without concentration gradient (σ_f is constant), and satisfy the Laplace equations. It can be proved easily, by substituting to Eq. (32), that the following is the solution for the exterior electric field:

$$\nabla\phi_e = \frac{\nabla\phi_{e0}}{1+c}. \tag{42}$$

Then, by taking divergence to the above equation and substituting the results to the nondimensionalized Poisson equation, $\rho^f = -\nabla^2\phi$, the charge density is obtained as

$$\rho^f = \frac{\nabla c\cdot\nabla\phi_{e0}}{(1+c)^2}. \tag{43}$$

On the other hand, the leading order terms of flow velocity and electrolyte concentration are of the order of λ. The term in RHS of Eq. (34) is in the order of $Pe_{\mathrm{el}}\lambda^2$ while the term in LHS is in the order of λ. Therefore, under the assumption that $Pe_{\mathrm{el}}\lambda \ll 1$, the concentration distribution satisfies the following Laplace equation of $\nabla^2 c = 0$, to the first order of λ. Moreover, the zeroth order electrostatic potential and c_1 satisfy the same boundary conditions of

$$\mathbf{E}_{e0}|_{r\to\infty} = -\nabla c_1|_{r\to\infty} = \mathbf{e}_x,$$
$$\mathbf{E}_{e0}\cdot\mathbf{n}|_{r=1} = \nabla c_1\cdot\mathbf{n}|_{r=1} = 0.$$

Thus, it is evident that $\phi_{e0} = c_1$ and $\mathbf{E}_{e0} = -\nabla c_1$ which become [8]

$$\phi_{e0} = c_1 = -\left(r + \frac{1}{2r^2}\right)\cos\theta, \tag{44}$$

$$\mathbf{E}_{e0} = -\nabla c_1 = \mathbf{e}_r\left(1 - \frac{1}{r^3}\right)\eta - \mathbf{e}_\theta\left(1 + \frac{1}{2r^3}\right)\sqrt{1-\eta^2}, \tag{45}$$

where $\eta = \cos\theta$.

Note that the expression for the external electric field given in Eq. (42) is accurate for any given concentration distributions. Accordingly, the electric field is readily obtained by using Eqs. (42) and (44) for any given concentration field. Equation (42) can be expanded in the form of $\nabla\phi_e = \nabla\phi_{e0}(1-\lambda c_1) + \cdots$ which can be rewritten as $\nabla\phi_e = \nabla(\phi_{e0} - \lambda\phi_{e0}^2/2) + \cdots$. Thus, the exterior electrostatic potential, to the first order, becomes

$$\phi_e = \phi_{e0} - \frac{\lambda}{2}\phi_{e0}^2 + \phi_{e,\mathrm{ref}}, \tag{46}$$

where the reference potential is chosen $\phi_{e,\mathrm{ref}} = 0$. It follows that the exterior electrical field can be expanded in the form of $\mathbf{E}_e = -\nabla\phi_e \cong \mathbf{E}_{e0} + \lambda\mathbf{E}_{e1} + \cdots$, and can be written to the first order of λ as follows:

$$\mathbf{E}_e \cong -\nabla\phi_{e0}(1-\lambda c_1) = \mathbf{E}_{e0} - \lambda c_1\mathbf{E}_{e0}. \tag{47}$$

The zeroth order solution for the exterior electric field ($\mathbf{E}_{e0}$) is given by Eq. (45), and the first-order external electric field ($\mathbf{E}_{e1}$) becomes $\mathbf{E}_{e1} = -c_1\mathbf{E}_{e0} = c_1\nabla c_1$.

For the interior electric field, we can express the solution by using the Legendre polynomial $P_n(\eta)$ as

$$\phi_i = \sum_n (a_n + \lambda b_n + \cdots) r^n P_n(\eta).$$

According to Eq. (40), the electrostatic potential at the particle surface is continuous, i.e., $\phi_e = \phi_i$ at $r = 1$. This condition is applied to determine the unknown coefficients, and the resulting electrostatic potential and the electric field, to the first order, become

$$\phi_i = -\frac{3}{2}r\eta - \frac{3\lambda}{8}r^2(3\eta^2-1) - \frac{3\lambda}{8}, \tag{48}$$

$$\mathbf{E}_i = \frac{3}{2}\left[\eta\mathbf{e}_r - \sqrt{1-\eta^2}\mathbf{e}_\theta\right] + \frac{3\lambda}{4}r\left[(3\eta^2-1)\mathbf{e}_r - 3\eta\sqrt{1-\eta^2}\mathbf{e}_\theta\right]. \tag{49}$$

Note that the electric field inside the particle is independent of electrical properties of the particle. This is because we assumed the particle is composed of a nonconducting

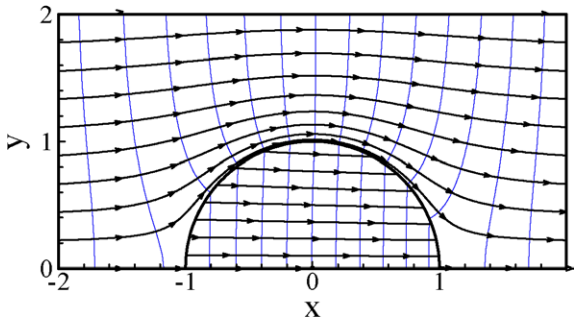

Fig. 5. Electrostatic potential (line without arrow, blue colored) and electric field (line with arrows, black colored) to the first order of λ when $\lambda = 0.1$. (Color drawing in the online version.)

material, and there is no current flow inside the particle. The electric field inside the particle is rather passively determined by the exterior electric field. It is also noticeable that, different from the free charge density, the exterior electric field is independent of the electrical permittivity.

Fig. 5 shows the electrostatic potential and the electric field, to the first order of λ, at interior and exterior of the particle, in which $\lambda = 0.1$. The exterior electric field should be generally stronger at RHS than at LHS since the concentration is relatively smaller at RHS than at LHS. The interior electric field also directs to the positive x-direction. The zeroth order interior electric field is in fact represented by $\mathbf{E}_{i0} = (3/2)\mathbf{e}_x$, and thus, it is uniform inside the particle and has only x-directional component. The θ-directional term in the first order terms of $\mathbf{E}_i$ is positive at RHS and negative at LHS. Therefore, at interior of the particle, the electric-field strength should be somewhat greater at RHS.

4.2. Hydrodynamic field

In order to find the EHD component of the CP force, the hydrodynamic field should be analyzed. This can be done by following the well-established analytical procedures for the axisymmetric creeping flow [19,20]. The present problem is a little distinct as the Coulombic force is included.

The momentum equation in Eq. (31) can be rewritten, to the first order of λ, as

$$-\nabla p_1 - \nabla \times \Omega_1 + \mathbf{B}_1 = 0, \tag{50}$$

where $\mathbf{B}_1$ is the first-order component of the Coulombic body force and $\Omega_1 = \nabla \times \mathbf{u}_1$ is the vorticity vector. We introduce the vector potential for vorticity ($\mathbf{A}_1$) which is related with vorticity as $\Omega_1 = \nabla \times \nabla \times \mathbf{A}_1$. This vector potential and the vorticity vector have only φ-directional component, which is denoted by A_1 and ω_1, owing to the axisymmetry of the flow assumed in this study. This function is related to the stream function ψ_1 as $A_1 = \psi_1/(r\sin\theta)$. The stream function automatically satisfies the continuity equation, and is related with vorticity as follows:

$$\Omega_1 = \omega_1 \mathbf{e}_\varphi = -\frac{\mathbf{e}_\varphi}{r^2\sqrt{1-\eta^2}} E^2 \psi_1, \tag{51}$$

where

$$E^2 = \frac{\partial^2}{\partial r^2} + \frac{(1-\eta^2)}{r^2}\frac{\partial^2}{\partial \eta^2}.$$

The first order velocity components in r and θ directions are represented by the stream function as

$$u_{1r} = -\frac{1}{r^2}\frac{\partial \psi_1}{\partial \eta}, \tag{52}$$

$$u_{1\theta} = -\frac{1}{r\sqrt{1-\eta^2}}\frac{\partial \psi_1}{\partial r}. \tag{53}$$

If the curl is taken to both sides of Eq. (50), the following fourth-order differential equation is obtained

$$E^4 \psi_1 = -r\sqrt{1-\eta^2}(\nabla \times \mathbf{B}_1). \tag{54}$$

The first order components of the charge density are obtained by way of the Taylor series expansion of Eq. (43) as follows:

$$\rho_1^f = -\nabla c_1 \cdot \mathbf{E}_{e0} = \lambda \mathbf{E}_{e0} \cdot \mathbf{E}_{e0},$$

where $\nabla c_1 = -\mathbf{E}_{e0}$. Then, the first order Coulombic body force becomes $\mathbf{B}_1 = (\mathbf{E}_{e0} \cdot \mathbf{E}_{e0})\mathbf{E}_{e0}$ which can be written as

$$\mathbf{B}_1 = \left\{1 + \frac{1}{r^3} + \frac{1}{4r^6} + \left(-\frac{3}{r^3} + \frac{3}{4r^6}\right)\eta^2\right\}\mathbf{E}_{e0}. \tag{55}$$

Since $\nabla \times \mathbf{E}_{e0} = 0$, the source term in the stream-function equation, Eq. (54), can be further simplified to $\nabla \times \mathbf{B}_1 = [\nabla(\mathbf{E}_{e0} \cdot \mathbf{E}_{e0})] \times \mathbf{E}_{e0}$. Then, Eq. (54) is rewritten as

$$E^4 \psi_1 = -r\sqrt{1-\eta^2}\big[\nabla(\mathbf{E}_{e0} \cdot \mathbf{E}_{e0})\big] \times \mathbf{E}_{e0}.$$

Substituting $\mathbf{E}_{e0}$ in Eq. (45) into the above equation, we obtain

$$E^4 \psi_1 = 9\left(\frac{1}{r^6} + \frac{1}{5r^9}\right) Q_1(\eta) - 24\left(\frac{1}{r^3} - \frac{1}{2r^6} - \frac{1}{20r^9}\right) Q_3(\eta). \tag{56}$$

The Gegenbauer polynomials $Q_n(\eta)$ are related with the Legendre polynomial $P_n(\eta)$ as $Q_n(\eta) = \int_{-1}^{\eta} P_n(\eta)\,d\eta$, and their first- and third-order polynomials are written as $Q_1(\eta) = (1/2)(\eta^2 - 1)$ and $Q_3(\eta) = (1/8)(5\eta^2 - 1) \times (\eta^2 - 1)$. The stream function should satisfy the no-slip condition at the particle surface, which is written as

$$\frac{\partial \psi_1}{\partial r} = 0 \quad \text{at } r = 1,$$
$$\psi_1 = 0 \quad \text{at } r = 1.$$

The stream function is decomposed into homogeneous solution (ψ_{1h}), which satisfies $E^4 \psi_1 = 0$, and the particular solution (ψ_{1p}). The homogeneous solution can be represented in terms of the Gegenbauer function as follows:

$$\psi_{1h} = \sum_{n=1}^{\infty}\big[A_n r^{n+3} + B_n r^{n+1} + C_n r^{2-n} + D_n r^{-n}\big] Q_n(\eta),$$

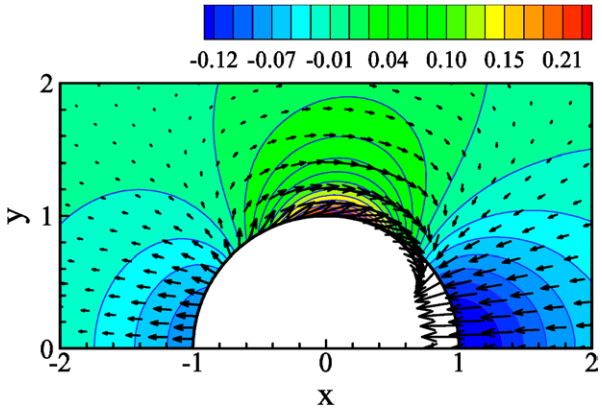

Fig. 6. Effective body force (vector) when $\lambda = 0.1$. Contour represents the x-directional component (b_{1x}) of the effective body force.

where A_n, B_n, C_n, and D_n are unknown coefficients. By substituting the functional form of $r^\alpha Q_n(\eta)$ into Eq. (56), we obtain the following particular solution:

$$\psi_{1p} = \left(\frac{1}{8r^2} + \frac{1}{840r^5}\right) Q_1(\eta) + \left(-\frac{r}{5} - \frac{1}{4r^2} + \frac{1}{660r^5}\right) Q_3(\eta).$$

Then, the solution for Eq. (56) can be written in the form of

$$\psi_1 = \sum_{n=1}^{\infty} \left[A_n r^{n+3} + B_n r^{n+1} + C_n r^{2-n} + D_n r^{-n}\right] Q_n(\eta) + \left(\frac{1}{8r^2} + \frac{1}{840r^5}\right) Q_1(\eta) + \left(-\frac{r}{5} - \frac{1}{4r^2} + \frac{1}{660r^5}\right) Q_3(\eta).$$

The unknown coefficients are obtained by applying the no-slip boundary condition. All the coefficients vanish except $C_1 = 109/1680$, $C_3 = 139/264$, $D_1 = -107/560$, and $D_3 = -103/1320$. Accordingly, it becomes

$$\psi_1(r,\eta) = \left(C_1 r + \frac{D_1}{r} + \frac{1}{8r^2} + \frac{1}{840r^5}\right) Q_1(\eta) + \left(-\frac{r}{5} + \frac{C_3}{r} - \frac{1}{4r^2} + \frac{D_3}{r^3} + \frac{1}{660r^5}\right) Q_3(\eta). \tag{57}$$

On the other hand, the body force can be decomposed into the far-field component $\mathbf{B}_{1\infty} = \mathbf{B}_1(r \to \infty)$ and its remainder $\mathbf{b}_1 = \mathbf{B}_1 - \mathbf{B}_{1\infty}$. According to Eq. (55), $\mathbf{B}_{1\infty} = \mathbf{e}_x$ and $\nabla \times \mathbf{B}_{1\infty} = 0$ to the first order. Therefore, the far-field component $\mathbf{B}_{1\infty}$ does not contribute to fluid motion, and is exactly balanced by pressure. Only the effective body force $\mathbf{b}_1$ contributes to the generation of the hydrodynamic flow. Fig. 6 shows the effective body force (vector), when $\lambda = 0.1$, together with the x component of the body force (contour). The effective body force decays with r^{-3}, and therefore, it is confined within a region very close to the particle. As shown in the figure, the body force is positive at the top of the particle (around $\theta = \pi/2$) and negative at both sides of the particle (around $\theta = 0$ and π). It can be inferred from the figure that the pressure unaffected by fluid motion (i.e., pressure due to the concentration polarization) may exert force to the right at the top region, while the force is towards the left at both sides of the particle.

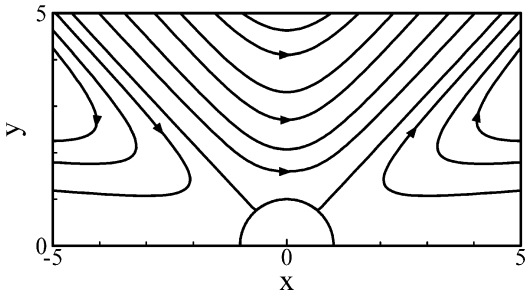

Fig. 7. Streamline contour for ψ_1.

Fig. 7 shows several streamlines plotted by using ψ_1 in Eq. (57). Far from the particle, $\psi_1|_{r\to\infty} = C_1 r Q_1(\eta) - (r/5) Q_3(\eta)$, in which $\psi_{1\infty}|_{r\to\infty} = 0$ at $\eta = \sqrt{(2C_1+1)/5}$. These angles correspond to $\theta = 47.4°$ and $137.4°$. As discussed in Fig. 6, the effective body force $\mathbf{b}_1$ directs to the right at the top of the particle. This force generates the rightward fluid motion around the top of the particle. At both RHS and LHS of the particle, however, the force directs to the left. In the vicinity of LHS and RHS of the particle, these forces accelerate the fluid to the left. Accordingly, the flow driven to the right at the top of the particle collides with the flow driven to the left at both sides, somewhere over the particle. The two nearly straight streamlines, touching the particle surface, correspond to those representing $\psi_{1\infty}|_{r\to\infty} = 0$.

4.3. Force acting on the particle

The EHD contribution to the CP force in Eq. (18) is nondimensionalized and is further decomposed to pressure f_p and the viscous contributions f_v as follows:

$$f^h = \frac{F^h}{\varepsilon_f E_0^2 a^2} = f_p + f_v,$$

where

$$f_p = -2\pi\lambda \int_0^\pi p_{1s} \cos\theta \sin\theta \, d\theta = -2\pi\lambda \int_{-1}^{1} p_{1s}\, \eta \, d\eta, \tag{58}$$

$$f_v = 2\pi\lambda \int_{-1}^{1} \left[2\eta \frac{\partial u_{r1}}{\partial r}\bigg|_{r=1} - \sqrt{1-\eta^2}\,\tau_{s1}\right] d\eta. \tag{59}$$

In the above, p_{1s} and τ_{s1} are the (dimensionless) first-order pressure and skin friction at particle surface, both of which are nondimensionalized by $\varepsilon_f E_0^2$.

For the evaluation of each component, we derive the expressions for p_{1s} and τ_{s1}. If we substitute Eq. (51) into the reduced momentum equation of Eq. (50), the gradient of

EHD pressure in θ direction becomes

$$\frac{\partial p_1}{\partial \theta} = -\frac{1}{\sqrt{1-\eta^2}}\frac{\partial E^2\psi_1}{\partial r} + r\mathbf{B}\cdot\mathbf{e}_\theta. \tag{60}$$

We decompose the pressure to $p_1 = p_1^{(h)} + p_1^{(\mathrm{el})}$, where the first and the second terms in RHS of the preceding equation are associated with the hydrodynamic pressure $p_1^{(h)}$ and the electrical pressure $p_1^{(\mathrm{el})}$, respectively. Note that $p_1^{(\mathrm{el})}$ represents the pressure induced by the concentration polarization, which is unaffected by fluid motion. The two contributions are integrated separately. We integrate the first term, by using the following relation:

$$E^2\psi_1 = \left[-\frac{2C_1}{r} + \frac{1}{2r^4} + \frac{1}{30r^7}\right]Q_1(\eta) + \left[\frac{12}{5r} - \frac{10C_3}{r^3} + \frac{3}{2r^4} + \frac{3}{110r^7}\right]Q_3(\eta).$$

Then, it becomes

$$p_{1s}^{(h)} = \left(-\frac{113}{2640} + C_1 - \frac{15C_3}{4}\right)\eta + \left(-\frac{315}{176} + \frac{25C_3}{4}\right)\eta^3 + p_{1s}^{(h)}(\eta=0). \tag{61}$$

Substituting $\mathbf{B}_1$ in Eq. (55) into Eq. (50), we obtain the following equation for the pressure due to the concentration polarization:

$$\frac{\partial p_1^{(\mathrm{el})}}{\partial \theta} = \left[r + \frac{1}{r^2} + \frac{1}{4r^5} + \left(-\frac{3}{r^2} + \frac{3}{4r^4}\right)\eta^2\right]\mathbf{E}_{e0}\cdot\mathbf{e}_\theta.$$

Then, by integrating the preceding equation, we get

$$p_{1s}^{(\mathrm{el})} = \frac{27}{8}\eta - \frac{9}{8}\eta^3 + p_{1s}^{(\mathrm{el})}(\eta=0). \tag{62}$$

Therefore, the total surface pressure on the particle surface $p_{1s} = p_{1s}^{(h)} + p_{1s}^{(\mathrm{el})} + p_1(\eta=0)$ becomes

$$p_{1s} = \left(\frac{8797}{2640} + C_1 - \frac{15C_3}{4}\right)\eta + \left(-\frac{513}{176} + \frac{25C_3}{4}\right)\eta^3 + p_{1s}(\eta=0). \tag{63}$$

Strange as may sound, the (dimensional) pressure is not dependent on fluid viscosity, although the flow itself is a kind of friction-dominating flow. This is because of the peculiar feature of the creeping flow. For creeping flows, the pressure as well as the shear stress is in general proportional to a product of the viscosity and characteristic velocity. However, the velocity which is driven by the Coulombic body force is inversely proportional to the viscosity, e.g., $u_c = \varepsilon_f E_0^2 a/\mu$. Therefore, the pressure becomes independent of the viscosity and is proportional to the driving force $(\varepsilon_f E_0^2)$. Consequently, the resulting force is also independent of viscosity. As will be shown, the same is true for the shear stress and the resulting force.

The pressure is integrated over the particle surface as $f_p^{(h,\mathrm{el})} = -2\pi\lambda\int_{-1}^{1} p_{1s}^{(h,\mathrm{el})}\eta\,\mathrm{d}\eta$ to obtain the following forces:

$$f_p^{(h)} = 2\pi\lambda\left(\frac{67}{90} - \frac{2C_1}{3}\right) = \frac{589}{420}\pi\lambda, \tag{64}$$

$$f_p^{(\mathrm{el})} = -\frac{18}{5}\pi\lambda. \tag{65}$$

Note that the force directly induced by the concentration polarization $f_p^{(\mathrm{el})}$ directs to the negative x-direction while the hydrodynamic pressure force $f_p^{(h)}$ acts to the positive x-direction. The total pressure force acting on the particle is

$$f_p = f_p^{(h)} + f_p^{(\mathrm{el})} = -\frac{923}{420}\pi\lambda. \tag{66}$$

Let us consider the viscous stress. The skin friction on the particle surface is reduced to $\tau_{s1} = \partial u_{1\theta}/\partial r|_{r=1}$, due to the no-slip condition. If we represent $u_{1\theta}$ by the steam function, we obtain

$$\tau_{s1} = \frac{1}{\sqrt{1-\eta^2}}\left\{\left(-\frac{25}{24} + C_1 - 3D_1\right)Q_1 + \left(\frac{1153}{660} - 3C_3 - 15D_3\right)Q_3\right\}. \tag{67}$$

The integral $\int_{-1}^{1} 2\eta(\partial u_{r1}/\partial r)_{r=1}\,\mathrm{d}\eta$ in f_v, in Eq. (59), vanishes. Then, the force due to the skin friction is obtained from Eq. (59) as follows:

$$f_v = 2\pi\lambda\left(\frac{25}{36} - \frac{2C_1}{3} + 2D_1\right) = \frac{113}{210}\pi\lambda. \tag{68}$$

The dielectric contribution to the force can be obtained by performing the integration in Eq. (17) and using the above-derived expressions for $\mathbf{E}_e$ and $\mathbf{E}_i$. The force in Eq. (17) is nondimensionalized and is further decomposed to

$$f^M = \frac{F^M}{\varepsilon_f E_0^2 a^2} = f_e^M + f_i^M.$$

Here, f_e^M and f_i^M represent the contribution from the exterior and interior electric field, respectively, and become

$$f_e^M = 2\pi\lambda\int_{-1}^{1}\left[\eta T_{e,rr} - \sqrt{1-\eta^2}T_{e,r\theta}\right]\mathrm{d}\eta,$$

$$f_i^M = -2\pi\lambda\int_{-1}^{1}\left[\eta T_{i,rr} - \sqrt{1-\eta^2}T_{i,r\theta}\right]\mathrm{d}\eta,$$

where $T_{e,rr} = (1/2)(E_{e,r}^2 - E_{e,\theta}^2)$, $T_{e,r\theta} = E_{e,r}E_{e,\theta}$, $T_{i,rr} = (\varepsilon_p/2\varepsilon_f)(E_{i,r}^2 - E_{i,\theta}^2)$, and $T_{i,r\theta} = (\varepsilon_p/\varepsilon_f)E_{i,r}E_{i,\theta}$. Since $E_{e,r} = \mathbf{E}\cdot\mathbf{e}_r = 0$, it becomes $T_{e,rr} = -(1/2)E_{e,\theta}^2$ and $T_{e,r\theta} = 0$.

Fig. 8 shows the traction vector of the Maxwell stress $(\mathbf{T}\cdot\mathbf{n})$ computed at the particle surface. The figure is plotted by computing $T_{e,rr}$, $T_{e,r\theta}$, $T_{i,rr}$, and $T_{i,r\theta}$ by using the electric field obtained earlier in Eqs. (47) and (49). The stress from the exterior electric field (filled arrows) has only radial

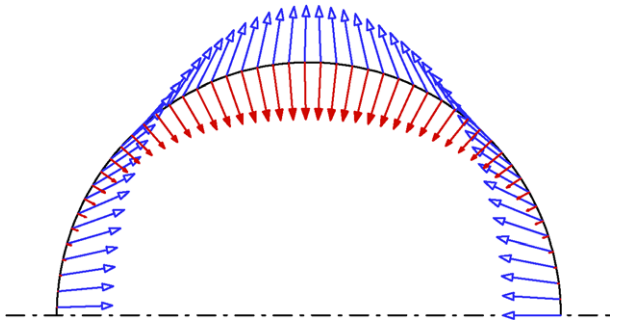

Fig. 8. Distribution of the traction vector of the Maxwell stress ($\mathbf{T}\cdot\mathbf{n}$) on the particle surface. Filled (in a red line) and hollow (in a blue line) arrows represent the stress due to the exterior and interior electric field, respectively. The electrical permittivities are set equal, i.e., $\varepsilon_f = \varepsilon_p$. (Color drawing in the online version.)

component which is generally greater at RHS of the particle. If we perform integration for the interior surface, it becomes $f_i^M = 0$, and the total force due to the Maxwell stress becomes

$$f^M = f_e^M = -\frac{9}{5}\pi\lambda. \tag{69}$$

It should be noted that, to the first order of λ, f^M is independent of hydrodynamic field. Additionally, f^M is independent of the electrical permittivity of the particle, which is consistent to the DEP force in a conducting medium [17]. This force is different from the conventional DEP force in a dielectric medium in that this force does not vanish even when $\varepsilon_f = \varepsilon_p$.

The total force acting on the particle is the sum of the pressure force, the frictional force, and the dielectric force, i.e.,

$$f_{\mathrm{CP}} = f^h + f^M = f_p^{(\mathrm{el})} + f_p^{(h)} + f_v + f_e^M + f_i^M.$$

Finally, the nondimensional CP force becomes

$$f_{\mathrm{CP}} = \pi\lambda\left(-\frac{13}{18} - \frac{8C_1}{3} + 4D_1 - \frac{9}{5}\right) = -\frac{1453}{420}\pi\lambda. \tag{70}$$

Since $\lambda = ka/c_0$ and $k = -\nabla c_\infty$, the CP force can be expressed in a dimensional form as

$$F_{\mathrm{CP}} = \frac{1453}{420}\pi a^3\varepsilon_f E_0^2\left(\frac{\nabla c_\infty}{c_0}\right). \tag{71}$$

If we define $i = \sigma_0 E_0$ as the current that is constant throughout the domain, the above expression can be alternatively expressed, in terms of current, as

$$F_{\mathrm{CP}} = \frac{1453}{420}\pi a^3\varepsilon_f i^2\frac{\nabla\sigma_\infty}{\sigma_0^3}. \tag{72}$$

The direction of the CP force is determined by the concentration gradient, and is always towards the region of the higher ionic concentration. For a given concentration (conductivity) gradient, the CP force is inversely proportional to the cube of local concentration. For the future comparison with the conventional DEP force, we change the above form to a similar form to the conventional DEP force. Since $\sigma_\infty E_\infty$ is constant, it becomes $E_\infty\nabla c_\infty = -c_\infty\nabla E_\infty$. Introducing the preceding relation to Eq. (71) and noting that $c_\infty(x=0) = c_0$ and $E_\infty(x=0) = E_0$, we obtain

$$F_{\mathrm{CP}} = -2\pi\varepsilon_f a^3 K\nabla E_\infty^2, \tag{73}$$

where $K = 1453/210$.

Table 2
Relative contribution of each component to the CP force

Contributions	Symbol	f/f_{CP}
Pressure (electrical)	$f_p^{(\mathrm{el})}$	1.04
Pressure (viscous)	$f_p^{(h)}$	−0.41*
Skin friction	f_v	−0.16
Maxwell stress (exterior)	f_e^M	0.52
Maxwell stress (interior)	f_i^M	0
Sum		1

* Minus sign represents the direction of the component opposite to the CP force.

Table 2 shows the relative contribution of each component to the total CP force. In the table, the minus sign represents the direction of the force component opposite to that of total CP force. The pressure force directly induced by the concentration polarization, $f_p^{(\mathrm{el})}$, is the largest contribution among the components of the CP force. The dielectrophoretic contribution, $f_e^M + f_i^M$ in which $f_i^M = 0$, is just half of that due to the electrical pressure contribution. In the present investigation, we assumed that the conductivity of the particle is zero. In reality, the particle has a finite conductivity, although it may be much smaller than that of electrolytes. If the particle has a finite conductivity, some current may flow thought the particle. Then, the strength of the electric field around the particle may be decreased, and the overall magnitude of the CP force may be reduced.

4.4. Numerical verifications

A numerical analysis is performed to verify the analytical results. In the numerical analysis, a set of coupled equations, Eqs. (31)–(41), were solved by using the finite volume method. A spherical surface that has its origin at the center of the particle and has the radius of $100a$ is chosen as the outer boundary surface, in which the particle surface forms the inner boundary. The no-slip condition is applied at the outer boundary surface. In the simulation, λ and Pe_{el} are chosen as 10^{-3} and 1, respectively.

Fig. 9 compares the analytical and the numerical results for the pressure and the wall-shear stress on the particle surface, which show a fairly good agreement. In the figure, $p_{1s}(\eta = 0)$ in Eq. (63) is set to zero. Due to the pumping action of the particle, a slow but large-scale circulating flow is generated inside the numerical domain. However, the flow field, the pressure in the vicinity of the particle, and the wall-shear stress on the particle surface are hardly influenced by the large-scale circulating flow.

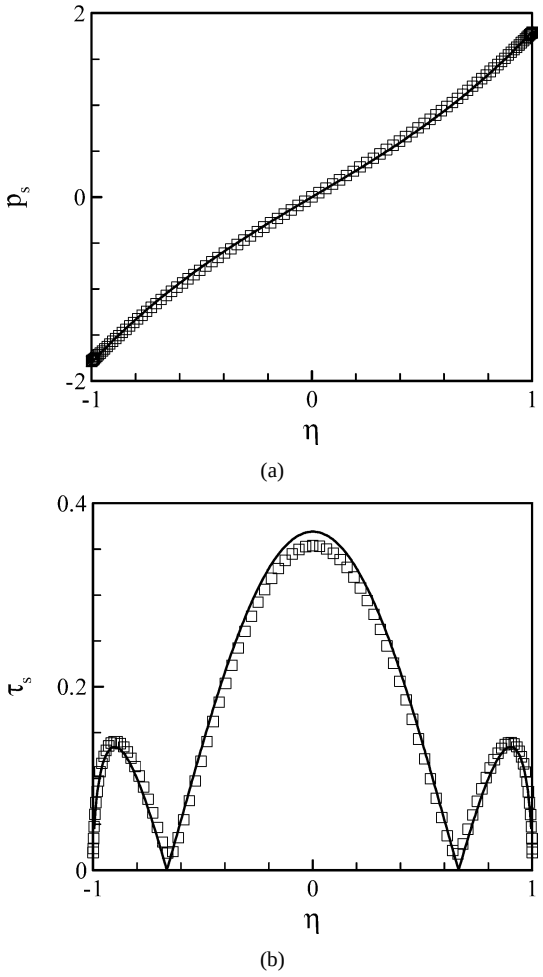

Fig. 9. Comparison of the analytical and the numerical results for (a) pressure (b) wall-shear stress. Solid line represents the analytical result, and the symbols are for the numerical results. These stresses are scaled by $\varepsilon_f E_0^2$, where $E_0 = E_\infty(x = 0)$.

The high value of λ essentially requires a consideration for boundary surfaces (refer to Fig. 4). Additionally, for large Peclet numbers, the effect of hydrodynamic flow will become more significant; consequently, the boundary effect on flow would be much pronounced due to the slow decaying character of the creeping flow. In the present investigation, we developed an analytical result that is independent of the geometrical shape of boundary surfaces. The goal of the present numerical analysis is limited to validation of analytical results. Investigating the effect of boundary surface is necessary in the future.

5. Discussions

The translating velocity (U_{CP}) of the particle due to the CP force can be obtained by balancing the CP force with the Stokes force ($F_s = 6\pi\mu a U_{\mathrm{CP}}$) [8], which yields

$$U_{\mathrm{CP}} = \frac{|F_{\mathrm{CP}}|}{6\pi\mu a} = \frac{1453}{2520}\frac{a\varepsilon_f E_0^2\lambda}{\mu}. \tag{74}$$

The influence of the convective transport of species due to U_{CP} can be represented by the (hydrodynamic) Peclet number defined as $Pe = aU_{\mathrm{CP}}/D$. From the preceding equation and the definition of the electrical Peclet number, i.e., $Pe_{\mathrm{el}} = \varepsilon_f E_0^2 a^2/(\mu D)$, the Peclet number can be related to the electrical Peclet number as

$$Pe = \frac{1453}{2520} Pe_{\mathrm{el}}\lambda.$$

Since $0 < Pe_{\mathrm{el}}\lambda \ll 1$, it confirms that the effect of translation-induced convective transport on the concentration field can be neglected within the validity of the present theory. Even if we take into account of the translating velocity in the analysis from the beginning, its effect appears at best in the second order terms.

Let us consider the effect of the applied electric-field direction on the direction of the CP force. Assume, in Fig. 1, the direction of the external electric field is reversed while maintaining the concentration gradient as before. Then, only the sign of the electric field will be changed. Since the charge density is related to the concentration and the electric field as $\rho^f = -\varepsilon\nabla\sigma\cdot\mathbf{E}/\sigma$, the sign of free charge due to concentration polarization is also reverse. For the particular case shown in Fig. 1, then, negative charges will be generated. There is no change, however, in the Coulombic body force ($\rho_e\mathbf{E}$), and eventually, the hydrodynamic field and the hydrodynamic force acting on the particle will not change. In the Maxwell stress, every component is in the form of square of electric field. The sign change of the electric field, therefore, does not affect the resulting force. As to the direction of the concentration gradient, as explained before, the CP acts towards the region of higher concentration. Consequently, the CP force always directs to the region of "higher" concentration, irrespective of the direction of **E** or **i**.

In principle, the linear theory presented in this paper is applicable to cases of small effective concentration gradients and small electrical Peclet numbers Pe_{el}. It is desirable to compare the magnitude of the CP force with other forces on the basis of the present linear theory. Here, we compare the CP force with the DEP, the electrophoretic force, and the gravity forces.

The dielectrophoretic force for a conducting particle submerged in a conducting liquid is [17]

$$F_{\mathrm{DEP}} = 2\pi\varepsilon_f a^3 K_{\mathrm{CM}}\nabla E_\infty^2,$$

where $K_{\mathrm{CM}} = (\sigma_p - \sigma_f)/(\sigma_p - 2\sigma_f)$ is the Clausius–Mossotti factor in this case. For a limiting condition of a nonconducting particle, it becomes $K_{\mathrm{CM}} = -1/2$, and the relative magnitude of the DEP force relative to the CP force becomes

$$\left|\frac{F_{\mathrm{DEP}}}{F_{\mathrm{EP}}}\right| = \left|\frac{K_{\mathrm{CM}}}{K}\right| = \frac{105}{1453}. \tag{75}$$

As shown, the above comparison shows that the CP force is ten times greater than the DEP force under the same electric-field gradient. However, for the CP force the electrical-field gradient is produced due to the concentration gradient,

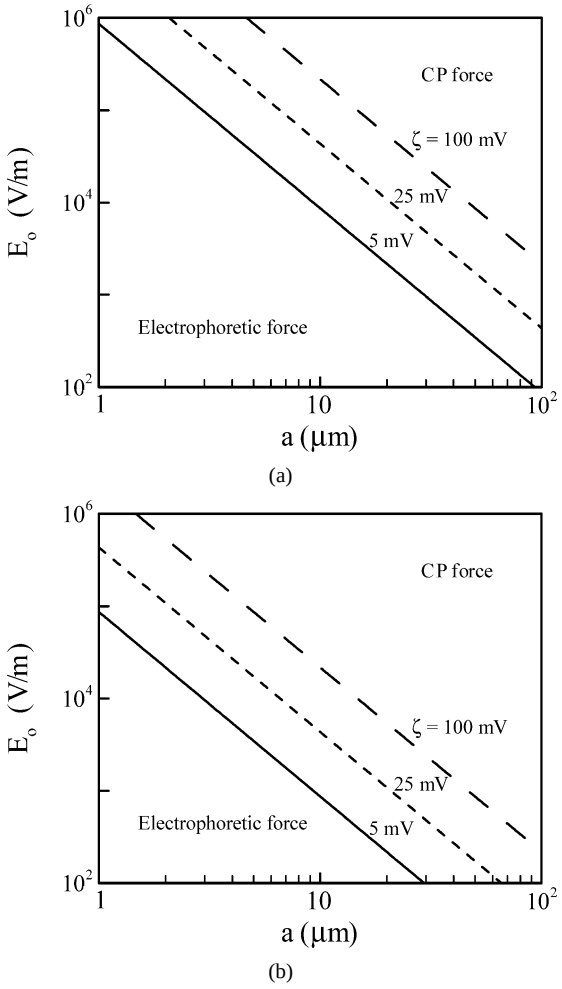

Fig. 10. Comparison of magnitude of the CP force and the electrophoretic forces: (a) $|k|/c_0 = 10^4\ \mathrm{m}^{-4}$; (b) $|k|/c_0 = 10^5\ \mathrm{m}^{-4}$.

while for the DEP force, the field gradient is independent of the concentration gradients, and determined by the difference in electrodes' geometries and distance, etc. Thus, the above comparison does not mean that the CP force is always greater than the DEP force.

Under a DC field, the CP force shows very similar characteristics to those of the DEP force. Alike the DEP force, as explained earlier, the direction of the CP force does not depend on the polarity of the electric field. Both are proportional to the square of the electric-field strength and volume of a body. These characteristics have a potentially important implication. That is, analogous to the DEP force, the CP force may have a nonzero value for even an AC field.

The electrophoretic force acting on a particle that has a zeta potential of ζ can be represented by [8]

$$F_{\mathrm{EP}} = 6\pi\zeta\varepsilon_f E_0 a.$$

Then, the ratio of the electrophoretic force to the CP force becomes

$$\left|\frac{F_{\mathrm{CP}}}{F_{\mathrm{EP}}}\right| = \frac{1453}{2520}\frac{a^2 E_0|k|}{c_0\zeta}.$$

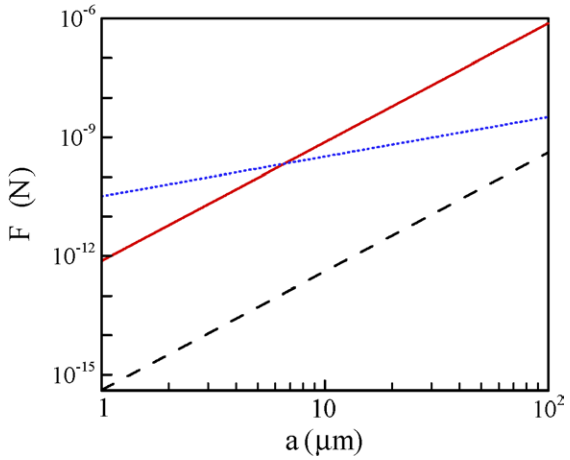

Fig. 11. Comparison of magnitude of forces for $E_0 = 10^5$ V/m, $\zeta = 25$ mV, $|k|/c_0 = 10^4\ \mathrm{m}^{-4}$, $g = 9.81\ \mathrm{m}^2/\mathrm{s}$, $\rho_p = 1.010\ \mathrm{kg/m}^3$, $\rho = 998\ \mathrm{kg/m}^3$, and $\varepsilon_f = 6.9 \times 10^{-10}\ \mathrm{C}^2/(\mathrm{J\,m})$. The solid, dotted, and dashed lines correspond to those of the CP force, the electrophoretic force, and the gravity force, respectively.

The two forces may have the same magnitude when

$$E_0 = \frac{2520}{1453}\left(\frac{c_0\zeta}{|k|}\right)\frac{1}{a^2}, \tag{76}$$

where $k = -\nabla c_\infty$. In Figs. 10a and 10b, the above relation is plotted for $|k|/c_0 = 10^4\ \mathrm{m}^{-4}$ and $10^5\ \mathrm{m}^{-4}$, respectively. Fig. 10 represents the region where the CP force and the electrophoretic force become greater than the other. Such values of $|k|/c_0$ may be typical in microchannel environments. For instance, consider the case considered in Section 3.2, in which two streams of different electrolyte concentration of 50 $\mathrm{mol/m}^3$ (=50 mM) and 1 $\mathrm{mol/m}^3$ (=1 mM) flowing in parallel in a microchannel having a width of 100 μm. The diffusional mixing at the interface will generate a concentration gradient. Then, k may be in the order of $5 \times 10^5\ \mathrm{mol/m}^4$, and consequently, $|k|/c_0$ may be in the order of $2 \times 10^4\ \mathrm{m}^{-4}$. When $a = 10$ μm, the value of $|k|/c_0 = 10^4\ \mathrm{m}^{-4}$ corresponds to $\lambda = 0.1$. In the figure, the CP force is greater at the upper region of each line, and the electrophoretic force is greater at the lower region.

The CP force is proportional to a^3 while the electrophoretic force is proportional to a. Thus, as shown in Fig. 10, as the size of a particle increases, the relative magnitude of the CP force will increase compared to the electrophoretic force. Additionally, the CP force is proportional to the square of the electric-field strength while the electrophoretic force is directly proportional to the electric-field strength. Thus, the CP force may become more important for high electric fields compared to the electrophoretic force. It should be emphasized that the CP force exists even for an electrically neutral particle. Additionally, the AC field does not generate any electrophoretic force while the CP force under an AC field may still exist.

Fig. 11 compares the absolute magnitude of the CP force with that of electrophoretic force and the gravity force $F_g = (4/3)\pi a^3(\rho_w - \rho_p)g$ in which ρ_p is the particle density. It

is chosen so that $E_0 = 10^5$ V/m, $\rho_p = 1,010$ kg/m^3, $\zeta = 25$ mV, and $|k|/c_0 = 10^4$ m^{-4}. As shown, the CP force is greatest among the three forces when the particle size ranges from 7 to 100 μm, although the validity of the present theory for a large particle is not assured.

To avoid any complication that could arise from the conventional electrokinetic effects, we assumed that the particle has no surface charge. In practice, however, most of particles have surface charges. It is believed that, at least to the leading order, the surface charges may not significantly affect the magnitude of the CP force. The reason is that the electrical double layer on the particle surface may screen the electric field induced by the surface charge. Therefore, the exterior and interior electric field may not be significantly distorted from the state without the surface charge, and subsequently, the leading order term of the CP force will be hardly influenced. For a charged particle, therefore, the CP force is additional to the electrophoretic force.

6. Summary

A particle submerged in an electrolyte solution experiences a new kind of force, the concentration-polarization (CP) force. The CP force in a DC electric field originates from two different mechanisms: (i) the gradient of EHD pressure which is developed by the Coulombic force acting on "induced" free charges, and (ii) the dielectric force due to nonuniform electric field induced by the conductivity gradient.

The CP force is proportional to the square of the applied electric field and acts towards the region of higher ionic concentration, regardless of direction of the electric field. The CP force is effective on electrically neutral (uncharged) particles, which may have practical significance to handle particles having same electrokinetic properties. The present investigation also suggests that the concentration gradient can become a means to generate a DEP-like force.

The magnitude of the CP force is compared to the electrophoretic, the convectional DEP, and the gravity forces. It demonstrates, within the validity of the present linear theory, that the magnitude of the CP force is at least comparable to the electrophoretic force for particles having radius of tens of micrometer. The CP force is ten times greater than the DEP force for a given value of ∇E_∞^2.

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