Electro-coalescence of an aqueous droplet at an oil–water interface

M. Mousavichoubeha,b, M. Ghadiri,a,* M. Shariati-Niasarb

a Institute of Particle Science and Engineering, University of Leeds, Leeds LS2 9JT, UK
b School of Chemical Engineering, College of Engineering, University of Tehran, 11365-4563 Tehran, Iran

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ABSTRACT

The coalescence of an aqueous droplet at an oil–water interface under an electric field has been investigated, with a view to quantify conditions that give rise to secondary droplet formation. Two patterns of drop-interface coalescence may occur: complete coalescence and partial coalescence. The former is obviously the desirable pattern for industrial coalescers. However in practice, the process of coalescence could actually produce smaller droplets, which become more difficult to remove, and hence undesirable. This is caused by either necking, due to extensive elongation of the droplet, or reaction to a fast and energetic coalescence and is referred to as partial coalescence. The volume of the droplets formed in this way has been analyzed as a function of the initial droplet size, electric field strength and the distance between the droplet and the interface. The expansion speed of the neck connecting the droplet and interface at the beginning of the pumping process has also been quantified. These results are useful in optimizing the electro-coalescence process.

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1. Introduction

In the chemical, processing and manufacturing industries, immiscible liquids are often mixed such that one phase is fully dispersed in another, e.g. in extraction and leaching. The aim is to get a large interfacial area for the enhancement of mass transfer between the two immiscible liquids [1–4]. In crude oil extraction from oil wells [5], an aqueous saline phase is often well-dispersed in the crude oil. However these emulsions or dispersions have to be separated into their constituent phases before the next operating steps or as required by process requirements, environmental regulations and customer specifications as in the case of crude oil industry [6]. There are several techniques for enhancing the separation of water-in-oil emulsions, such as the addition of chemical demulsifier [7], pH adjustment and filtration [8], gravity or centrifugal settling [9], heat treatment and electrostatic demulsification [10,11]. In terms of energy efficiency, electrostatic demulsification is considered to be the best among the above methods [10]. The state of the art has been reviewed by Eow and Ghadiri [6]. Here the rate of coalescence can significantly be enhanced by the application of an electric field. The coalescence occurs in three stages [9,12,13]. In the first stage, the drops approach each other or the interface and are separated by a film of the continuous phase. The second stage involves the thinning of this film. When the film reaches a critical thickness any disturbance or instability causes it to rupture, following which coalescence occurs [14–16]. Film thinning is often the overall controlling step in the absence of an electric field. In order to increase the separation rate, the film-thinning process needs to be faster and this can be done by the application of an electric field. High electric fields have been used to separate water-in-oil dispersions in crude oil and extraction industries [6]. To apply this method, the continuous phase needs to be much more electrically insulating compared with the dispersed phase, in order to set up an electric field [17]. The current understanding of the electrocoalescence phenomenon has been reviewed by Eow et al. [18].

2. The effect of applied electric field on drop interface coalescence

In the absence of an electric field the coalescence of a drop at a liquid–liquid interface sometimes produces smaller drops as observed by Charles and Mason [19,20]. During the rupture of the film between the main drop and the interface, the excess internal pressure from the curved interface produces a cylindrical liquid column. The radius of this column decreases rapidly until its circumference becomes smaller than its height. As a result of a Rayleigh wave disturbance, a secondary droplet is often formed [20]. The mean rest-time of a drop at an interface can be significantly reduced by applying an electric field because the rate of film thinning is increased [19–22]. It has been reported that in the presence of an external electric field, the generation of the secondary droplet does not occur [19,20,22]. Aryafar and Kavehpour [23] have recently proposed that the partial coalescence in the absence of an electric field may be described by Ohnesorge number,
The properties of the liquid used in the experiment.

<table>
<thead>
<tr>
<th>Liquids</th>
<th>Conductivity ($\mu$S m$^{-1}$) ($\pm$5%)</th>
<th>Viscosity (mPa s) ($\pm$5%)</th>
<th>Surface tension (mN m$^{-1}$) ($\pm$5%)</th>
<th>Density (kg m$^{-3}$) ($\pm$5%)</th>
<th>Dielectric constant</th>
</tr>
</thead>
<tbody>
<tr>
<td>De-ionized water</td>
<td>1.1 x 10$^{-3}$</td>
<td>1.00</td>
<td>73</td>
<td>1196</td>
<td>80</td>
</tr>
<tr>
<td>Sunflower oil</td>
<td>3.6 x 10$^{-6}$</td>
<td>46.5</td>
<td>33</td>
<td>998</td>
<td>4.9</td>
</tr>
</tbody>
</table>

* The measured surface tension is with respect to air at 1 atm and 20°C.

3. Objectives of the present work

For the separation of water-in-oil emulsions, the smaller the dispersed phase droplet size is, the more difficult would the separation be. An important problem in this separation process is the formation of the secondary droplet by partial coalescence. This leads to a lowering of the separation efficiency as the much tinier droplets are more difficult to separate. Therefore basically it is better to prevent these secondary droplets from forming in the first place. In this work our observations on the formation of the secondary droplets during the electro-coalescence of the primary droplets are reported and the parameters that affect the process are quantified.

4. Experimental set-up and procedure

The experimental cell used in this work is shown in Fig. 1. The cell was made of Perspex to facilitate visualization of the phenomenon. The electrodes were polished brass plates. The plates had dimensions of 90 mm x 25 mm. The high voltage electrode was attached to the moveable upper part of the cell. The distance between the two electrodes could therefore be varied by moving the upper part up or down although it was set at 53 mm in this work. The Perspex block had a thickness of about 6 mm. There is a small hole through the middle point of the moveable upper part of the cell and the brass plate for a hypodermic needle to go through it. The needle attached to a syringe (Hamilton micro-liter syringe) was used to produce small aqueous droplets in the cell. The high voltage electrode was connected to a positive polarity high voltage direct current source (Model: PS/EH60R01.5-22, manufactured by Glassman High Voltage Inc.). The bottom electrode was grounded.

5. Results and discussion

5.1. The mechanism of drop-interface coalescence under electric field

The coalescence events of water droplets at an oil/water interface were recorded at 20,000 fps and are shown in the sequence of images in Figs. 2 and 3. In these figures, the length scale is the same for all images, facilitating the comparison between the primary droplet and the secondary droplets. The deformation of the droplet and interface before coalescence in the absence of an electric field results in the

![Diagram](image-url)
local deformation of interface and falling droplet (see Fig. 4(b)). An uncharged droplet subject to an electric field is polarized and when the deformed droplet approaching the deformed interface the electric field strength increases exponentially at small separations to the extent that electroclamping phenomenon becomes operative [27] giving rise to the neck formation as shown in Fig. 4(b). The high speed of video recording has made this observation very clear. When the droplet is sufficiently close to the interface (a few micrometers apart) in a time period less than 16 μs the high strength electric field in the gap between the droplet and interface causes clamping between them (Fig. 4(b)), resulting in the formation of a narrow channel. The droplet will now be acquiring the same charge as the adjacent electrode and will be experiencing a repelling Coulombic force. For a solid electrode plate, the droplet will be repelled from the electrode. However for an aqueous liquid interface, the surface tension will be pushing the liquid in the droplet into the continuous phase via the channel formed, and rapidly enlarging the neck. It is likely that the current constriction of the electrical clamping process ruptures the thin film (oil phase) between the droplet and the interface. A hole is formed in this way at the interface by which the process of coalescence is initiated. The hole expands very rapidly and the liquid is pumped rapidly into its bulk phase. Two patterns of coalescence are observed here: "complete coalescence" and "partial coalescence", as shown in Figs. 2 and 3, respectively.

There are two rate processes operating: pumping of droplet into its bulk phase (due to surface tension) and the necking process. Whether a secondary droplet is formed depends on the process which is dominating. The predominance of each of these processes, i.e. necking and pumping depends on some parameters and will be discussed later.

5.2. Complete and partial coalescence patterns

It can be seen from Fig. 2 that with the start of droplet pumping into its bulk phase (due to surface tension) and the necking process. Whether a secondary droplet is formed depends on the process which is dominating. The predominance of each of these processes, i.e. necking and pumping depends on some parameters and will be discussed later.

5.3. Factors affecting the volume of the secondary droplet

High speed video observations indicate that three parameters are influential in determining the coalescence pattern: droplet size (d), electric field strength (E) and the height of falling droplet from interface (λ). In this work the dispersed phase is de-ionized water and does not contain any surfactants which could affect the interfacial tension.

5.3.1. The effect of primary droplet size

The effect of droplet size in the range 576 ± 1–1196 ± 4 μm on the volume of the secondary droplet has been investigated for constant electric field strength and λ. The results are shown in Fig. 5(a)–(d).
As observed from these figures, by increasing the primary droplet size (d) the volume of detached body increases, except for λ = 0 (not shown here) where the coalescence is complete. It means when the droplet is sitting on the interface there will not be any detached body and all coalescence processes lead to a complete coalescence pattern. The formation of bigger detached bodies as the primary droplet size is increased can be explained by considering the effect of interfacial tension. According to Young-Laplace, $\Delta p = \frac{2\sigma}{R}$, where $\Delta p$, $\sigma$ and $R$ are internal pressure, interfacial tension and the droplet radius, the bigger droplets have lower internal pressure, as compared with the smaller ones [28]. They will therefore be more deformable and form a long neck more readily. Another factor is that the polarization of larger droplets can be more effective in producing a neck. Thus less rigidity and larger attractive force by the far electrode on a bigger droplet result in detaching a bigger volume from primary droplets.

5.3.2. The effect of electric field strength

As the results of Fig. 5(a)-(d) can be re-plotted in terms of the electric field strength to show its trend, one of which is shown in Fig. 6. It can be seen that when the falling droplet is subjected to higher

Fig. 3. Partial coalescence of a droplet of 1196 ± 4 μm diameter under two electric field strengths: (a) 124 V/mm and (b) 181 V/mm.

Fig. 4. Deformation and start of coalescence of a 1196 ± 4 μm droplet with an interface under the electric field strength of 181 V/mm, showing the formation of a narrow channel.
Fig. 6. The effect of electric field strengths on detached body volume for various drop size and $\lambda = 101 \pm 10 \mu m$. 

As mentioned, the volume of detached body is being controlled by the mutual interaction between the necking and pumping process. Eow et al. [25] have shown that by increasing the electric field strength the deformation of a given droplet size increases.

Table 2

<table>
<thead>
<tr>
<th>Electric field strength (V/mm)</th>
<th>90</th>
<th>124</th>
<th>158</th>
<th>181</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D = d_{maj}/d_{min}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.058</td>
<td>1.075</td>
<td>1.154</td>
<td>1.158</td>
<td></td>
</tr>
</tbody>
</table>

As shown in Fig. 7 and Table 2, by increasing the strength of electric field from 90 V/mm to 181 V/mm the deformation of the droplet with the diameter of 984 ± 2 μm increases and that the side of droplet close to the interface gets more elongated locally under higher electric fields. This has previously been shown by Eow et al. [25]. The deformations in Table 2 are the ratio of the major to minor diameters of the drop.

As the narrow channel forms between droplet and interface the pumping process competes with the necking, with the latter brought about by the change in droplet polarity, due to contact, and consequent repelling from the adjacent electrode. So the pumping process is not able to overcome the necking process. In this state and for a given droplet size and falling height, the volume of detached body in a higher electric field will be bigger than that for lower elec-
The effect of height of droplet from interface ($\lambda$)

The data of Fig. 5(a)–(d) can be expressed in terms of $\lambda$ to see its effect on secondary droplet volume clearly. This is done for one electric field strength in Fig. 10. Increasing the height of falling droplet from interface for a given droplet size and under a constant electric field strength shows the radial change ($dr_{\text{channel}}/dt$) and “equator reduction” ($dr_{\text{equator}}/dt$) and “peak falling” ($d\lambda_{\text{peak}}/dt$) show the radial reduction and falling of peak point of droplet respectively. To understand the pumping behavior of the droplet these three speeds should be considered at the same time. As it can be seen in Fig. 9, at the beginning of the coalescence by formation of the narrow channel between droplet and interface, the speed of channel expansion is very fast (in this case about 245 mm/s) and it decreases continuously. Moreover, the speeds of equator reduction and peak falling are initially zero.

5.3.3. The effect of height of droplet from interface ($\lambda$)

The droplets located further from the interface, i.e. the larger value of $\lambda$, experience stronger attractive Columbic forces ($F \propto 1/r^2$ where $r$ is the distance between charge sources) exerted by the high potential electrode. So less rigidity and larger Columbic force helps necking process significantly.

6. Conclusions

Drop-interface coalescence under an electric field could lead to the formation of a detached body. This is of course highly undesirable, and should be avoided by optimizing the process. The parameters that affect the formation of detached body have been analyzed. These are the droplet size, electric field strength and distance of droplets from the electrodes. The results show that by increasing any of these parameters the volume of detached body increases. This information is useful in optimizing the electro-coalescence process. For example, by the use of appropriate pulsatile electric field it could be possible to enhance the electro-coalescence and at the same time suppress the formation of secondary droplets.

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