Dielectrophoresis of a deformable fluid particle in a nonuniform electric field

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In nonuniform electric fields, the dielectrophoretic behavior and the interface shape of uncharged fluid particles freely suspended in an immiscible fluid may be significantly influenced by the induced electrohydrodynamic flow. This work presents a theoretical investigation of the primary effects of electrohydrodynamic flow associated with the dielectrophoresis of a deformable fluid particle with a leaky dielectric model. The electrohydrodynamic flow is shown to either enhance or hinder the dielectrophoretic motion of fluid particles. A variety of shapes of deformable fluid particles are predicted. [S1063-651X(96)03410-1]

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In a nonuniform electric field, an uncharged particle may be subjected to a dielectrophoretic force. The dielectrophoresis of particles is important in a variety of applications including biophysics, bioengineering, multiphase separation, among others [1]. When the particles of concern are fluid drops or bubbles, the fluid interface between the particle and surrounding medium can be mobile as well as deformable. If finite electrical conductivity of the dielectric fluids comes into play, local charge accumulation at the mobile fluid interface can induce a tangential electric stress that drags fluids into motion [2]. The leaky dielectric theory put forward by Taylor [3] has been successful in describing the electrohydrodynamic flow driven by the tangential electric stress around an uncharged fluid drop in a uniform electric field (see also [4–6]). Theoretical analyses of the dielectrophoresis of uncharged drops and bubbles in nonuniform fields, however, have not considered the effects of the electrohydrodynamic flow and particle shape deformations [1]. To elucidate the primary effects of electrohydrodynamic flow in the dielectrophoresis of deformable fluid particles, such as drops and bubbles, this work presents a theoretical analysis of the poorly understood phenomena with a leaky dielectric model.

The key ingredients in the leaky dielectric model, as recognized by Taylor [3], are the finite electrical conductivity and viscosity of the fluids. Accounting for the fluid conductivity in the theory need not be sophisticated; constant Ohmic conductivity in each fluid phase can be sufficient to induce tangential electric stress at the particle interface. The electrohydrodynamic flow, due to the tangential electric stress, is a consequence of the fluid viscosities that transfer momentum from the interface, where the driving force resides, to bulk fluids. Many functional forms may be used to describe fluid viscosities [7]; among all the possibilities, Newtonian fluids of constant viscosities may be considered as a simple yet adequate first approximation in the leaky dielectric theory. Even for Newtonian fluids, nonlinearities may appear in general governing equations because of the fluid inertia and the capillarity of deformable fluid interfaces. The nonlinear behavior of significantly deformed leaky dielectric drops in a uniform electric field was investigated by Feng and Scott [6] with finite element computations. Nonlinearities may also arise from the charge convection by fluid flow at the interface, which have not been rigorously investigated in existing theoretical work (see the discussions in Refs. [4–6]). To understand the basic features of the electrohydrodynamic flow in the dielectrophoresis of deformable fluid particles, the theoretical model considered here is kept simple by neglecting all nonlinear effects. As demonstrated by Feng and Scott [6], the inclusion of nonlinearities in the numerical computations may provide more accurate results than a linearized asymptotic analysis. Appropriate procedures for computing the solutions of the nonlinear equations may also be used to predict the critical electric field strength when the fluid particle becomes unstable, when the mathematical singular points are evaluated in parameter space. Nevertheless, analytical solutions of the linearized problem can offer important physical insights into the primary electrohydrodynamic effects and provide invaluable guidance to more comprehensive numerical computations (cf. [6]).

The problem of concern here consists of a fluid particle of volume $(4/3)\pi a^3$, density ρ_i, viscosity μ_i, conductivity σ_i, and dielectric constant κ_i immersed in an unbounded immiscible fluid of density ρ_o, viscosity μ_o, conductivity σ_o, and dielectric constant κ_o. The deformable interface separating the particle from its surrounding fluid has constant interfacial tension γ. In what follows, the subscripts i and o denote the variables associated with the fluids inside and outside the particle, respectively; the variables without subscripts i and o are applicable to either fluid. To illustrate the most significant effects of local charge accumulation at the particle interface, the present analysis only considers a dc electric field and associated axisymmetric steady states. In spherical coordinates $(r,\ \theta)$ with the origin fixed at the particle's center of mass, the externally applied electric field can be described in terms of the electric potential V ($\underline{\mathbf{E}} = -\boldsymbol{\nabla} V$) which is assumed to have the form

$$V = -E_0[rP_1(\zeta) + \Lambda r^2 P_2(\zeta)], \tag{1}$$

where $\zeta \equiv \cos\theta$ and $P_l(\zeta)$ denotes the Legendre polynomials. The factor Λ is a measure of the relative magnitude of the field nonuniformity. The applied field given by (1) could be the simplest possible nonuniform electric field for dielectrophoresis [8].

For homogeneous fluids with constant physical properties, the electric potential distributions inside and outside a spherical particle are found to be

$$V_i(r,\theta)=-E_0\left[\frac{3}{2+R}rP_1(\zeta)+\Lambda\frac{5}{3+2R}r^2P_2(\zeta)\right], \quad (2)$$

and

$$V_o(r,\theta)=-E_0\left[\left(r+\frac{1-R}{2+R}\frac{a^3}{r^2}\right)P_1(\zeta)+\Lambda\left(r^2+2\frac{1-R}{3+2R}\frac{a^5}{r^3}\right)P_2(\zeta)\right]. \quad (3)$$

Equations (2) and (3) satisfy the Laplace equation and all the boundary conditions, such as that given by (1) in the far field, as well as the continuity of the tangential component of the electric field and that of the normal component of the electrical current density ($\sigma\underline{\mathbf{E}}$) at the fluid interface $r=a$. Here $R\equiv\sigma_i/\sigma_o$ is the conductivity ratio. Consequently, the steady surface charge density distribution on the fluid interface is given by

$$q_s(\theta)=\epsilon_0(\kappa_oR-\kappa_i)E_0[\eta_1P_1(\zeta)+2\eta_2P_2(\zeta)] \quad (4)$$

as consistent with the jump in the normal component of the electric displacement vector $\epsilon_0\kappa\underline{\mathbf{E}}$ at $r=a$, with $\eta_1\equiv3/(2+R)$, $\eta_2\equiv5\Lambda a/(3+2R)$, and ϵ_0 denoting the permittivity of free space. Clearly, the overall particle is electrically neutral even though the local charge density is nonzero around the interface. These nonzero local surface charges can be acted upon by the tangential component of electric field at the fluid interface to generate a tangential electric stress (cf. Ref. [2,3]), which can be written as

$$q_sE_\theta=\epsilon_0(\kappa_oR-\kappa_i)E_0^2\left[\eta_1^2P_1\frac{\partial P_1}{\partial\theta}+\eta_1\eta_2\left(P_1\frac{\partial P_2}{\partial\theta}+2P_2\frac{\partial P_1}{\partial\theta}\right)+2\eta_2^2P_2\frac{\partial P_2}{\partial\theta}\right]. \quad (5)$$

Because the fluid interface cannot support tangential stresses, the tangential electric stress must be offset by the viscous stresses arising from the induced electrohydrodynamic flow.

Without omitting the salient features in dielectrophoresis, the effects of the fluid inertia and particle shape deformation are neglected for the moment. Thus the primary electrohydrodynamic flow field can be determined by solving a linear fourth-order equation for the stream function [9]

$$\left[\frac{\partial^2}{\partial r^2}+\frac{\sin\theta}{r^2}\frac{\partial}{\partial\theta}\left(\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\right)\right]^2\psi=0$$

that yields

$$\psi_i(r,\theta)=\frac{(\kappa_oR-\kappa_i)\epsilon_0E_0^2}{\mu_o+\mu_i}\sum_{l=2}^{5}\beta_l\frac{(r^{l+2}-a^2r^l)}{a^{l-1}}G_l(\zeta), \quad (6)$$

and

$$\psi_o(r,\theta)=\frac{(\kappa_oR-\kappa_i)\epsilon_0E_0^2}{\mu_o+\mu_i}\sum_{l=2}^{5}\beta_l\left(\frac{1}{r^{l-3}}-\frac{a^2}{r^{l-1}}\right)a^lG_l(\zeta), \quad (7)$$

where $G_l(\zeta)$ are the Gegenbauer functions of the first kind [9] and $\beta_2\equiv\eta_1\eta_2/15$, $\beta_3\equiv\eta_1^2/5+6\eta_2^2/35$, $\beta_4\equiv24\eta_1\eta_2/35$, $\beta_5\equiv4\eta_2^2/7$. The solutions (6) and (7) satisfy the traction boundary condition for the tangential stresses at a spherical fluid interface and the natural boundary conditions that require the boundedness of the flow field at infinity and coordinate origin. With the relationship between the flow velocity $\underline{\mathbf{u}}$ and the stream function given by

$$u_r=\frac{1}{r^2\sin\theta}\frac{\partial\psi}{\partial\theta}$$

and

$$u_\theta=-\frac{1}{r\sin\theta}\frac{\partial\psi}{\partial r},$$

the corresponding tangential flow velocity along the fluid interface, driven by the tangential electric stress, can be derived as

$$u_\theta=\frac{(\kappa_oR-\kappa_i)\epsilon_0E_0^2a}{35(\mu_o+\mu_i)}\left[\frac{\eta_1\eta_2}{3}\sin\theta(90\cos^2\theta-11)+\eta_2^2\sin\theta\cos\theta\left(35\cos^2\theta-9+\frac{7\eta_1^2}{\eta_2^2}\right)\right]. \quad (8)$$

The first term in (8) represents the asymmetric flow with respect to the particle's equatorial plane, which is associated with a net force. The fact that the asymmetric flow is proportional to $\eta_1\eta_2$ shows that both the P_1 and P_2 components in the applied electric field are necessary for generating a net force. In contrast, the second term in (8) consists of components associated with either η_1^2 or η_2^2, indicating that the symmetric flow may appear in any configuration of the applied electric field. Taylor's results for the electrohydrodynamic flow about a drop in a uniform field [3] are recovered at $\Lambda=0$ in the more general results presented here.

To quantitatively calculate the contributions of electrohydrodynamic flow to the dielectrophoretic force as well as the fluid interface deformation, the normal stress balance at the fluid interface must be evaluated. For convenience, the problem is considered in a reference frame with the coordinate origin fixed at the particle's center of mass. Thus the motion of the particle is equivalently represented by a uniform flow velocity at infinity [10]. The steady motion of the particle can be readily accounted for by adding a term for uniform flow past a spherical fluid particle at zero Reynolds number [9] in (6) and (7). Moreover, the buoyant force due to gravity is assumed to be aligned with the applied electric field so that axisymmetry is preserved. Thus the equation for the normal stress balance at the fluid interface takes the form

$$\underline{\mathbf{n}}\,\underline{\mathbf{n}}:[(\underline{\underline{\mathbf{T}}}+\underline{\underline{\mathbf{T}}}^E)_o-(\underline{\underline{\mathbf{T}}}+\underline{\underline{\mathbf{T}}}^E)_i]=\frac{\gamma}{a}\boldsymbol{\nabla}\cdot\underline{\mathbf{n}}, \quad (9)$$

where $\underline{\mathbf{n}}$ is the outward unit normal vector at the fluid interface, $\underline{\underline{\mathbf{T}}}=-p\underline{\underline{\mathbf{I}}}+\mu[\boldsymbol{\nabla}\underline{\mathbf{u}}+(\boldsymbol{\nabla}\underline{\mathbf{u}})^T]$ and $\underline{\underline{\mathbf{T}}}^E=\epsilon_0\kappa(\underline{\mathbf{E}}\,\underline{\mathbf{E}}-(1/2)\underline{\mathbf{E}}\cdot\underline{\mathbf{E}}\,\underline{\underline{\mathbf{I}}})$ [2] with superscript T standing for the transpose and $\underline{\underline{\mathbf{I}}}$ the identity tensor. The term on the right side of (9) is the capillary force due to fluid interface curvatures.

Each term in (9) can be written as an expansion in P_l where l ranges from 0 to 4, with the terms on the left side approximated by the results for spherical particles and the capillary force term on the right side retaining the first-order effects of interface deformations. Only the terms associated with $l=1$ in (9) contribute to a net force, which is of primary concern in dielectrophoresis. By gathering the terms associated with P_1 in (9), the equation describing net force balance (in the direction of $\theta = 0$ [11]) can be obtained as

$$\frac{4\pi\kappa_i\epsilon_0 E_0^2 \Lambda a^3}{(2+R)(3+2R)}\left[2(2SR^2-3S+1)+(SR-1)\frac{M+2}{M+1}\right] = \frac{4}{3}\pi a^3 g(\rho_i-\rho_o)+2\pi\mu_o\mu_i a U\frac{3+2M}{1+M}, \tag{10}$$

where $S\equiv\kappa_o/\kappa_i$, $M\equiv\mu_o/\mu_i$, g is the acceleration of gravity, and U denotes the dielectrophoretic velocity. The term associated with U represents the viscous drag on a creeping spherical particle with a mobile fluid interface. In (10), the left side accounts for the forces of electric orgin with the first term arising from the normal electric stress ($\underline{\mathbf{n}}\,\underline{\mathbf{n}}:\underline{\underline{\mathbf{T}}}^E$) and the second term from electrohydrodynamic flow ($\underline{\mathbf{n}}\,\underline{\mathbf{n}}:\underline{\underline{\mathbf{T}}}$), whereas the right side represents the forces of nonelectric origin, namely, buoyancy and viscous drag. When $U = 0$, (10) describes the force balance in the situation of dielectrophoretic levitation of fluid particles against gravity [12].

The intensity of electrohydrodynamic flow increases as $R\to 0$ and vanishes as $R\to 1/S$ because of the disappearance of surface charge accumulation [see (4)]. Actually electrohydrodynamic flow also vanishes as $R\to\infty$. In this case, the fluid inside the particle is much more conductive than the surrounding fluid and the tangential component of electric field becomes zero, leading to zero tangential electric stress [see (5)] despite a substantial amount of surface charge existing at the fluid interface.

For systems with R approaching unity, dielectrophoresis becomes impossible for the solid spheres [1]. According to (10), however, the dielectrophoretic force on a fluid particle differs from that on a solid sphere by a term proportional to $M(SR-1)/(M+1)$. Hence, dielectrophoresis of fluid particles with mobile interfaces may still occur at $R=1$ provided that $\kappa_o\neq\kappa_i$ and μ_o/μ_i is not diminishing. Of course, circumstances also exist for the dielectrophoretic force to vanish on a fluid particle but remain effective on a solid particle. For two-phase fluid systems with closely matched conductivities and surrounding fluids much more viscous than that inside the particles, significant differences are expected between the dielectrophoretic motion of fluid and solid particles. If $\mu_i\gg\mu_o$, the electrohydrodynamic flow contribution to the dielectrophoretic force becomes the same as that due to the jump in the tangential electric stress at the surface of a solid sphere. Therefore, when the fluid inside the particle is much more viscous than the outside one, the dielectrophoretic motion of a spherical fluid particle resembles that of a solid sphere, despite the fundamental difference in physical mechanisms and the appearance of the electrohydrodynamic flow.

One of the fundamental differences between fluid and solid particles is the fluid interface deformation. With the fluid interface described by $r=a[1+F(\theta)]$ and the shape function written as $F(\theta)=\Sigma_{l=2}^4\alpha_l P_l(\zeta)$, the interface curvature term $\nabla\cdot\underline{\mathbf{n}}$ in (9) takes the approximate form (see, e.g., [10]) $2+\Sigma_{l=2}^4(l^2+l-2)\alpha_l P_l$ provided that $|\alpha_l|\ll 1$. Thus (9) yields

$$\alpha_2=\frac{\kappa_i\epsilon_0 E_0^2 a}{4\gamma}\left[\frac{\eta_1^2}{3}(SR^2+S-2)+\frac{\eta_2^2}{7}(4SR^2-3S-1)+\left(\eta_1^2+\frac{6\eta_2^2}{7}\right)\frac{(SR-1)(2M+3)}{5(M+1)}\right], \tag{11}$$

$$\alpha_3=\frac{\kappa_i\epsilon_0 E_0^2 a\,\eta_1\eta_2}{50\gamma}\left[6(SR^2+S-2)+\frac{12(SR-1)(3M+4)}{7(M+1)}\right], \tag{12}$$

$$\alpha_4=\frac{\kappa_i\epsilon_0 E_0^2 a\,\eta_2^2}{105\gamma}\left[6(SR^2+S-2)+\frac{(SR-1)(4M+5)}{M+1}\right]. \tag{13}$$

With respect to the particle's equatorial plane, the symmetric shape component is represented by α_2 and α_4, whereas the asymmetric deformation is represented by α_3. Like the asymmetric component in the tangential velocity at the interface [see (8)], α_3 is proportional to $\eta_1\eta_2$. In the absence of the P_2 component of the applied field, i.e., $\Lambda\to 0$, α_3 and α_4 vanish and α_2 recovers Taylor's results [3] for drop shapes in a uniform electric field. In any configuration of the applied electric field, the prolate-oblate type of two-lobed shape deformation described by $\alpha_2 P_2(\zeta)$ may always appear. Moreover, both signs and relative magnitudes of α_l (with $l=2$, 3, 4) can be changed by varying R, S, M, and Λ; therefore, a fluid particle may exhibit a great variety of shapes in response to the stress distributions in the dielectrophoretic process.

As a result of (9) for $l=0$, the uniform excess pressure inside a fluid particle of volume $(4/3)\pi a^3$ can be determined as

$$\Delta p_0=\frac{2\gamma}{a}-\frac{\kappa_i\epsilon_0 E_0^2 a}{2}\left\{\frac{\eta_1^2}{3}(SR^2-2S+1)+\frac{2\eta_2^2}{5}(2SR^2-3S+1)+\frac{SR-1}{5(M+1)}\left[\left(\eta_1^2+\frac{6\eta_2^2}{7}\right)\times(2M-7)-\frac{\eta_2^2}{14}(84M-165)\right]\right\}. \tag{14}$$

For liquid drops immersed in an immiscible fluid, Δp_0 may not cause any noticeable physical consequences due to the material incompressibility. For gas bubbles in liquids, however, Δp_0 is intimately related to the bubble volume which may be of practical concern in many applications.

The analysis of this work provides the predictions of the primary effects of electrohydrodynamic flow driven by the tangential electric stress in the dielectrophoresis of a fluid particle and the nature of electrified fluid interface deformations. In general, for systems with $(R-1)(SR-1)<0$, a fluid particle is subjected to a stronger dielectrophoretic force than a solid sphere due to the electrohydrodynamic flow. If $(R-1)(SR-1)>0$, the electrohydrodynamic flow tends to hinder the dielectrophoretic motion of a fluid particle in comparison with a solid sphere subjected to the same nonuniform electric field. In response to stresses arising from the electric field and electrohydrodynamic flow, a fluid particle can be deformed in a variety of shapes depending on the values of R, S, M, and Λ. A particle of a different shape may render different polarization features and in turn alter the electric and flow fields in different ways. For example, the dielectrophoretic force may increase when the fluid particle is elongated in the electric field direction because of a greater induced dipole moment, or vice versa. Hence, judiciously arranging the fluid properties so that the fluid particles are elongated or compressed in the direction of the applied electric field may become an effective means to control the strength of the dielectrophoretic force to achieve desired dielectrophoretic performance.

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[8] Dielectrophoretic forces are actually proportional to the local $\boldsymbol{\nabla} E^2$. In the present reference frame, net force effects cannot be obtained from any single term of P_l alone. Combinations of several P_{2n} and P_{2n+1} are usually needed for net forces to appear. Of many possibilities, the combination of P_1 and P_2 seems to be the simplest one.

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[11] In this work, the applied electric field is considered to be mainly in the direction of $\theta=0$ as consistent with (1), whereas the gravity and uniform flow velocity U, with respect to the reference frame fixed in the drop, are in the direction of $\theta=\pi$.

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