ELECTROHYDRODYNAMIC DEFORMATION AND INTERACTION OF A PAIR OF EMULSION DROPS

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ABSTRACT

The response of a pair of emulsion drops to the imposition of a uniform electric field is examined. The case studied is that of equal-sized drops whose line of centers is parallel to the axis of the applied field. A new boundary integral solution to the governing equations of the leaky dielectric model is developed; the formulation accounts for the electrostatic and hydrodynamic interactions between the drops, as well as their deformations. Numerical calculations show that, after an initial transient during which the drops primarily deform, the pair drift slowly together due to their electrostatic interactions.

INTRODUCTION

Externally-applied electric fields provide a well-known means for maniupulating suspenions of drops and bubbles. Common applications span a variety of multiphase flows, including enhanced coalescence, emulsion breaking and demixing operations for dispersions [1], electrophoretic migration of charged drops [2], enhanced heat and mass transfer owing to electroconvection [3], and aqueous two-phase partitioning [4]. Additional impetus for understanding the behavior of drops and bubbles in externally-applied electric fields arises from spaced-based materials processing, where nonuniform fields may be used to position fluid globules [5].

Previous research on the deformation of emulsion drops by electric fields has focused primarily on the behavior of isolated drops in uniform applied fields. In this context, the seminal contribution was made by Taylor [6], who demonstrated that conductive processes play a substantive role in determining how a drop, dispersed in another liquid, deformed in response to the imposed field. Taylor's analysis was pedicated on a model which has since come to be known as *the leaky dielectric*. Subsequent investigations have confirmed the essential premise of Taylor's analysis, i.e. that conduction processes cannot be altogether ignored, and have shown the leaky dielectric to be a useful quantitative model [7,8].

Several investigators have focused on the effect of a uniform electric field on two neighboring drops [9-12], motivated by the observation that the drop pair tends to coalesce due to the imposition of the field. As is the case for an isolated drop, the drops deform as the applied field strength increases, though now there are electrostatic and hydrodynamic interactions between the drops. Sozou [10] employed the leaky dielectric model to study the two drop problem in the limit that drop deformation was negligible, while others [11,12] have considered finite drop deformation but have neglected viscous effects. Taken together, these studies omit two important features of the problem for emulsion drops: first, dielectrophoretic forces, which bring the drops together, depend on charge conduction *and* drop shape; and second, resistance to the relative motion between the drops is dominated by viscous effects.

Here we apply the leaky dieletric model to investigate axisymmetric electrohydrodyanmic interactions between two emulsion drops of equal size. Boundary integral methods are used to obtain a numerical solution that accounts for the aforementioned problem features,viz., electrical conduction, drop deformation, and viscous-dominated hydrodynamic interations.

GOVERNING EQUATIONS

Balance Laws and Boundary Conditions

The balance laws of the leaky dielectric are well-known [13]. The velocity is governed by the Stokes equations, viz.

$$0 = -\nabla p + \mu \nabla^2 \mathbf{u} \qquad \nabla \cdot \mathbf{u} = 0, \tag{1}$$

and the electric field is solenoidal and irrotational, i.e.

$$\nabla \cdot \mathbf{E} = 0 \qquad \nabla \times \mathbf{E} = 0. \tag{2}$$

As a consequence of Eqn. (2), the electric field can be written as the gradient of a harmonic function, i.e.

$$\mathbf{E} = -\nabla\phi \qquad \nabla^2 \phi = 0, \tag{3}$$

where ϕ is the electric potential.

The electromechanical boundary conditions that apply at the drop surface couple the electrostatic field to the velocity. Classical electrostatics gives that

$$-\epsilon \nabla \phi \cdot \mathbf{n} + \bar{\epsilon} \nabla \bar{\phi} \cdot \mathbf{n} = \frac{q}{\epsilon_0}, \tag{4}$$

and

$$[\nabla \phi - \nabla \bar{\phi}] \times \mathbf{n} = 0, \tag{5}$$

where ϵ is the dielectric constant, ϵ_0 is the permittivity of free space, $\mathbf{n}$ is the unit normal pointing outward from the drop, and q is the local free surface charge density. Here overbars are used to indicate that a particular quantity refers to the drop interior.

The charge density q is not known independently of ϕ. Thus, while the electric potential must still satisfy Eqns. (4) and (5), the description of the electrostatic conditions at the interface must be augmented to unambiguously determine the electric field. If lateral transport of charge within the interface is neglected, conservation of charge requires that

$$\sigma \nabla \phi \cdot \mathbf{n} = \bar{\sigma} \nabla \bar{\phi} \cdot \mathbf{n}. \tag{6}$$

The velocity is continuous and the stresses balance at the interface, so

$$\mathbf{u} = \bar{\mathbf{u}} \tag{7}$$

and

$$\mathbf{n} \cdot (\mathbf{T}^N + \mathbf{T}^M) - \mathbf{n} \cdot (\bar{\mathbf{T}}^N + \bar{\mathbf{T}}^M) = \gamma (\nabla_s \cdot \mathbf{n})\mathbf{n}. \tag{8}$$

In Eqn. (8), γ is the interfacial tension, $\mathbf{T}^N$ and $\bar{\mathbf{T}}^N$ are Newtonian stress tensors, i.e.

$$\mathbf{T}^N = -p\mathbf{I} + \mu(\nabla \mathbf{u} + \nabla \mathbf{u}^T) \qquad \bar{\mathbf{T}}^N = -\bar{p}\mathbf{I} + \bar{\mu}(\nabla \bar{\mathbf{u}} + \nabla \bar{\mathbf{u}}^T), \tag{9}$$

and $\mathbf{T}^M$ and $\bar{\mathbf{T}}^M$ are Maxwell stress tensors, i.e.

$$\mathbf{T}^M = \epsilon \epsilon_0 [\mathbf{E}\mathbf{E} - \frac{1}{2}(\mathbf{E} \cdot \mathbf{E})\mathbf{I}] \qquad \bar{\mathbf{T}}^M = \bar{\epsilon}\epsilon_0[\bar{\mathbf{E}}\bar{\mathbf{E}} - \frac{1}{2}(\bar{\mathbf{E}} \cdot \bar{\mathbf{E}})\mathbf{I}]. \tag{10}$$

The remaining boundary conditions are: that the dependent variables remain bounded within the drops; and that, in the distance, the velocity vanish and $-\nabla\phi$ go over to $\mathbf{E}^\infty$, the externally-applied electric field.

Boundary Integral Formulation

A definition sketch for the problem to be solved is given in Fig. 1, and, as noted, the drops deform under the action of the imposed field. Because the drop shapes are not known *a priori*, analytic solutions to the governing equations are impracticable. The problem does, however, lend itself to boundary integral methods since the Green's functions for the balance laws are known.

The boundary integral representation for $E_n(\mathbf{x_s})$, the normal component of the electric field at some position $\mathbf{x_s}$ on a drop surface, is

$$E_n(\mathbf{x_s}) = \frac{2}{1 + \bar{\sigma}/\sigma} \mathbf{n} \cdot \left[\mathbf{E}^\infty(\mathbf{x_s}) + \frac{1}{4\pi}(\bar{\sigma}/\sigma - 1) \int_S |\mathbf{x_s} - \mathbf{y}|^{-3}(\mathbf{x_s} - \mathbf{y}) E_n(\mathbf{y}) dS_y \right]. \tag{11}$$

The component of the electric field tangent to the surface, i.e. $E_t \equiv \mathbf{t} \cdot \mathbf{E}(\mathbf{x_s})$, follows directly from the integration of an expression involving E_n, viz.

$$E_t(\mathbf{x_s}) = \mathbf{t} \cdot \left[\mathbf{E}^\infty(\mathbf{x_s}) + \frac{1}{4\pi}(1 - \bar{\sigma}/\sigma) \int_S |\mathbf{x_s} - \mathbf{y}|^{-3}(\mathbf{x_s} - \mathbf{y}) E_n(\mathbf{y}) dS_y \right]. \tag{12}$$

For the velocity, the boundary integral equation reads

$$\frac{1}{2}(1 + \frac{\bar{\mu}}{\mu})\mathbf{u}(\mathbf{x_s}) = \int_S \left[\mathcal{C}_{\text{el}}^{-1}\mathbf{n}(\nabla_s \cdot \mathbf{n}) - [\![\mathbf{n} \cdot \mathbf{T}^M]\!] \right] \cdot \mathbf{J}(\mathbf{x_s}|\mathbf{y}) dS_y - (1 - \frac{\bar{\mu}}{\mu}) \int_S \mathbf{n} \cdot \mathbf{K}(\mathbf{x_s}|\mathbf{y}) \cdot \mathbf{u}(\mathbf{x_s}) dS_y, \tag{13}$$

where

$$\mathbf{J}(\mathbf{x_s}|\mathbf{y}) = \frac{1}{8\pi} \left[\frac{\mathbf{I}}{|\mathbf{x_s} - \mathbf{y}|} + \frac{(\mathbf{x_s} - \mathbf{y})(\mathbf{x_s} - \mathbf{y})}{|\mathbf{x_s} - \mathbf{y}|^3} \right] \qquad \mathbf{K}(\mathbf{x_s}|\mathbf{y}) = -\frac{3}{4\pi} \frac{(\mathbf{x_s} - \mathbf{y})(\mathbf{x_s} - \mathbf{y})(\mathbf{x_s} - \mathbf{y})}{|\mathbf{x_s} - \mathbf{y}|^5}. \tag{14}$$

Note that Eqns. (11)-(14), which are constructed from the Green's functions for u and ϕ so as to satisfy Eqns. (4)-(8), are given in dimensionless form. The reference scales for the problem are: stress, $\epsilon\epsilon_0(E^\infty)^2$; velocity, $a\epsilon\epsilon_0(E^\infty)^2/\mu$; potential, aE^∞; and length, a. Accordingly, $\mathcal{C}_{\text{el}} \equiv a\epsilon\epsilon_0(E^\infty)^2/\gamma$ is an electric capillary number that characterizes the ratio of normal stresses due to electric and capillary forces; when $\mathcal{C}_{\text{el}}$ is small compared to unity, the imposed field causes little deformation of the emulsion drops.

RESULTS

Equations (11)-(13) were integrated numerically to obtain solutions for the electric and velocity fields. One of the virtues of the formulation summarized above is that, for a given drop shape, one can solve for the electric field without making reference to the velocity. Moreover, the components of the electric field can be obtained sequentially. Equation (11) is solved first for E_n, then E_t follows from Eqn. (12). Once the electric field is determined, the jump in the Maxwell stress is computed and substituted into the first integral on the RHS of Eqn. (13), along with the capillary stresses associated with the given drop shape. Owing to the need for the hydrodynamic stresses to balance these Maxwell and capillary stresses, the surface velocity field evinces a non-trivial normal component. Inasmuch as

$$\frac{d\mathbf{x_s}}{dt} = (\mathbf{n} \cdot \mathbf{u})\mathbf{n} = (\mathbf{n} \cdot \bar{\mathbf{u}})\mathbf{n}, \tag{15}$$

the drop shape is updated by moving the points $\mathbf{x_s}$, which are located on the interface, a distance $\delta\mathbf{x_s} = (\mathbf{n} \cdot \mathbf{u})\mathbf{n}\delta t$, where δt denotes the time step for the calculation. After updating the interface position, the calculation for the electric field is repeated, and so on, ...

Figures 2-4 show the results of such a calculation. In Fig. 2, the drop shapes and positions are shown as functions of time, which is scaled on $a\mu/\gamma$. The drops are assumed to be spherical and situated at $z_0 = 3.25$ when the field is imposed ($t = 0$). The behavior exhibited by the drop pair shown is characteristic of the results obtained in the study. The drops deform more rapidly than they translate. So long as the drops are not positioned such that their initial deformations bring their surfaces into virtual contact, translation is the rate limiting process by which the drops come together. (The phrases *virtual contact* and *come together* do not necessarily imply coalescence, since factors such as DLVO interactions have not been considered in the

365

calculations). The drops translate slowly toward one another because the electrical forces that drive them together are dielectrophoretic and, thus, die off roughly as one over the drop separation raised to the fourth power.

In Fig. 3, the velocity of the front and back edges of the right-hand drop are plotted against time; the front edge is that point closest to the origin (cf. Fig. 1). After the inital transient due to deformation, the front and back of the drop move with essentially the same velocity as the drop pair slowly drifts together. At approx. $t = 35$, the front edge of the drop accelerates as it comes under the increasingly stronger influence of the neighboring drop. Judging from Fig. 4, it would appear that this occurs when the dimensionless separation is about unity. Just before $t = 44$, when the numerical scheme says the drops touch, the front edge of the drop slows appreciably as the thin film between the pair drains, giving rise to lubrication forces.

CONCLUSIONS

A new boundary integral formulation for the electrohydrodynamic interaction between pairs of emulsion drops has been developed. The method accounts for the effects of conduction processes, viscous stresses and drop deformation on the interactions. Numerical results show that after an initial transient due to deformation, the drops converge comparatively slowly owing to the weakness of the electrostatic (dielectrophoretic) interactions. Since dielectrophoretic translation rates are shape dependent, the results imply that meaningful descriptions of emulsion behavior in electric fields ought to account for drop deformation induced by the imposed field.

ACKNOWLEDGEMENTS

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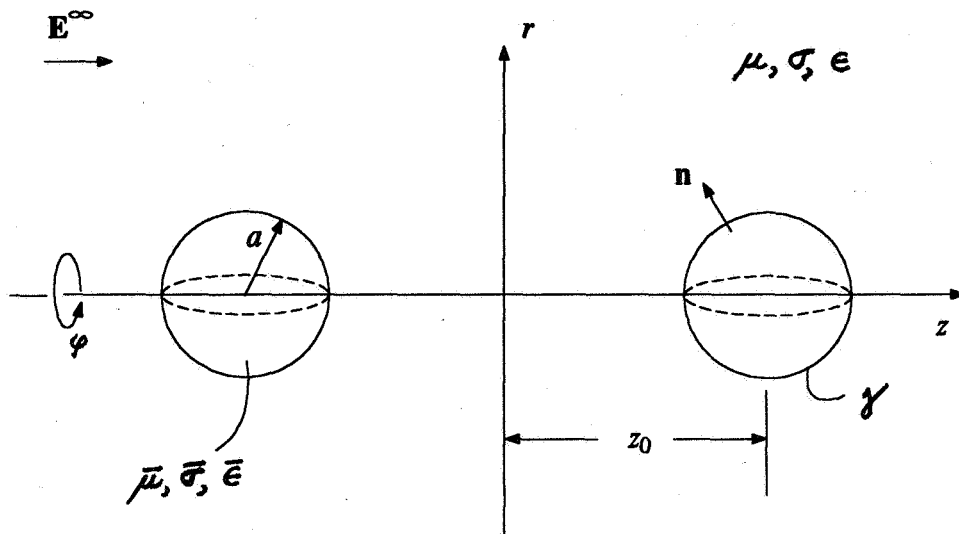13. J. R. Melcher and G. I. Taylor, *Ann. Rev. Fluid Mech.*, **1** (1969) 111-146.
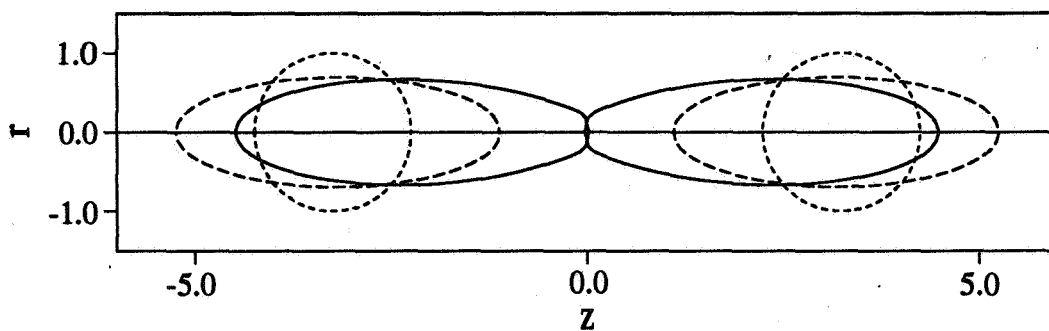
Figure 1: Definition sketch.

Figure 2: Drop shape and position as a function of time. $C_{el} = 5; \bar{\mu}/\mu = 1; \bar{\epsilon}/\epsilon = 3; \bar{\sigma}/\sigma = 3; z_0 = 3.25$.
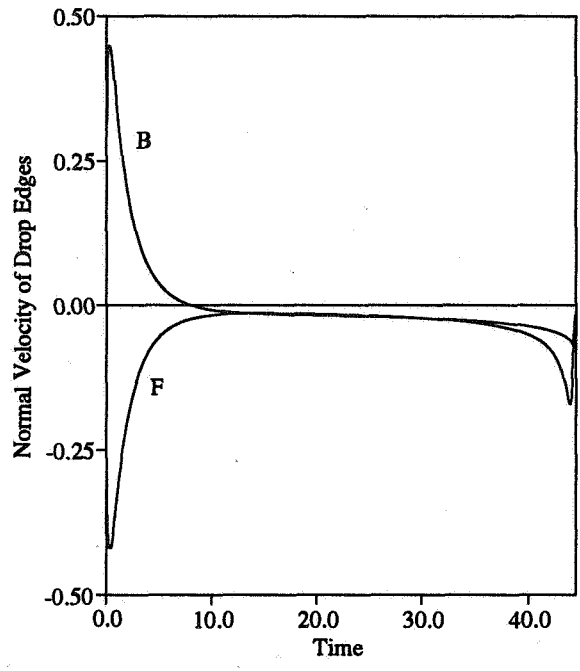Legend: ------------, $t = 0$; --------, $t = 10.4$; —————, $t = 44.0$.

Figure 3: Velocity of front (F) and back (B) edges of drop as a function of time. Parameter specifications are as for Figure 2.
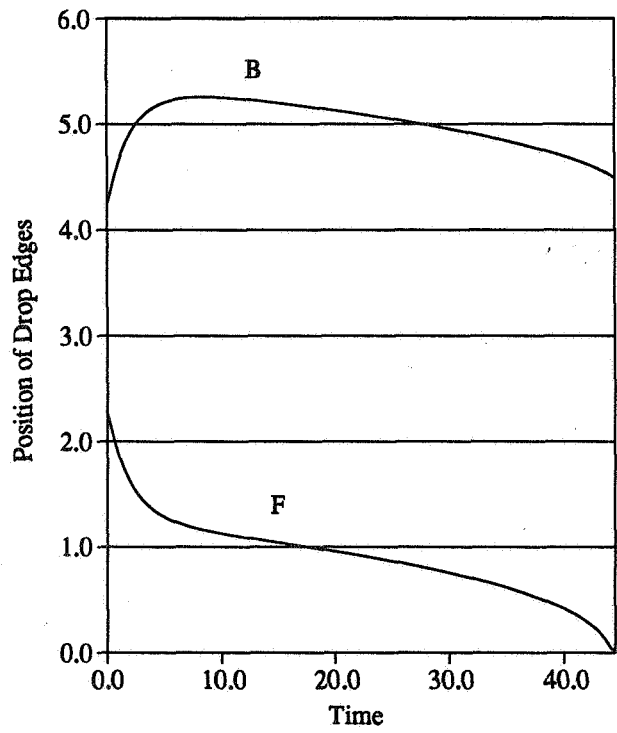

Figure 4: Position of front (F) and back (B) edges of drop as a function of time. Parameter specifications are as for Figure 2.