Deformation of Drop-within-Drop System under the Influence of High Intensity Oscillating Electric Field

Diptendu Das[1], Rajkumar S.Patil[1], Sonali Kulkarni[1], V.A. Juvekar [1] and Ramdas Bhattacharya [2]

[1] Department of Chemical Engineering, IIT Bombay, Mumbai -400076, India

[2] Atomic Energy Regulatory Board, Mumbai-400094, India

Abstract: This paper deals with the simulation study on the deformation of drop-within-drop system in a high intensity oscillating electric field. It consists of a composite drop suspended in a continuous phase. This composite drop has a smaller inner drop suspended within the larger outer drop. For the sake of simplicity we consider the case where the inner drop and the continuous phase are made of the same liquid. The outer drop is made up of another immiscible liquid. The entire system is contained within a cylinder with insulating curved surface. A high intensity sinusoidally varying axial field is imposed on the system. In this paper, we have studied deformation of the individual drops and compared the deformation of the outer drop in the presence/ absence of the inner drop. The viscous effect slows down the response of the outer drop and hence it cannot regain its original spherical shape during the quarter cycle period of ebbing-field. Hence, with each cycle, the drop deformation continues to increase till steady state is reached. At high field intensities, the deformation becomes boundless. Although, the outer drop shows the same steady deformation in the presence and in the absence of the inner drop, the presence of the inner drop slows down the response of the outer drop.

This study is very important in understanding the stability of liquid-liquid interface for designing the liquid emulsion membranes.

Keywords: Double emulsion, Leaky dielectric model, electro hydrodynamic flows, drop deformation, emulsion stability

1. Introduction

Double emulsions are formed by dispersing an emulsion in another immiscible liquid phase. Examples are: Water-oil-water (W/O/W), oil-water-oil (O/W/O) or oil-oil-oil (O'/O/O') emulsions. Double emulsions have tremendous potential applications in various fields particularly in metal extraction, drug delivery, cosmetics, agriculture, food, photography, leather etc [1-3].

In Liquid Emulsion Membrane (LEM) extraction technique, a valuable solute at very low concentrations in external continuous phase is extracted into the internal strip-phase. The two phases are separated by the middle membrane phase. This process has wide applications in the separation and recovery of organic solutes, minerals and toxic materials like phenols, uranium and cadmium, etc. from effluents and also in waste-water treatment [4-8].

Application of LEM technology to industrial scale is, however, hindered by the challenges in achieving stability of the double emulsion during the extraction. Swelling of the emulsion, rupture of the membrane and the leakage of internal phase into the external phase are the associated problems. They are all caused by deformation of the interfaces. The shear stresses aggravate these problems. So it is essential to study the effect of shear on the overall stability of double emulsions.

It is observed that when an electric field (AC/DC) is applied to a liquid drop which is suspended in another immiscible liquid, it undergoes deformation due to electrohydrodynamic stresses at the interface. If the intensity of the applied electric field is below the critical value and it is oscillating then the drop elongates and contracts. The frequency of elongation and contraction is equal to double the frequency of the applied electric field with some phase lag. The restoring force is interfacial tension which balances this electric force which causes regaining of spherical shape of drop. This drop elongation and relaxation causes the fluids to flow in and around the drop. This velocity field gives rise to a shear field around the internal droplets. Hence applying oscillating electric field it is possible to study shear induced instability. Lot of experimental and theoretical studies[9-12] have been done on drop deformation under electric field which is useful for understanding the stability of single emulsion under shear. The deformation of double emulsion globule has been experimentally studied by Ha et al.(1999) [13, 14]. However, no simulation studies have been conducted so far.

In this paper we have simulated the deformation of a drop-within-drop system in a high intensity oscillating electric field. The system consists of a composite drop suspended in a continuous phase. This composite drop consists of a smaller inner drop suspended within the larger outer drop. This study is the first step towards understanding the deformation of a double emulsion globule.

2. Physical Model and Mathematical Formulations

2.1 Problem Description

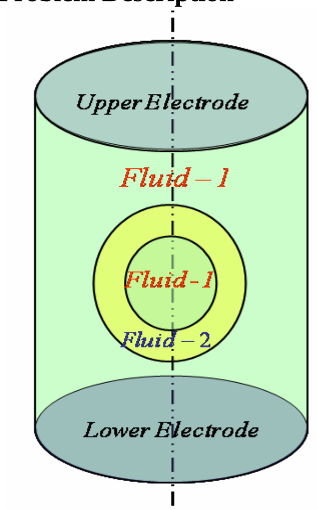

Figure 1: Schematic representation of drop within drop system.

Fig 1 shows our physical model for the study of deformation of drop within drop system. In this system, the fluid-1 (density ρ_1, viscosity μ_1, electrical conductivity κ_1 and permittivity ε_1) and fluid-2 (density ρ_2, viscosity μ_2, electrical conductivity κ_2 and permittivity ε_2) are immiscible. The drop assembly is placed at the centre of cylindrical vessel which contains a continuous phase of fluid-1. The metal electrodes are fixed at the top and bottom surfaces of the vessel for applying high intensity oscillating electric field along the axis of the cylinder. The curved wall of the cylinder is insulating.

2.2 Problem Formulation

The problem is cast in 2D axisymmetric case as illustrated in Figure-2. The outer drop BCJB is contained within a cylindrical domain DEFAD. The inner drop GHIG is inside the outer drop. For the purpose of simulation, we consider three sub-domains. Subdomain 1: external continuous phase (DEFAD), subdomain 2: outer drop (BCJB) and subdomain-3, internal drop (GHIG). The radius of the cylinder and its height are respectively 5 mm and 10 mm. The outer drop radius is 2 mm and the inner drop radius is 1.5 mm.

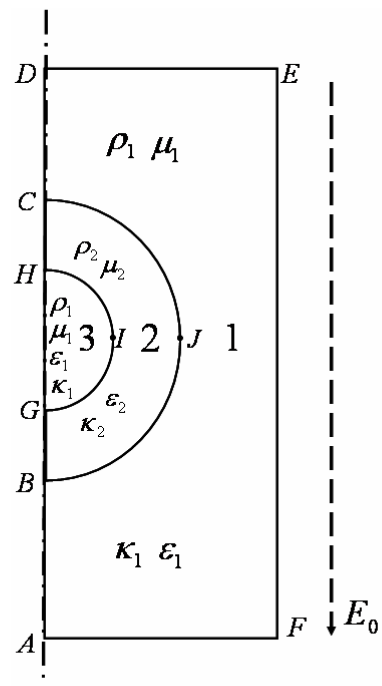

Figure 2: Schematic representation of the axisymmetric model of drop-within-drop system under oscillating Electric Field.

The circular end surfaces AF and DE are the electrodes having respective sinusoidal potentials V_+ and V_- such that $V_+ = (1/2)V_0 \sin(\omega t)$ and $V_- = -V_+$. The densities of the fluids are assumed to be equal so that the effect of gravity can be neglected. The value of the interfacial tension, σ, is chosen as 5 mN.m^{-1}. The viscosity ratio $M=\mu_2/\mu_1$ is chosen as unity. The conductivity ratio $R=\kappa_2/\kappa_1$ of 37.5 and the ratio of dielectric constants $Q=\varepsilon_2/\varepsilon_1$ of 0.1 are used. Two different values of V_0 are chosen for the simulation, viz. 800 V and 1000 V. The frequency of oscillation is maintained at 1 Hz.

2.2 Governing Equations for the Flow

The fluid velocity is obtained by solving the equations of continuity and motion. These can be expressed as follows.

$$\overline{\nabla} \cdot \overline{v}_i = 0 \quad \text{i = 1, 2, 3} \tag{1}$$

$$\rho_i \frac{\partial \overline{v}_i}{\partial t} + \rho_i \left(\overline{v}_i \cdot \overline{\nabla}\right)\overline{v}_i = \nabla \cdot \left[p_i \overline{\overline{I}} + \mu_i \left(\overline{\nabla} \overline{v}_i + \left(\overline{\nabla} \overline{v}_i \right)^T \right) \right] + \overline{f}_{Ei} + G \overline{\nabla} \phi \tag{2}$$

where, v_i = velocity of fluid in the domain i , p_i is the pressure, G is the Gibbs free energy of the system, ϕ is the phase field function to track the interface deformation and $\overline{f}_E$ is the electrical force per unit volume due to Maxwell stress tensor $\overline{\overline{\tau}}_E$ and is given by

$$\overline{f}_E = \overline{\nabla} \cdot \overline{\overline{\tau}}_E \tag{3}$$

The dynamics of the interface deformation due to stresses can be solved numerically by using Cahn-Hilliard diffusion equation. In COMSOL Multiphysics, the Cahn-Hilliard diffusion equation is split into two parts for numerical convenience are given by Eq. 4 and by Eq. 5

$$\frac{\partial \phi}{\partial t} + \overline{v} \cdot \overline{\nabla} \phi = \nabla \cdot \left(\alpha \beta / \delta^2 \right) \overline{\nabla} \psi \tag{4}$$

$$\psi = -\nabla \cdot \delta^2 \nabla \phi + \phi\left(\phi^2 - 1\right) \tag{5}$$

ϕ is the dimensionless phase field variable, α is the mobility, β is the energy density and δ scales the interface thickness. In the above Eq. 4, the interfacial tension value is incorporated through the energy density β and interface thickness δ by following expression.

$$\sigma = \frac{2\sqrt{2}}{3} \cdot \frac{\beta}{\delta} \tag{6}$$

In this system, we have two immiscible fluids; the phase field function distributes its value as a +1 for bulk fluid-1, -1 for the bulk fluid-2 and zero at the interface.

2.2 Governing Equation for Electric Field

In subdomains 1, 2 and 3 we used Maridional Electric current equation to describe the potential distribution and the current density distribution in the system. The total charge conservation in the system is given by

$$\frac{\partial C}{\partial t} = -\overline{\nabla} \cdot \overline{j} \tag{7}$$

C is the volume charge density in the bulk fluids and $\overline{j}$ is the flow of current density in the system due to free ions in the bulk fluids. The current density in the system is given by Eq 8

$$\overline{j} = -\kappa_r \overline{\nabla} \overline{V} \tag{8}$$

The κ_r is the relative conductivity in the system. It is expressed in terms of volume fractions of each phase in the system multiplied by their corresponding conductivity value.

$$\kappa_r = \kappa_1 \times Vf_1 + \kappa_2 \times Vf_2 \tag{10}$$

The Vf_1 is the volume fraction of fluid 1 in the system and is equal to $(1+\phi)/2$, similarly the Vf_2 is the volume fraction of fluid 2 in the system and is equal to $(1-\phi)/2$.

The charge distribution in the system is given by Poisson Equation

$$\overline{\nabla} \cdot \left(\varepsilon_r \overline{\nabla} \overline{V} \right) = -\frac{C}{\varepsilon_0} \tag{11}$$

ε_r is the effective dielectric constant in the system and is defined as

$$\varepsilon_r = \varepsilon_1 \times Vf_1 + \varepsilon_2 \times Vf_2 \tag{12}$$

ε_0 is the relative permittivity of the free space.

The leaky dielectric theory was incorporated in our problem formulation by using a governing equation of Maridional Electric current in COMSOL Multiphysics which is given by Eq 13.

$$\overline{\nabla} \cdot \frac{\partial}{\partial t}\left(\varepsilon_0 \varepsilon_r \overline{\nabla} \overline{V}\right) = \overline{\nabla} \cdot \left(\kappa_r \overline{\nabla} \overline{V}\right) \qquad (13)$$

The Maxwell stress tensor is given by

$$\bar{\bar{\tau}}_{m\,i} = \varepsilon_i \varepsilon_0 \left[\overline{\nabla} V_i \overline{\nabla} V_1 - \frac{1}{2}\left(\overline{\nabla} V_i \cdot \overline{\nabla} V_i\right) \bar{\bar{I}} \right] \qquad (14)$$

and the electric force per unit volume is given by

$$\bar{f}_{E,i} = \overline{\nabla} \cdot \bar{\bar{\tau}}_{m,i} \qquad (15)$$

Initial Conditions:

At time $t = 0$
The fluid velocity $v_i = 0$
The potential at electrodes $V = 0$

Boundary conditions

Fluid Flow:

On the boundaries AF, FE, ED: The velocity of fluid is zero, ($v_i = 0$) since they are walls.
The boundary ABGHCD is the axis of symmetry,

The stress continuity is applied at both the interfaces, BJC and GIH:

$$n \cdot \left(\mu_1\left(\nabla v_1 + (\nabla v_1)^T\right) - p_1 \bar{\bar{I}} - \mu_2\left(\nabla v_2 + (\nabla v_2)^T\right) - p_2 \bar{\bar{I}}\right) = 0 \qquad (16)$$

Electric potential

At AF: $V = -\dfrac{V_0}{2} sin(\omega t)$ (17)

At ED: $V = \dfrac{V_0}{2} sin(\omega t)$ (18)

At FE: Electrical insulation $n \cdot \bar{j} = 0$

At BJC and GIH: The Gauss Law

$$-\varepsilon_1 \varepsilon_0 \frac{\partial V}{\partial n} + \varepsilon_2 \varepsilon_0 \frac{\partial V}{\partial n} = \sigma \qquad (19)$$

The charge balance equation

$$-\kappa_1 \frac{\partial V}{\partial n} + \kappa_2 \frac{\partial V}{\partial n} = \frac{\partial \sigma}{\partial t} \qquad (20)$$

The above two boundary condition equations are incorporated in the governing equation. Here, we are not imposing explicitly any boundary conditions at the interface.

3. Results and Discussions

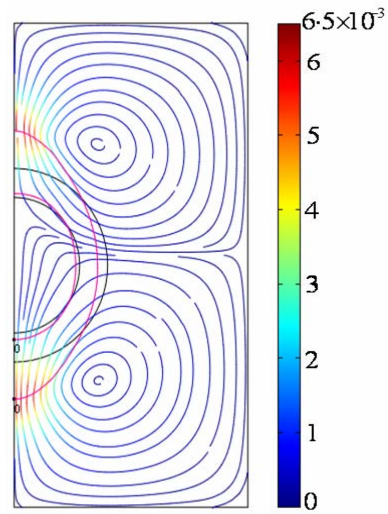

Figure 3. Simulated velocity field at t= 0.96 s. $V_0 = 1000$ V.

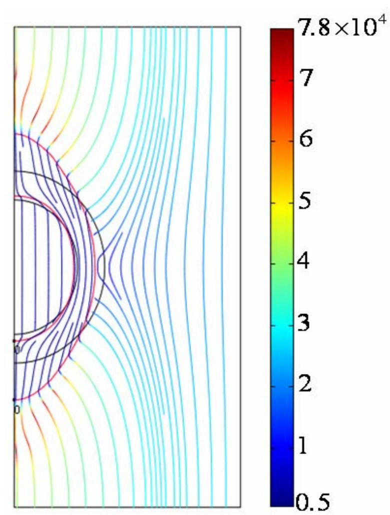

Figure 4. Simulated electric field at t= 0.96 s. $V_0 = 1000$ V.

Figure -3 and 4 show the velocity field and the electric field respectively, at one instant of time. Two vortices are seen in Figure-3. They are generated by tangential components of the Maxwell stresses. The highest stress exits at the poles of the deformed drop and hence the fluid velocity is also highest at these points. Also the electric field is highest at the poles due to the induction effect. It is also seen that the electric field is constant inside the inner drop. Since the conductivity of the inner drop is much lower than the outer drop, no current flows through the inner drop and hence it acts as a dielectric sphere.

Figure 5 shows deformation D of drops as a function of time, in the presence and the absence of the inner drop. Figure 5a corresponds to the amplitude of potential of $V_0 = 800$ V. and Figure 5b corresponds to the amplitude of potential of $V_0 = 1000$ V.

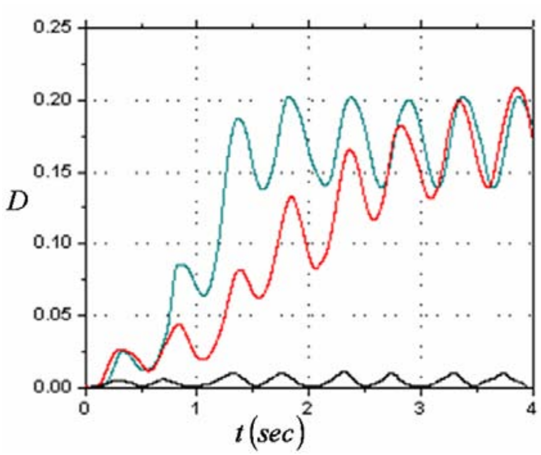

Figure 5a Deformation of the drops in electric field. $V_0 = 800$ V. Green line shows the deformation of the outer drop in the absence of the inner drop. Red line shows the deformation of the outer drop in the presence of the inner drop and the Black line represents the deformation of the inner drop.

Deformation is defined as

$$D = \frac{L - B}{L + B} \tag{21}$$

Where L and B is the lengths of the major and minor axes of the deformed drop, respectively,

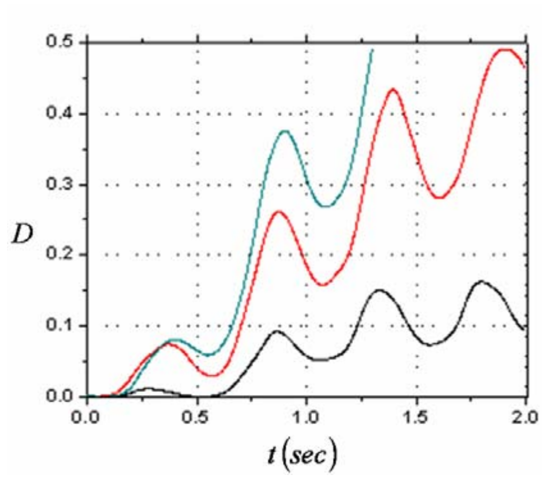

Figure 5b Deformation of the drops in electric field. $V_0 = 1000$ V. Green line shows the deformation of the outer drop in the absence of the inner drop. Red line shows the deformation of the outer drop in the presence of the inner drop and the Black line represents the deformation of the inner drop.

It is seen from these figures that deformation oscillates with the field. However, the deformed drop is not restored to its original shape when the field becomes zero. Some residual deformation is retained. As a result, its deformation increases with every cycle, till a steady oscillatory deformation is attained. In the absence of the inner drop, the steady state is regained faster than in the presence of the inner drop. The reason is that the inner drop produces a retardation effect on the deformation of the outer drop due to phase lag between its own deformation and that of the outer drop. This phase lag is caused by the difference in the conductivities of the two drops.

At higher fields, the deformation becomes unbounded as seen in Figure 5 b. It is important to note that this transition occurs over a narrow range of the electric field.

The deformation of the inner drop is significantly lower than that of the outer drop. The reason is, the outer drop acts as an electric shield, thereby reducing the filed intensity in the vicinity of the inner drop. The shield effect progressively reduces as the field intensity increases. Thus, at high field strengths the deformation of the inner drop is substantial as shown in Figure 5b .

5. Conclusions

From these studies, we find that the deformation of the composite drop in oscillating electric field is governed by a variety of factors. The inner drop retards the deformation of the outer drop and hence delays the attainment of the steady state. On the other hand, the outer drop acts as an electric shield and reduces the deformation of the inner drop. The shielding effect diminishes as the field intensity is increased.

Note that these results are valid only for the chosen fluid properties and do not cover the entire range of the fluid properties.

8. References

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9. Acknowledgements

This research was supported by Board of Research in Nuclear Science (BRNS) and Unilever Industries Limited.