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ELECTROHYDRODYNAMIC INTERACTION OF A PAIR OF SPHERICAL DROPS

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ABSTRACT

The axisymmetric electrohydrodynamic interaction between two spherical emulsion drops has been examined, using the leaky dielectric model to represent the constitutive behavior of the liquid phases. The results follow from the general solutions in bispherical coordinates to the Laplace equation for the electric potential and the Stokes equations for the velocity field. For drops of similar composition, the electrical interactions induced between the drops by the imposition of the electric field are always attractive, meaning they favor coalescence of the drop pair. The hydrodynamic interactions, however, are not always favorable and, indeed, are shown in certain circumstances to drive the drops apart.

INTRODUCTION

The response of individual emulsion drops to the imposition of electric fields has been studied for a number of decades. The archetypal work on the subject is that due to Taylor (ref. 1), who first elucidated the role played by weak electrical conduction processes in the context of drop deformation caused by externally-imposed electric fields. Central to the drop deformation, Taylor showed, were steady fluid circulations driven in and about the drop. The circulations stem from interfacial electrical stresses that arise as a consequence of ohmic conduction processes in the liquids.

The circulations described by Taylor have since been recognized to be of technological significance as a tool to enhance heat and mass transfer in liquid-liquid dispersions. In the work that we summarize below, we show that the circulations are also significant with regard to the interactions that occur between neighboring drops in a space-filling dispersion. That is, we find the imposition of an electric field drives relative motion between a pair of drops that is a strong function of the hydrodynamics. The behavior of the emulsified drops thus contrasts with that of aerosols, where electrical interactions dominate the pairwise behavior.

PROBLEM STATEMENT

Consider two spherical emulsion drops immersed in a fluid with which they are immiscible. In general, the drops need not be the same size, and may possess electromechanical properties (e.g. viscosity μ, electrical conductivity σ and dielectric constant ϵ) distinct from one another as well as from those of the surrounding fluid (Fig. 1). Suppose now that a uniform electric field of strength E_∞ is externally applied along the line of centers of the drops. If the fluids are poor conductors, free charge will accumulate at the interfaces with the result that: one, the drops exert electrical forces on one another; and two, tangential

Maxwell stresses drive fluid circulation in and around the drops (ref. 1). Depending on the resultant of the electrical and hydrodynamic interactions, the relative motion between the drops may be such that they are either drawn together, or they move apart.

In the present analysis, we use the leaky dielectric model to represent the constitutive behavior of the fluids. Furthermore, we examine the case where surface tension is sufficiently high so as to hold the drops spherical. This fixed nature of the geometry facilitates an analytic solution of the electrohydrodynamic problem in the form of an expansion in bispherical harmonics.

Provided the charge relaxation time $\epsilon_3\epsilon_0/\sigma_3$ is small compared to the characteristic time associated with that of fluid motion, $\mu_3/\epsilon_3\epsilon_0 E_\infty^2$, one can solve for the electric potential independently of the flow-field. The electrostatics are governed by the Laplace equation

$$\nabla^2\Phi_i = 0 \qquad \text{for} \qquad i = 1, 2, 3, \tag{1}$$

where Φ_i is the potential in each of the three regions (Fig. 1). The boundary conditions on the drop surfaces are those of continuity of both Φ and the normal component of electric current density. Far from the drops, $-\nabla\Phi$ goes over to the applied field.

The flow is slow and axisymmetric, so in cylindrical coordinates,

$$E^4\psi = 0, \qquad \text{with} \qquad E^2 = r\frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial}{\partial r}\right) + \frac{\partial^2}{\partial z^2}, \tag{2}$$

and ψ is the stream function. The velocity is continuous, and the tangential components of the stress balance on the drop surfaces. The normal stress balance is not considered explicitly; instead we apply a kinematic condition to the normal component of the velocity.

The solution to the Stokes equation gives us the forces on the drops due to the electrically-driven circulations, as well as the hydrodynamic resistance to the relative motion of the drops. The translational velocities V_1 and V_2 of the drops, both of which are in the z direction, follow from balancing the force due to the electrical interactions between the drops with the two hydrodynamic forces, viz. the hydrodynamic force that stems from the electrically-driven circulations and that due to the hydrodynamic resistance to the drops' relative motion.

For simplicity, all variables are made dimensionless through division by the following characteristic quantities: length, a_1; electric potential, $a_1 E_\infty$; stress, $\epsilon_3\epsilon_0 E_\infty^2$; and velocity, $a_1\epsilon_3\epsilon_0 E_\infty^2/\mu_3$.

Owing to the geometry of the problem, it is convenient to introduce bispherical coordinates ξ and η, which are related to (dimensionless) cylindrical coordinates r and z in the following manner:

$$z = \frac{a \sinh\xi}{\cosh\xi - \cos\eta}; \qquad r = \frac{a \sin\eta}{\cosh\xi - \cos\eta}. \tag{3}$$

$\xi > 0$ for $z > 0$, $\xi < 0$ for $z < 0$, with $\xi = 0$ on the plane $z = 0$ and for infinite distance from the drops. η represents the inverse tangent of the angle between two lines drawn to a point from the drop centers. The surfaces of drops 1 and 2 have constant values of ξ, denoted ξ_1 and ξ_2. These values are related to the drop radii and distances d_i from the plane $z = 0$ by $a_i/a_1 = \pm a \operatorname{cosech}\xi_i$ and $d_i/a_1 = \pm a \coth\xi_i$, with the plus and minus signs for $i = 1$ and 2, respectively.

RESULTS AND DISCUSSION

The solution to the electrostatics problem in the bispherical system is (ref. 2)

$$\Phi_1 = -z + (\cosh\xi - \mu)^{\frac{1}{2}} \sum_{n=0}^{\infty} C_n \exp\left[-\left(n + \frac{1}{2}\right)\xi\right] P_n(\mu), \tag{4}$$

$$\Phi_2 = -z + (\cosh\xi - \mu)^{\frac{1}{2}} \sum_{n=0}^{\infty} D_n \exp\left[\left(n+\frac{1}{2}\right)\xi\right] P_n(\mu), \tag{5}$$

$$\Phi_3 = -z + (\cosh\xi - \mu)^{\frac{1}{2}} \sum_{n=0}^{\infty} \left\{ A_n \exp\left[-(n+\frac{1}{2})\xi\right] + B_n \exp\left[(n+\frac{1}{2})\xi\right]\right\} P_n(\mu) \tag{6}$$

where $\mu = \cosh\eta$, and $P_n(\mu)$ is the nth Legendre polynomial. The constants A_n, B_n, C_n, and D_n are determined via recursion relations obtained by applying the boundary conditions and using the orthogonality of the Legendre polynomials.

Knowing the potential enables us to get an explicit expression for the electric stress at the surface, which in turn gives us an expression for the total electric force on each drop, as we integrate the stress over each drop surface, viz.

$$F_i^{\text{elec}} = 2\pi \int_{-1}^{1} \left\{ \frac{1}{2}\frac{1-\mu\cosh\xi}{\cosh\xi-\mu}\left[-\frac{\sigma_i}{\sigma_3}^2\left(\left.\frac{\partial\Phi_i}{\partial\xi}\right|_{\xi_i}\right)^2 + (1-\mu^2)\left(\left.\frac{\partial\Phi_i}{\partial\mu}\right|_{\xi_i}\right)^2\right] - \frac{\sigma_i}{\sigma_3}\sinh\xi\frac{1-\mu^2}{\cosh\xi-\mu}\left.\frac{\partial\Phi_i}{\partial\xi}\right|_{\xi_i}\left.\frac{\partial\Phi_i}{\partial\mu}\right|_{\xi_i}\right\} d\mu, \qquad i=1,2. \tag{7}$$

Note the tangential component of the electric stresses also contribute to the tangential stress balance condition for the drop surfaces and this drives the velocity field.

The general solution to the Stokes equation for the stream function ψ is

$$\psi_i = (\cosh\xi - \mu)^{-\frac{3}{2}}(1-\mu^2)\sum_{n=1}^{\infty} U_{in}(\xi) P_n'(\mu), \qquad i = 1,2,3, \tag{8}$$

where

$$U_{in}(\xi) = a_{in}\sinh[-(n-\frac{1}{2})\xi] + b_{in}\cosh[(n-\frac{1}{2})\xi] + c_{in}\sinh[-(n+\frac{3}{2})\xi] + d_{in}\cosh[(n+\frac{3}{2})\xi], \qquad i=1,2,3. \tag{9}$$

Thus the boundary conditions can be recast as equations in $U_1(\xi_1)$ and $U_2(\xi_2)$, which comprise a linear system of equations for the stream function coefficients a_{in}, b_{in}, c_{in}, and d_{in} for each n from 1 to ∞.

Sozou (ref. 3) has implemented this scheme for the particular situation involving identical drops, subject to the restriction that there be no relative motion between the drops. Here we relax Sozou's constraints, allowing for relative motion and considering drops of different size and electromechanical properties. The principal results that come from our analysis, then, are the drop velocities V_1 and V_2, *and* the conditions for which no relative motion obtains.

The coefficients for the external flow field are used to compute the net hydrodynamic force F_i^{hyd} exerted on the drops, through the well-known formulae (ref. 4)

$$F_i^{\text{hyd}} = F_i^{\text{rel}} + F_i^{\text{circ}} = -\frac{2\sqrt{2}\pi}{a}\sum_{n=1}^{\infty} n(n+1)\left(a_{3n} \pm b_{3n} + c_{3n} \pm d_{3n}\right), \tag{10}$$

with the plus and minus signs corresponding to $i = 1$ and 2, respectively. The coefficients a_{in}, b_{in}, c_{in}, and d_{in} are linear in V_1 and V_2, and F_i^{hyd} involves a contribution from the relative motion F_i^{rel} and from the electrically-driven circulation F_i^{circ}. Since F_i^{rel} is a linear combination of V_1 and V_2, one may write

$$F_1^{\text{rel}} = \text{DC}_{11}V_1 + \text{DC}_{12}V_2, \qquad F_2^{\text{rel}} = \text{DC}_{21}V_1 + \text{DC}_{22}V_2, \tag{11}$$

where $DC_{11}, DC_{12}, DC_{21}$, and DC_{22} are the drag coefficients for the relative motion (refs. 5 and 6). Finally, balancing F_i^{elec}, F_i^{rel}, and F_i^{circ} on each drop yields the drop velocities, viz.

$$V_1 = -\left[DC_{22}(F_1^{elec} + F_1^{circ}) - DC_{12}(F_2^{elec} + F_2^{circ})\right]/\Delta, \tag{12}$$

$$V_2 = \left[DC_{12}(F_1^{elec} + F_1^{circ}) - DC_{22}(F_2^{elec} + F_2^{circ})\right]/\Delta, \tag{13}$$

where

$$\Delta = DC_{11}\,DC_{22} - DC_{12}\,DC_{21}\,. \tag{14}$$

In Fig. 2, we give the translational velocity of drop 1 as a function of center-to-center separation for the special case that the drops are identical. For this circumstance, the electric forces on the drops are attractive. The electrically-driven circulations are attractive when $\sigma_1/\sigma_3 < \epsilon_1/\epsilon_3$, repulsive for $\sigma_1/\sigma_3 > \epsilon_1/\epsilon_3$, and vanish when $\sigma_1/\sigma_3 = \epsilon_1/\epsilon_3$. Plots are given for various conductivity ratios σ_1/σ_3, with viscosity ratio μ_1/μ_3 fixed at 1.0 and dielectric constant ratio ϵ_1/ϵ_3 fixed at 2.0. A negative velocity indicates that the drops are moving toward one another, so one can see that for certain values of $\sigma_1/\sigma_3 > \epsilon_1/\epsilon_3$, the drops may move apart. Physically, this means that the tangential electric stresses acting on the drop surfaces drive circulations that not only oppose drop motion, but are strong enough to overcome the attractive force due to electric interactions.

In Fig. 3 there are given plots of combinations of conductivity ratio σ_1/σ_3 and dielectric constant ratio ϵ_1/ϵ_3 for various drop separations at which the relative motion between the drops vanishes. Again, for simplicity, the results shown are for identical drops. Given a curve for a particular separation $D = (d_1+d_2)/a_1$, the area underneath the curve represents combinations of σ_1/σ_3 and ϵ_1/ϵ_3 for which the drops are driven apart by the fluid flow when the separation is at least D. For values of σ_1/σ_3 and ϵ_1/ϵ_3 that lie above the curve, the drops translate toward each other when at separations less than D. Thus, we see in Fig. 3 that a smaller variety of electrical properties facilitate coalescence for larger separations. This is mainly because F_i^{elec}, $i = 1, 2$, decays inversely with drop separation to the fourth power, whereas the interactions due to the circulations decay as one over the separation squared.

CONCLUDING REMARKS

The results presented here are a useful leading-order analysis for electrohydrodynamic interactions involving drops that may deform modestly under the action of an electric field. In general, keeping track of drop shape must be done numerically. The analytic results offer the advantage of providing qualitative behavior, such as the direction of the drops' translation for different values of σ_i/σ_3, ϵ_i/ϵ_3, μ_i/μ_3, and drop separation. Such information is rather cumbersome to generate numerically, especially when the relative motion is weak. Denoting the interfacial tension as γ, we note that the capillary number $Ca = a_1\epsilon_3\epsilon_0\,E_\infty^2\,/\gamma$ is small ($\ll 1$) in our study, and thus one can add corrections to the solution presented, using Ca as a perturbation parameter.

The results are most useful for appreciable drop separations, i.e. when the gap between the drop surfaces is at least the radius of the smaller drop. Referring to Eqs. (6), (7), and (8), we note that for smaller separations, it takes more terms in the sums for Φ and ψ to make these expressions accurate. Simple expressions for hydrodynamic resistance have been derived (refs. 7, 8) based on asymptotic analysis of the infinite sum that appears in Eq. (10) for the case of strictly spherical drops. In addition, it is at smaller separations where drop deformation becomes more significant. Indeed, it has been noted by Davis et al. (ref. 8) and others that, for small separations with the drops moving toward one another, the attendant increase in pressure within the lubrication layer results in dimpling of the drop surfaces. A subject for subsequent investigation, therefore, would be the influence of the electrohydrodynamic circulations on such dimpling.

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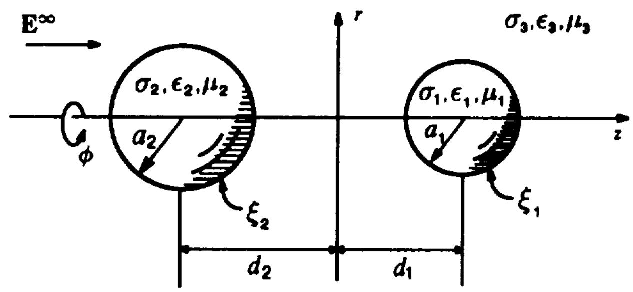

Figure 1: Definition sketch.

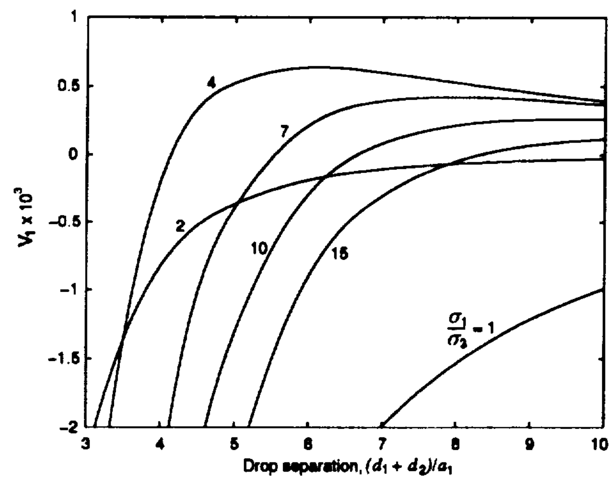

Figure 2: Velocity of drop 1 as a function of center-to-center separation, for various values of $\sigma_1/\sigma_3(=\sigma_2/\sigma_3)$. The unit of length is the radius of drop 1. $\mu_1/\mu_3 = \mu_2/\mu_3 = 1$; $\epsilon_1/\epsilon_3 = \epsilon_2/\epsilon_3 = 2$.

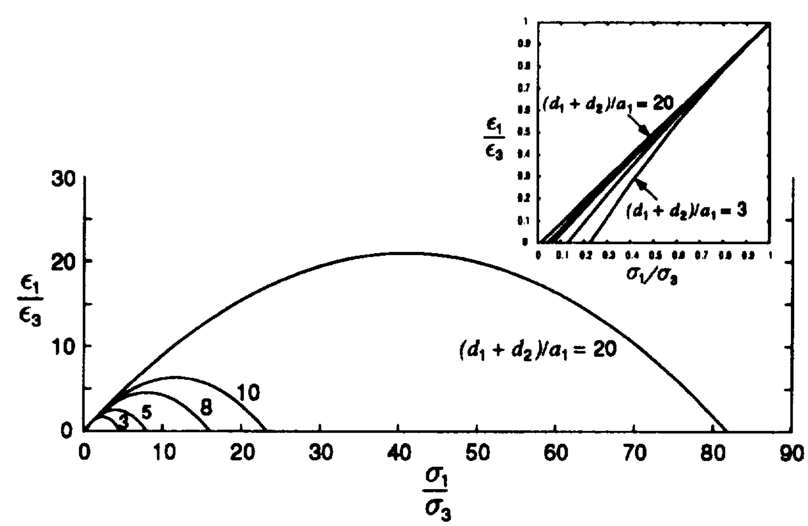

Figure 3: Combinations of conductivity and dielectric constant that hold two drops fixed in space, for various center-to center separations. $\mu_1/\mu_3 = \mu_2/\mu_3 = 1$; $a_1 = a_2$; $\sigma_1/\sigma_3 = \sigma_2/\sigma_3$; $\epsilon_1/\epsilon_3 = \epsilon_2/\epsilon_3$.