

Numerical and Experimental Investigation of Water Droplet Electrical Coalescence and Non-coalescence

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Abstract—A couple of water droplets suspended in dielectric oil and subjected to the action of strong electric field are the main objects of the study. The work was carried out on the basis of comparison of experimental research data on coalescence and non-coalescence with those of computer simulation. The latter was conducted in COMSOL Multiphysics software package basing on the phase field method that was modified to enable correct modeling of the space charge localization area, which is disregarded in most papers on the topic. The investigation yielded experimental data sets with complete description of all essential parameters, which can be used to verify quantitatively existing computer models. The results obtained in the modified computer model agree well with the experimental data.

I. INTRODUCTION

Electrophysics processes proceeding in two-phase immiscible fluids are of great interest owing to they underlie various up-to-date techniques, in particular, those for electrocleaning water-oil emulsions [1], merging liquid microscopic volumes for conducting biochemical analyses [2], and creating medicaments when the coagulation of super small quantity of active liquid substances is necessary. The corresponding process, when conductive droplets suspended in a low-conducting liquid can be merged under the effect of the electric field, is called electrocoalescence whereas the reverse situation when conductive droplets in the electric field are repelled is referred to as non-coalescence.

The basic physics of two these phenomena is quite complicated, which restrains the development of the corresponding technologies. Though there are a lot of high-quality experimental results (e.g., [3–5]), gaining more insight into the problem calls for reliable computer models whereas the generally used approaches lead frequently to partially incorrect results (see, e.g., [6,7]) when the electric field emerges inside the region of a conducting liquid.

Actually, the computer simulation of the corresponding processes is a quite complicated task due to sharp change in liquid properties and very specific area of force action: a very thin layer at the interface. There are two main approaches to simulate hydrodynamics of two-phase immiscible fluids: the arbitrary Lagrangian–Eulerian method (also re-

ferred to as moving mesh) [8] and phase field / level set method [9–11]. The former one (moving mesh) fails to compute coalescence of liquid droplets though it is the most widespread approach to calculate the deformation of a conducting liquid bulk under the action of the electric field (i.e., electrodeformation) [12,13]. In turn, the latter one (the phase field method) allow simulating the electrocoalescence; however, it has problems with an artificial charge «runaway» from the interface when there is a big difference in the electrical conductivity of two interfacial fluids [14]. The issue lead as well to the error in the computed value of the threshold between coalescence and non-coalescence even when improved model was used [15]. However, presently, the situation changed due to a new idea that is described hereinafter and yielded a good quantitative agreement with the experimental data.

II. EXPERIMENTAL AND SIMULATION TECHNIQUES

The work was carried out on the basis of comparison of experimental research data on coalescence and non-coalescence with those of computer simulation. A couple of water droplets suspended in dielectric (olive) oil and subjected to the action of strong electric field are the main objects of the study.

A. Experimental Setup

The corresponding properties for the olive oil / water couple are presented in Table 1. The olive oil was chosen as a dielectric liquid owing to the following. Firstly, it shows comparably high conductivity (with specific charge relaxation time about 1 s), which ensures the value of total charge to be zero at every droplets (before voltage turn-on); secondly, there is quite a reliable and reproducible value of interfacial tension for olive oil / water couple [16,17].

TABLE 1: PROPERTIES OF WORKING LIQUIDS

Property / Liquid	Olive oil	Distilled water
Mass density ρ [kg/m^3]	910	998
Relative permittivity ϵ	2.85	80
Dynamic viscosity η [$\text{Pa}\cdot\text{s}$]	0.065	0.91×10^{-3}
Electrical conductivity σ [S/m]	1.9×10^{-11}	10^{-4}
Interfacial tension γ [N/m]	0.016	

Fig. 1 shows the schematic diagram of the experimental set-up. The latter consists of test cell filled with a dielectric liquid, high-voltage source VIDN-30, digital-to-analog convertor (for voltage modulation), high-speed camera VS-Fast/G6, two micro syringes, and PC. Designing the test cell plays a key role in the research. The basic part of the set-up is a parallel-plate capacitor filled with a liquid dielectric. To obtain the data, undistorted by edge effects, the electrical field distribution should be uniform, which may be achieved by selection of the electrode size and electrode spacing. The absence of edge effects allows simulation of electrocoalescence in the two-dimensional axisymmetric statement, which noticeably reduces the computation time.

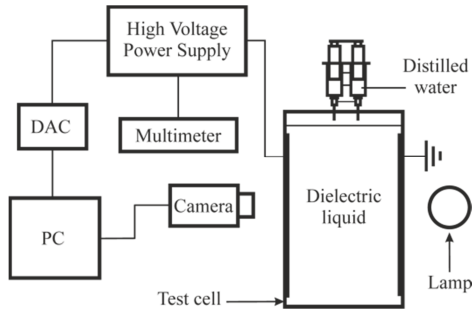


Fig. 1. Schematic diagram of the experimental setup.

Fig. 2 shows the schematic of the experimental cell and the distribution of the electric field strength for the final selection of sizes — electrode depth, height, and gap are 7, 8, and 3 cm, respectively. As can be seen, the field lines in the observation area are parallel, and the electric field distribution is uniform. The corresponding result is achieved when the lateral dimensions of planar electrodes are at least twice the length of the interelectrode gap. It should be noted that the design of the setup dealt with the most "difficult" (in terms of manifestation of edge effects) case, where the dielectric constant is 1. This was done so as to investigate liquids with different permittivity values later.

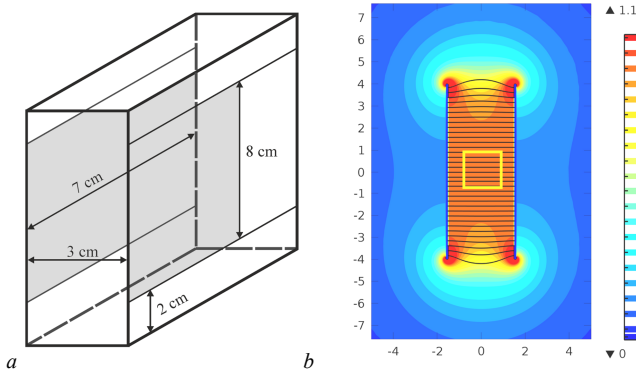


Fig. 2. Schematic of the experimental cell (a) and the computed distribution of the electric field normalized to its average value in the test cell for the final selection of electrode size and gap (b); yellow rectangle in the center represent the field of view.

B. Simulation Technique

To simulate electrocoalescence the phase field method was used. The technique bases on the calculation of the phase function distribution according to the following set of equations [18]:

$$\frac{\partial \phi}{\partial t} + \nabla(\vec{v}\phi) = \nabla\lambda\chi\nabla\psi \quad (1)$$

$$\lambda = \frac{3\varepsilon_{pf}\gamma}{\sqrt{8}} \quad (2)$$

$$\psi = -\nabla(\varepsilon_{pf}^2 \nabla \phi) + (\phi^2 - 1)\phi, \quad (3)$$

where t is the time, \vec{v} is the fluid velocity, λ is the mixing energy density, χ is the mobility turning parameter, ε_{pf} is the interface thickness, and γ is the interfacial tension. Equations (1), (3) are the decomposition of the fourth-order Cahn-Hilliard equation (which describes the process of phase separation) into two second-order ones. Parameter χ determines the time scale of the Cahn-Hilliard diffusion and thereby also governs the diffusion-related time scale of the interface. The value $\chi = 0.001 \text{ m}\times\text{s}/\text{kg}$ is used in the simulation. In turn, ε_{pf} should be set not greater than $D/50$ (where D is the droplet diameter) to obtain reliable numerical results.

The volume fraction of fluid 2 is computed as:

$$V_f = \min(\max\left(\left[\frac{1+\phi}{2}\right], 0\right), 1),$$

where the min and max operators are used so that the volume fraction has a lower limit of 0 and an upper limit of 1. There is an interface between two media in the computer model where V_f function changes from 0 (that corresponds to pure oil) to 1 (pure water).

To simulate electrophysics processes in two-phase medium, the model was supplemented with the following equations describing electric current passage and hydrodynamics:

$$\text{div } \vec{j} = 0 \quad (4)$$

$$\vec{j} = \sigma \vec{E} \quad (5)$$

$$\vec{E} = -\nabla V \quad (6)$$

$$\rho \frac{\partial \vec{v}}{\partial t} + \rho(\vec{v}, \nabla)\vec{v} = -\nabla P + \eta \nabla^2 \vec{v} + \vec{f}_{st} + \vec{f}_e \quad (7)$$

$$\text{div } \vec{v} = 0 \quad (8)$$

$$\vec{f}_{st} = \frac{\lambda \psi}{\varepsilon_{pf}^2} \nabla \phi \quad (9)$$

$$\vec{f}_e = E^2 \nabla \varepsilon \varepsilon_0 - \varepsilon \varepsilon_0 E^2 \nabla \sigma / \sigma, \quad (10)$$

where ρ , η , σ , ε , and γ are liquid properties (given in Table 1), P is the pressure, \vec{f}_{st} is the surface tension force [18], \vec{f}_e is the Coulomb force, \vec{j} is the electric current density, \vec{E} is the electric field strength, ϕ is the electric potential, and ε_0 is the vacuum permittivity.

The following non-linear relation between liquid properties and the volume fraction of water was used to avoid the problem of space charge “runaway” mentioned above [14]:

$$\sigma = C_1 \exp(C_2 V_f^2), \quad (10)$$

where C_1 and C_2 are the constants used to satisfy the following conditions:

$$\sigma(V_f = 0) = \sigma_{oil}$$

$$\sigma(V_f = 1) = \sigma_{water}.$$

The similar dependences were used to describe the change in other liquid properties.

III. RESULTS AND DISCUSSION

Though the corresponding approach, based on the non-linear relation between liquid properties, yielded acceptable results for droplet electrodeformation [14], it failed to satisfy experimental data on electrical coalescence [15]. The problem is in a small shift of space charge density in the outside direction from the interface, which is demonstrated in Fig. 3a, where space charge locates between levels $V_f = 0.5$ and 0.9 instead of $V_f = 0.1$ and 0.9 (see Fig. 3b). The latter leads to an improper increase in droplet diameter, thus distorting the model.

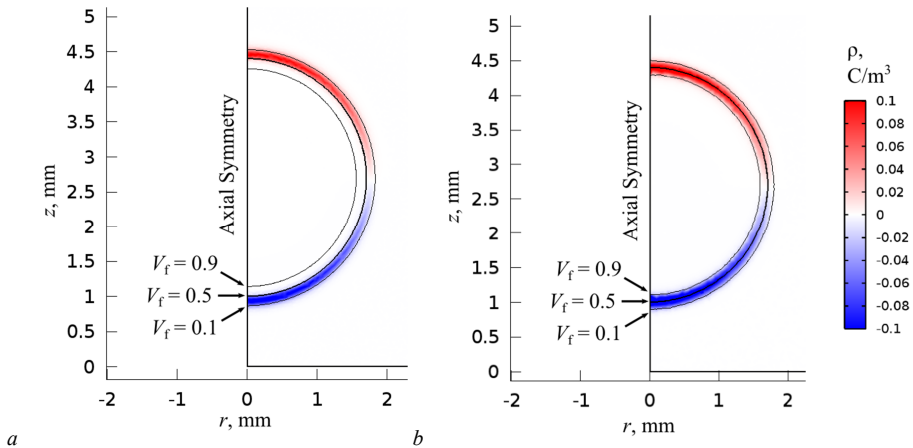


Fig. 3. Space charge distribution near the interface separating two immiscible liquids for two models: without (a) and with (b) the correction of the electrical conductivity of a dielectric liquid.

The following idea is used to resolve the problem. Since the electric field inside the conductive droplet screened nearly completely, there is no matter what is the value of electrical conductivity of suspending fluid if it is still much less than that of the droplet. Fig. 4 gives an example of the normalized electric field strength distribution for the case $\sigma_{water}/\sigma_{oil} = 10^3$ and $\sigma_{water}/\sigma_{oil} = 10^5$ (Fig. 4a and b) and the ratio of the maximum value of the electric field strength inside the droplet to its average value for $\sigma_{water}/\sigma_{oil} \in [10^0, 10^5]$ (Fig. 4c). The corresponding ratio is negligible and the electric field lines remain unchanged when the electric conductivity of the suspending phase $\geq 10^3$ times less.

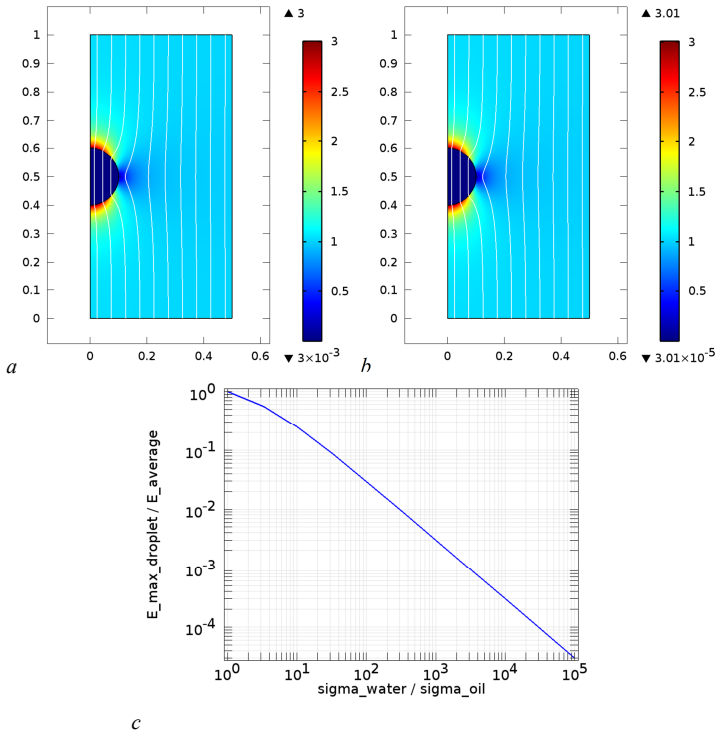


Fig. 4. The normalized electric field strength distribution for the case $\sigma_{\text{water}}/\sigma_{\text{oil}} = 10^3$ (a) and $\sigma_{\text{water}}/\sigma_{\text{oil}} = 10^5$ (b) and the ratio of the maximum value of the electric field strength inside the droplet to its average value for $\sigma_{\text{water}}/\sigma_{\text{oil}} \in [10^0, 10^5]$ (c).

Thus, the conductivity of dielectric liquid can be artificially raised without the distortion of the electric field distribution, which, in turn, reduces the conductivity difference between two phases and diminishes the problem of the artificial shift of space charge. Fig. 3b shows the distribution of the latter obtained in the modified model with $\sigma_{\text{water}}/\sigma_{\text{oil}} = 10^3$. As can be seen, the simulation yields quite a correct distribution with space charge density being localized near the level $V_f = 0.5$, i.e. exactly around the interface without a shift in one or another direction.

The suggested modified computer model was verified experimentally. Fig. 5 represents the compilation of the experimental data and shows the droplet radius dependence of the electric field threshold when the transition between coalescence (black circles) and non-coalescence (red crosses) takes place. Here, the radii of both droplets are believed to be the equal. The initial distance between droplets (the gap) lies mostly within 2–3 mm; however, some results were observed for less gap (within 1–2 mm). The latter data were reanalyzed basing on the computer simulation and the following conclusion was drawn: if two droplets are located so close to each other (at the time instant of voltage turn-on) that touching process takes place earlier than electrodeformation ends, coalescence can occur instead of non-coalescence. The corresponding experiments yielded coalescence and the data are believed to be distorted and marked with filled black circles.

(namely, the dependence of the experiment outcome on initial conditions that are frequently disregarded) and is to be taken into account when analyzing the observed results. On the other hand, it shows the possibility to increase the operating voltage of electrocoalescer when applying pulsed voltage.

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