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Deformation of a droplet in an electric field: Nonlinear transient response in perfect and leaky dielectric media

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Abstract

Deformation of a fluid drop, suspended in a second immiscible fluid, under the influence of an imposed electric field is a widely studied phenomenon. In this paper, the system is analyzed numerically to assess its dynamic behavior. The response of the system to a step change in the electric field is simulated for both perfect and leaky dielectric systems, exploring the influence of the fluid, interfacial, and electrical properties on the system dynamics. For the leaky dielectric case, the dynamic build up of the free charge at the interface, including the effects of convection along the interface due to electrohydrodynamic circulation, is investigated. The departure of the system from linear perturbation theory is explained using these dynamic simulations. The present simulations are compared with analytic solutions, as well as available experimental results, indicating that the predictions from the model are reliable even at considerably large deformations.

Keywords: Droplet deformation; Electric field; Navier–Stokes equations; Finite element; Perfect dielectric; Leaky dielectric

1. Introduction

A fluid drop suspended in another immiscible fluid will deform when subjected to an electric field. Applications of this phenomenon encompass spraying [1], aerosols, inkjet printing, coalescence of droplets for de-emulsification purposes [2–4], and electrowetting-based droplet manipulation in microfluidic systems [5–7], to name a few. The problem of droplet deformation under the influence of an imposed electric field has been extensively studied from different perspectives. The mathematical foundation of the subject has been rigorously established, and several aspects of the equilibrium and transient behavior of such systems have been explored.

Fig. 1 schematically depicts the fundamental parameters governing the extent of droplet deformation under the influence of an electric field. As noted in the figure, the key properties dictating the nature and extent of deformation are the viscosity, dielectric permittivity, and electric conductivity of the two fluids. The interfacial tension is unique for a given fluid–fluid interface. The deformation of a drop in an electric field is characterized by a balance of interfacial tension, hydrodynamic, and electrical stresses at the droplet interface. The electrical stresses cause the interface to distort, while interfacial tension tends to restore the original shape. Viscous stresses and fluid pressure gradients due to the flow fields can also alter the deformation substantially. The mathematical description of this system requires simultaneous solution of the fluid mechanical equations and the equations of electrostatics.

The first analytic result predicting the deformation of a drop in an electric field was derived by O'Konski and Thacher [8] for perfectly insulating (dielectric) drops in perfectly insulating media. Subsequently, Allan and Mason [9] performed a force balance over the surface of a dielectric drop in a dielectric medium. Their result was equivalent to the O'Konski and Thacher result. Using the notations of Fig. 1, the steady state deformation, d, of the droplet predicted by the O'Konski and Thacher/Allan and Mason (OTAM) expression is

$$d_\infty = \frac{R_0\epsilon_e|\mathbf{E}_0|^2}{\gamma}\frac{9(S-1)^2}{16(S+2)^2}, \tag{1}$$

where $|\mathbf{E}_0|$ is the magnitude of the electric field vector along the z coordinate, and the parameter S $(=\epsilon_i/\epsilon_e)$ represents the

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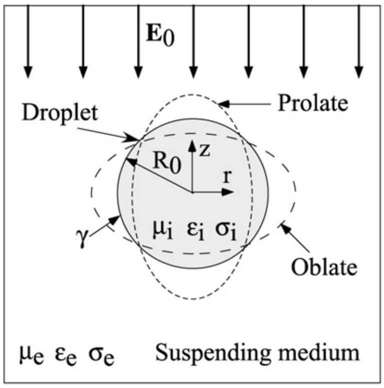

Fig. 1. Schematic representation of the deformation of a droplet suspended in a second fluid in the presence of an electric field, $\mathbf{E}_0$, acting along the z axis of the cylindrical coordinate system. The shaded circle represents the spherical nondeformed droplet before application of the field. The possible steady-state deformed shapes, prolate and oblate spheroids, are also depicted. The nondeformed droplet diameter is R_0. For the deformed spheroidal shapes, the equatorial radius (a) and polar (b) semi-axis are directed along the r and z coordinates, respectively. The relevant properties of the droplet and suspending medium are the viscosity, μ, dielectric permittivity, ϵ, and conductivity, σ. The subscripts i and e are used to represent the droplet and the suspending fluid, respectively. The interfacial tension, γ, constitutes the restoring force.

ratio of dielectric permittivities of the droplet and the suspending medium. The deformation parameter, d, is defined as

$$d = \frac{b-a}{b+a}, \tag{2}$$

where a is the equatorial radius of the spheroid and b is the polar half-axis. The symbol d_∞ is used in Eq. (1), and in the rest of the paper to indicate that the deformation is evaluated at steady state. According to Eq. (1), the deformation must always be positive, i.e., the resultant deformed shape is always a prolate spheroid, elongated in the direction of the electric field. Allan and Mason [9], however, observed discrepancies between the predictions of Eq. (1) and their experiments on electrical deformation of droplets. In some of their experiments oblate deformation (compression in the axis of the applied field) was observed, which was clearly beyond the scope of Eq. (1).

Taylor [10] and later Melcher and Taylor [11] developed an electrohydrodynamic model for the deformation of conducting droplets suspended in a conducting medium under the influence of an imposed electric field. This model, generally referred to as the "leaky dielectric model," has become a cornerstone of the theory of electrical drop deformation. The expression for the steady-state deformation in this model is

$$d_\infty = \frac{R_0 \epsilon_e |\mathbf{E}_0|^2}{\gamma} \frac{9}{16(2+H)^2} \times \left[H^2 + 1 - 2S + 3(H-S)\left(\frac{2+3M}{5+5M}\right)\right], \tag{3}$$

where

$$S = \frac{\epsilon_i}{\epsilon_e}, \qquad H = \frac{\sigma_i}{\sigma_e}, \qquad M = \frac{\mu_i}{\mu_e}.$$

Equation (3) successfully predicted the oblate deformations observed in the experiments of Allan and Mason [9]. The leaky dielectric model, however, suffers the limitation that it is invalid for perfect dielectric systems. This is immediately apparent from Eq. (3), where setting $\sigma_i = \sigma_e = 0$ does not yield the OTAM expression (Eq. (1)).

The leaky dielectric model has been elaborated upon in several subsequent works, notably [12–17], which specifically focus on the transport modes of the free charge carriers (ions). A comprehensive review of the theoretical developments in this area was given by Saville [15]. Zholkovskij et al. [16] provided a solution for the electrokinetic problem that resolved the disparity between the OTAM and Taylor results in the limit of zero conductivity. More recently, an extension of the Taylor model was proposed [17].

All the above mentioned approaches pertain to steady-state analysis of small deformations under the influence of small electric fields. These results do not apply to the dynamic problem, and cannot address large deformations or breakup of droplets. The transient electrohydrodynamic problem addressing the deformation of droplets is also an extensively studied subject [18–24].

Since the theoretical treatments of drop deformation in an electric field are limited to small deformations, or large deformations with assumptions placed on the shape, there has been a significant and continuing interest in finding numerical solutions to these problems. The pioneering computational studies on transient deformation of droplets appeared in the early 1970s [25,26]. These studies deal with conducting drops with constant surface potential, both isolated and in pairs, and isolated charged drops. A finite perturbation technique was employed to converge to the correct force balance for this system, yielding the steady-state result, which for relatively large deformations showed a small deviation from the spheroidal shape. The irrotational, inviscid fluid mechanics equations were solved to provide dynamic analyses of drop deformation and contact between drop pairs.

Sherwood [13] analyzed the leaky dielectric model by means of a boundary integral method. Viscosity was included in the model, but the droplet and medium were assumed to be equally viscous. The momentum term in the Navier–Stokes equations was neglected. Large deformations were obtained by stepwise increases in the applied field. Sherwood's results indicate that when the permittivity ratio ϵ_i/ϵ_e is sufficiently high, the drop develops the pointed ends characteristic of tip streaming, whereas a high conductivity ratio tends to produce a bulbous-ended breakup mode. The steady-state perfect dielectric problem was also solved using Newton's method with stepwise changes in applied field [27].

Finite element analysis of the shape patterns and stability of a charged drop in an electric field was conducted by Basaran and Scriven [28,29]. Their steady-state analysis assumed infinite drop conductivity, so that the final state did not involve flow, and hence the fluid mechanics equations were not necessary. Haywood et al. [30] developed a numerical technique to predict the transient deformation of a perfect dielectric system. Differences between the stability limits predicted from steady-state analysis and from their fully dynamic model were observed. Tsukada et al. [31] performed finite element cal-