

Electro-Coalescence of an Aqueous Droplet at an Oil–Water Interface with an Investigation of Secondary Droplets Formation

S. H. Mousavi¹, M. Shariaty-Niassar^{1}, H. Bahmanyar¹, M. A. Moosavian¹*
School of Chemical Engineering, College of Engineering, University of Tehran, 11365-4563 Tehran, Iran

Abstract

The coalescence of water droplets in oils may be enhanced by application of an electric field. This approach is commonly used in the crude oil and petroleum industry to separate water from crude oil extracted from oil well. By application of an electric field 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 change in interface tension between two phases as a result of surface active agents. There is a considerable growth in secondary droplets volume. Expansion speed of the neck connecting the droplet and interface at the beginning of the pumping process has also been quantified and partial coalescence has been explained as a result of competition between pumping and necking processes. These results are useful in optimizing the electro-coalescence process.

Keywords: *Coalescence, Electric Field, Electro-Coalescer, Interfacial Tension, Pumping Process*

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 obtain a large interfacial area for the enhancement of mass transfer between the two immiscible liquids [1-3]. In the petroleum industries, oil is usually found in the form of a water-in-oil

emulsion following the extraction stage [4]. Formation of emulsions takes place due to flow through porous media [5, 6], through shearing within choke valves and due to pressure drop resulting from pumping action in oil wells [4, 7]. The presence of natural surface-active agents such as resins and asphaltenes in crude oil aids the formation of stable water-in-oil emulsions [5, 8] by reducing the interfacial tension, thereby

* Corresponding author: mshariat@ut.ac.ir

promoting dispersion and emulsification of the droplets [9]. Such emulsions, despite being thermodynamically unstable can be kinetically very stable for long periods of time [5] and this is obviously undesirable. During the production life of an oil well, the water content increases with the age of the well [10]. To meet international specifications, and to produce an oil phase of sufficient quality to allow refinement, the oil and water have to be separated with the oil having less than 0.5% water content. Gravity separation is very difficult as the densities of oil and water are close; moreover, the high viscosity of oil makes settling of the aqueous drops too slow. Faster and more effective methods have therefore been developed [11], such as chemical demulsifiers [12], freezing (cryological) technologies [13], hydro-cyclones [14], pH adjustment techniques [15], centrifugal settling [16], filtration [15], heat treatment and electrostatic demulsification [17,18]. Of the aforementioned methods, the method utilising high electric fields is based on the phenomenon of enhanced coalescence of aqueous droplets suspended in the insulating oil under the action of an electric field [19]. Most of the commercially available electro-coalescers are large and bulky, because the design is based on slow migration of droplets rather than on the coalescence process. This is due to lack of underlying fundamental knowledge of the governing effects of high electric fields on drop-drop coalescence and drop-interface coalescence [20, 21]. In the absence of an electric field, the coalescence between drops or between a drop and its own homophase in an immiscible liquid medium occurs in several stages [16, 22]. In the first

stage, the drops approach each other (in drop-drop coalescence) or a drop reaches the interface (in drop-interface coalescence) while being separated by a thin film of the continuous phase. The next stage involves the thinning of this film to reduce the interfacial area; the thinning rate is affected by the capillary pressure and disjoining pressure and can be retarded due to the Marangoni effect if surfactant is present [23]. Once the film reaches a critical thickness, any significant disturbance or instability will cause it to rupture and coalescence occurs [22, 24]; film thinning is often the overall controlling step. Applying an electric field, in addition to speeding up the migration process, it enhances the film thinning process significantly, shortening the coalescence time substantially.

However, electro-coalescence suffers from break-up and formation of secondary droplets when either the electric field strength is too high or droplets have grown too large [11, 25-28]. Aryafar and Kavehpour [25, 26] and Blanchette and Bigioni [29] have recently proposed that the above process, referred to as partial coalescence, in the absence of an electric field may be described by Ohnesorge Number, $Oh = \mu / (r\rho\sigma)^{0.5}$, where μ is viscosity of dispersed phase, r is the radius of the drop, ρ is density and σ is interfacial surface tension. For $Oh > 1$ full coalescence occurs and for $Oh < 1$ only partial coalescence takes place.

2. Scope of the present work

Separation of water-in-oil emulsions becomes increasingly difficult with decreasing aqueous droplets size. Formation of secondary droplets by partial coalescence

causes problems, leading to decreased separation efficiency due to the smaller droplet sizes. Therefore preventing these secondary droplets from forming in the first instance would be beneficial. In this work we report our observations on the formation of secondary droplets during electro-coalescence of primary droplets. The effects of the most important factors, i.e. primary droplet size, interfacial tension and electric field strength are analysed. Also, we have made a closed observation on a single aqueous droplet partial coalescence.

3. Experimental set-up and procedures

The experimental cell used in this work is shown in Fig. 1. The cell was made of Perspex to facilitate visualisation of the phenomenon. The electrodes were polished brass plates with dimensions of 90 mm × 25 mm. The high voltage electrode was attached to the upper part of the cell, which was set at 51 ± 0.5 mm from the grounded electrode in this work. The Perspex block had a thickness of approximately 6 mm. There is a small hole through the mid-point of the upper part of the cell and brass plate to allow a hypodermic needle to pass through it. The needle, attached to a syringe (Hamilton micro-liter syringe) was used to produce small aqueous droplets in the cell. The hypodermic needle was grounded in order to produce uncharged drops. The high voltage electrode was connected to a positive polarity high voltage direct current source (TREK 20/20C). The bottom electrode was grounded. A high-speed digital video camera (Photron FASTCAM SA5), equipped with a micro lens (NAVITAR 12× Zoom Lens) was used

to observe the phenomena during the drop-interface coalescence. This camera was used with a frame speed of 20000 fps. The video camera was focused on the centre of the liquid-liquid interface. A halogen lamp (Dedolight DLHM4-300) with four flexible fibre optic heads was used for lighting; the intensity of the lighting could be accurately adjusted to facilitate focusing.

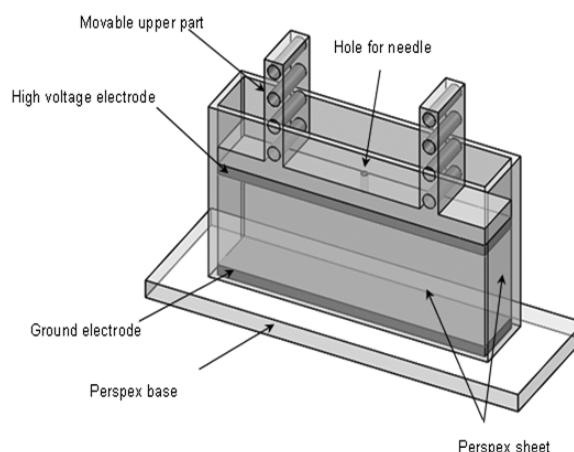


Figure 1. Test Cell [30].

De-ionized water was used as the dispersed phase, while the continuous phase was sunflower oil (obtained from Morrisons, UK Ltd). The properties of the liquids used in this research and interfacial tension values for solutions are given in Tables 1 and 2 respectively.

The conductivity was measured using a conductivity meter, Model 470 from JENWAY Products Inc. The voltage applied to the upper electrode was between 1 kV to 11 kV. The current was measured using an electrometer, Model 6514 from Keithley Instruments. The current-applied voltage relationship for sunflower and the solutions including Span 80 followed Ohm's law as can be seen in Fig. 2.

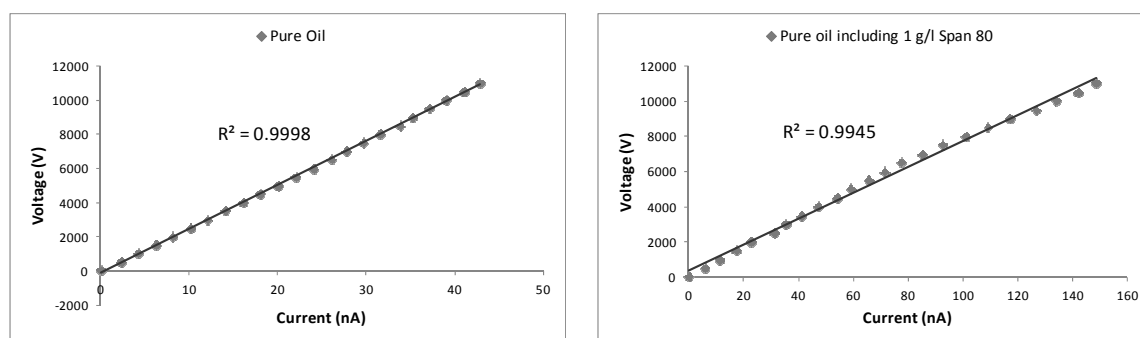
Table 1. The properties of the liquid used in the experiments.

Liquids	Conductivity (μSm^{-1}) ($\pm 5\%$)	Viscosity (mPa.s) ($\pm 5\%$)	Density (kgm^{-3}) ($\pm 5\%$)	Dielectric Constant
Deionised water	5.49	1.00	1000	80
Sunflower oil	7.62×10^{-5}	46.5	922	4.9
1 g/l Tween 20	10.2	≈ 1.00	≈ 1000	≈ 80
1 g/l SDS	242	≈ 1.00	≈ 1000	≈ 80
1 g/l Span 80	3.2×10^{-4}	≈ 46.5	≈ 922	≈ 4.9

Table 2. Interfacial tension.

Liquids	Interfacial Tension ^a (mN m^{-1})
Deionised water-Sunflower oil	25
1 g/l Tween 20 in dispersed phase- Pure sunflower oil	6
1 g/l SDS in dispersed phase- Pure sunflower oil	10
Deionised water-1 g/l Span 80 in Sunflower oil	16

^aInterfacial tension measured at 21 °C and 1 atm.

**Figure 2.** Satisfaction of Ohm's law.

Liquid viscosity was measured using Bohlin CVO rheometer (Malvern Instruments, UK). The density was measured using a volumetric flask. Interfacial tension was measured according to a technique based on the pendant drop method, using a contact-angle measuring instrument, EasyDrop from Kruss GmbH. The densities of sunflower oil and de-ionized water are quite close; this facilitates experimentation because it allows water droplets to fall slowly towards the liquid-liquid interface, thus giving sufficient

time to manage the recording process. Water droplets of different sizes were produced using hypodermic needles in droplet diameter range between 576 μm to 1196 μm . The glassware and needles were cleaned using both acetone and distilled water, this ensured any impurities with the potential to affect the coalescence phenomenon were removed. The droplets were released gently from the needle to avoid oscillation of the droplets and interface, and were then introduced at the middle of the planar

interface (sufficiently far from the edge where the interface was curved and the electric field not uniform). The diameter of the droplets was measured with different standard deviation from $\pm 1\mu\text{m}$ to $\pm 4\mu\text{m}$ by the use of Image-Pro and PFV (Photron Fastcam Viewer ver. 320) software. The diameter of the needle was measured by a microscope and was used for the in-situ calibration of the droplet size and measurements on the screen. The experiments were performed at $22\pm 2\text{ }^{\circ}\text{C}$.

4. Results and discussion

4-1. The mechanism of drop-interface coalescence under electric field

The coalescence events of water droplets at an oil/water interface were recorded at 20000 fps and are shown in the sequence of images in Figs 3 and 4.

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 has been reported previously by Charles and Mason [31] but this is notable only when the drop is resting on the interface. Applying a sufficiently strong electric field results in the local deformation of interface and falling droplet (see Fig. 5(b)). An uncharged droplet subject to an electric field is polarized and when the deformed droplet approaches the deformed interface the electric field strength increases exponentially at small separations to the extent that electroclamping phenomenon becomes operative [32], giving rise to the neck formation as shown in Fig. 5(b).

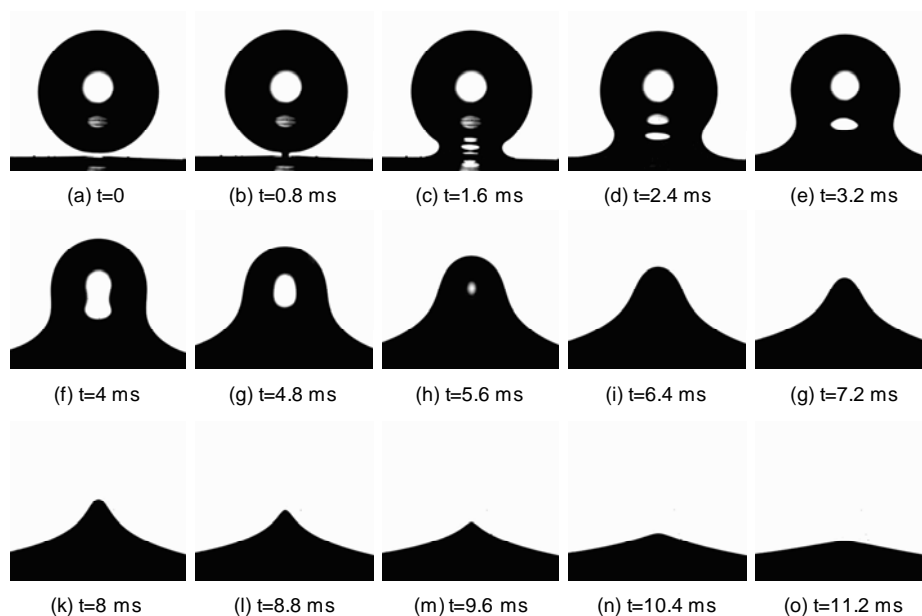


Figure 3. Sequence of a complete coalescence for a droplet of $984\pm 2\mu\text{m}$ diameter under electric field strength of 116 V/mm .

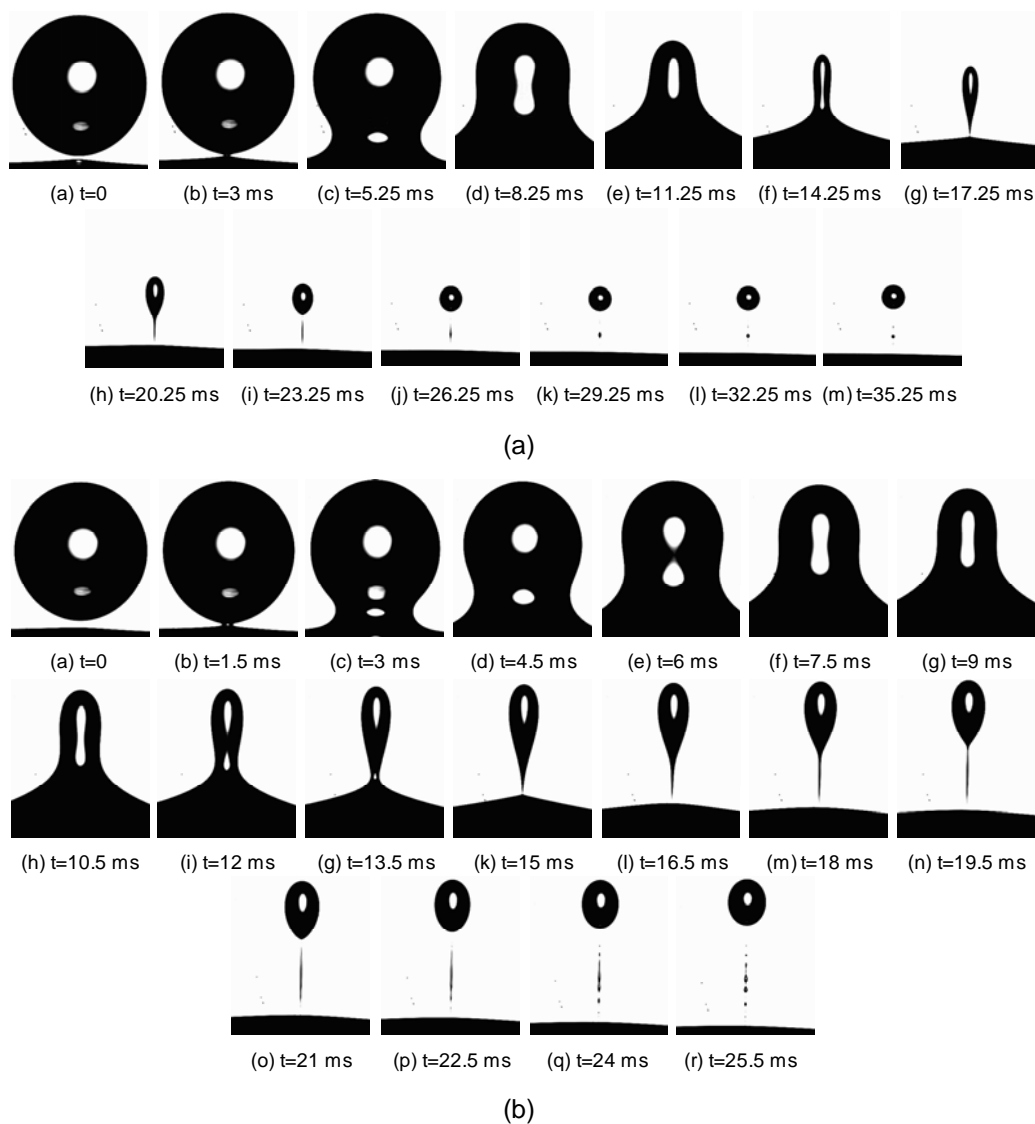


Figure 4. Partial coalescence of a droplet of 1196 ± 4 μm diameter under two electric field strengths: (a) 256 V/mm; (b) 373 V/mm.

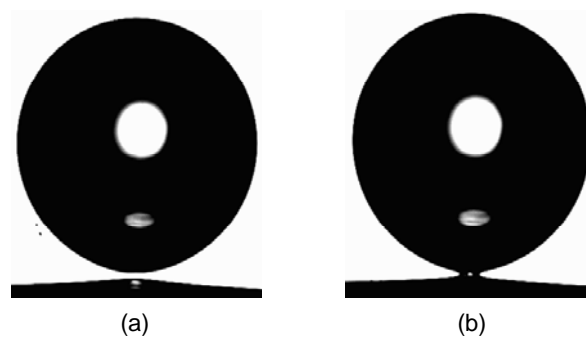


Figure 5. Deformation and start of coalescence of a 1196 ± 4 μm droplet with an interface under the electric field strength of 373 V/mm, showing the formation of a narrow channel.

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 μ s the high strength electric field in the gap between the droplet and interface causes clamping between them (Fig. 5(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 3 and 4, respectively. There are two rate processes operating: pumping of droplet into its bulk phase, due to surface tension and necking process. Whether a secondary droplet is formed depends on the dominant process. The predominance of each of these processes, i.e. necking and pumping depends on some parameters and will be discussed later.

4-2. Complete and partial coalescence patterns

It can be seen from Fig. 3 that with the start of droplet pumping into its bulk phase, no

necking occurs, leading to a complete coalescence pattern. In this coalescence pattern in some stages (Fig. 3. (l) and (m)), the peak of the droplet becomes sharp but finally, as a result of surface tension the process leads to a flat surface without any detached bodies. This means in such a condition the rate of pumping is faster than that of the necking process. Although the droplet experiences a repelling Coulombic force by the adjacent electrode, this force is not sufficiently strong (in the case of complete coalescence) to help the occurrence of necking process. Figure 4 shows the pattern of coalescence for droplets subjected to a higher electric field strength than that of Fig. 3, hence a necking process is observed. In Fig. 4(b), as a result of a faster development of the necking process in comparison with Fig. 4(a), the volume of the secondary droplet is bigger. In case (b) the repelling force between the droplet and the interface (after charging) is stronger than that for case (a) because of larger electric field strength and the necking process develops faster detaching a bigger secondary droplet. In both cases (a) and (b) the pumping process is in progress to drain the content of the primary droplet into its bulk phase but the necking process and its rate determine the volume of the secondary droplet. Thus under a given condition leading to a partial coalescence, the necking process and the pumping process are in competition with each other. Another noteworthy point is the difference between the tail length in Fig. 4 (a) and (b). In fact, in most cases when partial coalescence occurs there will be a main secondary droplet with forming a tail (see Fig. 6 for details).

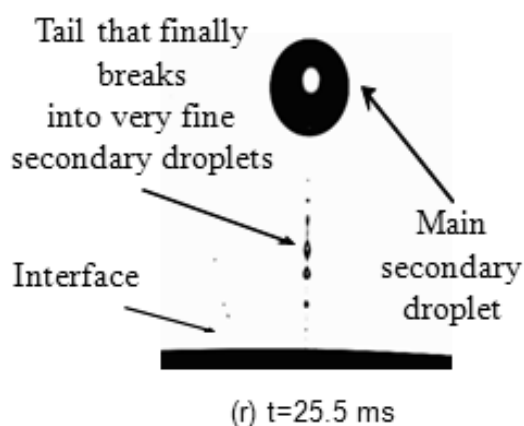


Figure 6. Illustration of the main secondary droplet and tail.

In Fig. 4(b) the droplet is subjected to higher electric field strength and the detached body has a longer tail than the other case. The secondary droplet is moving upwards, stretching the neck even further before it is broken into several much finer droplets entering the continuous phase (oil phase). In conclusion these two strong electric fields are detrimental for efficient electro-coalescers.

4-3. 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 deionised water and does not contain any surfactants which could affect the interfacial tension.

4-3-1. The effect of primary drop size and electric field strength

In the presence of an electric field and under the influence of some other parameters

(discussed later), the coalescence of single droplets with the interface usually results in the formation of secondary droplets: one main one and a number of very fine ones, with the latter appearing as a tail following the main secondary droplet (Fig. 6.). In this work, the total volume of all the secondary droplets has been measured. For these measurements it has been assumed that the shape of the droplets is symmetric around the axis passing through the droplets center, parallel to the direction of the electric field. The secondary droplets that form have enough time to return to a symmetric ellipsoidal shape from an asymmetric elongated shape and hence the volume of such bodies is easier to calculate.

Fig. 7 (a) and (b) shows the effect of primary drop size on the formation of secondary droplets volume under the action of various electric fields. The total volume of the secondary droplets has been normalized with respect to the initial drop volume. The data are for the electric field strengths in the range 116-373 V/mm, where it is observed that the ratio of volumes is relatively independent of the primary drop size at least in low electric field strength.

The data of Fig. 7 are replotted in terms of the electric field strength in Fig. 8 to show its effect. At low electric field strengths, the secondary droplet formation is small and insensitive to the droplet size. However, at higher field strength some dependency on droplet size is observed.

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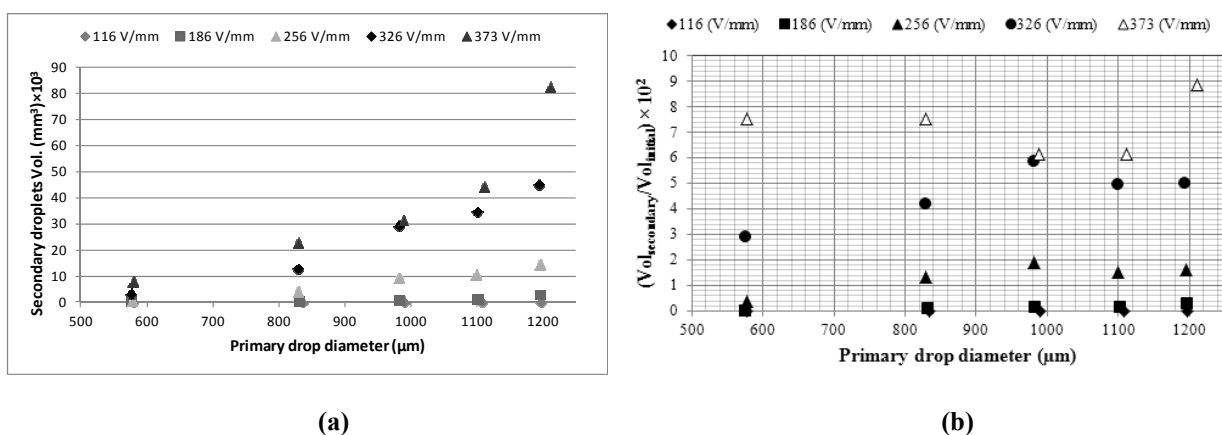


Figure 7. Effect of primary droplet diameter for a constant $\lambda = \lambda = 200 \pm 14 \mu\text{m}$ (a) Secondary droplets volume as a function of the primary droplet diameter (b) Normalized secondary droplets volume as a function of the primary droplet volume.

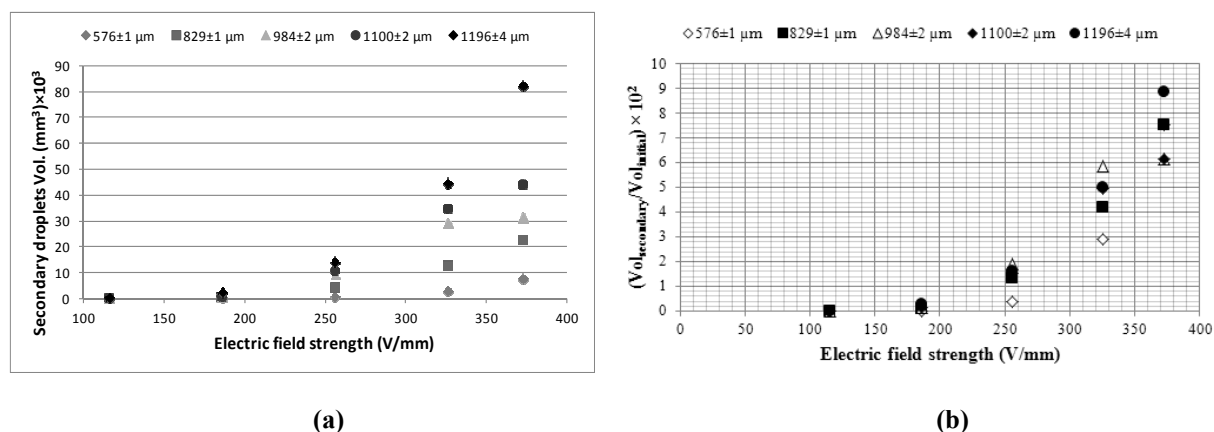


Figure 8. Effect of electric field strength for a constant $\lambda = \lambda = 200 \pm 14 \mu\text{m}$ (a) Secondary droplets volume as a function of electric field strength (b) Normalized secondary droplets volume as a function of electric field strength.

Eow et al. [30] have shown that by increasing the electric field strength the deformation of a given droplet size increases. As shown in Fig. 9 and Table 2, by increasing the strength of electric field from 186 V/mm to 373 V/mm the deformation of the droplet with the diameter of $984 \pm 2 \mu\text{m}$ has increased and the side of droplet close to the interface becomes more elongated locally under higher electric fields. 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 electric fields.

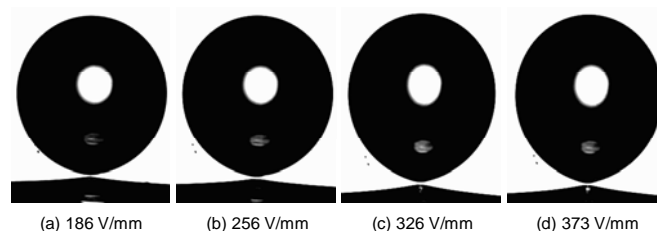


Figure 9. Different deformations degree for the same droplet diameter of $984 \pm 2 \mu\text{m}$ under different electric field strength.

Table 3. The deformation of the droplet with diameter of $984 \pm 2 \mu\text{m}$ under different electric field strength quantitatively.

Electric Field Strength (V/mm)	186	256	326	373
Deformation degree (d_{major}/d_{minor})	1.058	1.075	1.154	1.158

4-3-2. Addition of surface active agents

Adsorption of surfactants at water-oil interface is very important in terms of drop-drop and drop-interface coalescence time. So-called natural surfactants (e.g., asphaltenes, resins, inorganic particles, etc.) usually present in crude oil emulsion stabilise the water-in-oil emulsions [5, 8, 33] and increase time for separation. Another feature of surfactants that can influence the water in oil emulsion is related to the breakage and formation of the secondary droplets in the presence of surface active agents. Surfactants can significantly intensify the formation of the secondary droplets. In this work several surface active agents including Sodium Dodecyl Sulfate (SDS), Tween 20 (Polyoxyethylene sorbitan monolaurate) and Span 80 have been applied to investigate their effect on the secondary droplets formation. The effect of surfactants presence on secondary droplet formation has been shown in Fig. 10 (a) and (b).

Span 80 is highly soluble in oil phase whereas its solubility in the aqueous phase is very small. On the other hand, Tween 20 is

soluble in both aqueous and oil phases but its solubility in the aqueous phase is much higher than that in oil phase, and finally SDS is fully soluble in aqueous phase. The water soluble surfactants i.e. SDS and Tween 20 were added to aqueous phase as 1 g/l and under a specific electric field (232 V/mm); results of the presence of these surfactants have been compared with those for water phase without any surfactants under the same electric field. Also, the oil soluble surfactant was added to oil phase as 1 g/l as well but under a lower electric field strength i.e. 116 (V/mm) and the results have been compared with those for oil phase without any surface active agents under the same electric field strength. As it can be seen, addition of the surfactants to water or oil phase causes a significant effect on the formation of the secondary droplets. Under the same electric field strength of 232 (V/mm), by decreasing the interfacial tension from 25 (mN/m) for pure system to 6 (mN/m) for dispersed phase including 1 g/l Tween 20, the volume of the secondary droplets rapidly increases.

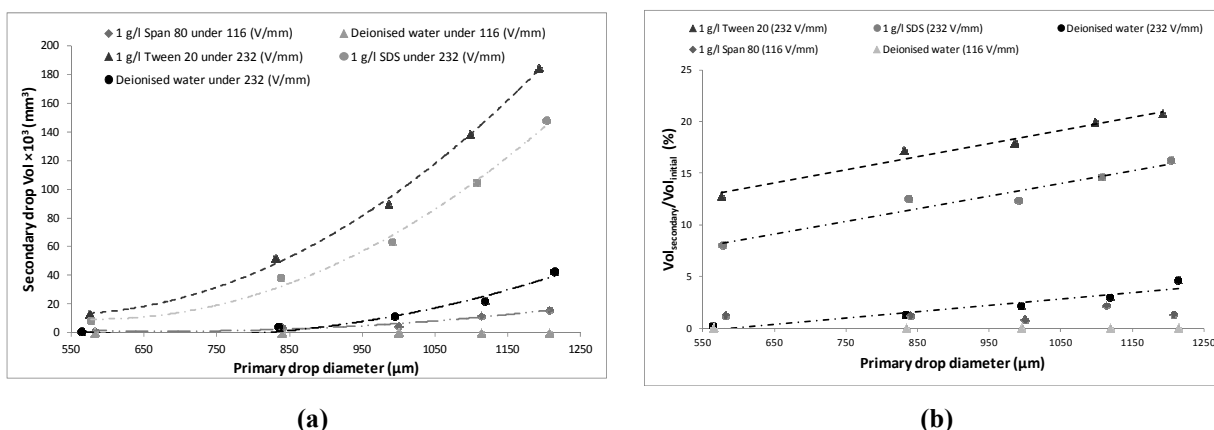


Figure 10. The effect of surfactants on the secondary droplets formation Secondary droplets volume as a function of primary droplet size Normalized secondary droplets volume as a function of primary droplet size.

Also, it is observed that for a pure system i.e. in the absence of any surfactant in aqueous and oil phase under the electric field strength of 116 (V/mm), no secondary droplets appear, but by adding just 1 g/l Span 80 into oil phase the secondary droplets start forming. These effects can be explained by the fact that surface active agents modify the interfacial tension as a result of their presence.

Unlike the pure system (Fig. 7 (b)), Fig. 10 (b) is showing addition of surfactants results in the primary drop size being effective on normalized volume; In fact, the presence of the surfactants in each phase can increase the probability of partial coalescence pattern's occurrence.

4-4. Detailed observation on partial coalescence

The temporal changes in some characteristic dimensions during coalescence of a single typical droplet are shown in Figs 11 and 12. In Fig. 12, “channel expansion” speed shows the radial change ($dr_{channel}(t)/dt$) of the narrow channel connecting the droplet and interface, and the terms “Equator reduction”

($dr_{equator}(t)/dt$) and “Peak falling” ($dh_{peak}(t)/dt$) speeds show reduction of equator and falling of peak point of droplet respectively.

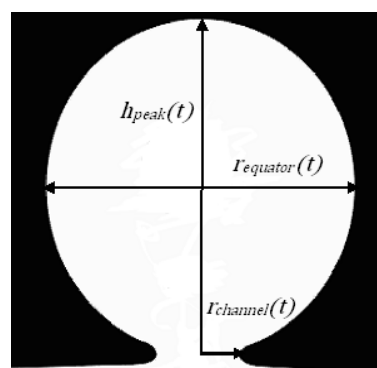


Figure 11. Characteristic dimensions to monitoring the speeds.

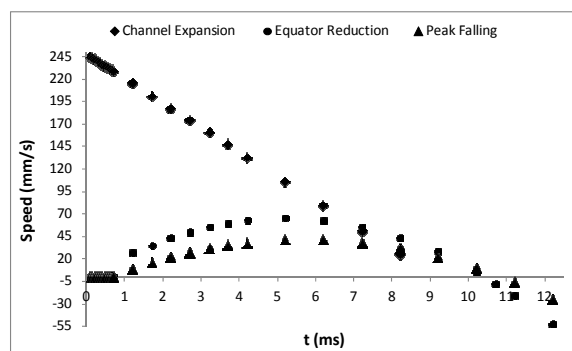


Figure 12. Speed of the channel expansion, equator deduction and peak falling for a drop with diameter 984 ± 2 μm under 373 V/mm.

To qualitatively understand the pumping and necking behavior at partial coalescence of a single droplet, these three speeds can be studied at the same time. At the first of coalescence phenomenon, channel expansion speed is rather high and its value is about 245 mm/s; this means after a few milliseconds of coalescence starting (after about 4 milliseconds), the channel diameter expands as the primary droplet diameter. In fact, this high speed of channel expansion points out a process called instantaneous rupture of the thin film between droplet and interface and then drainage of the droplet content into bulk phase. But this high speed of channel expansion exits just in the initial moment and reduces rapidly as after 6 ms of coalescence it reaches 78 mm/s showing about 68% reduction in expansion speed. This continuous and rapid reduction in channel speed implies that there is a considerable negative acceleration and according to Newton's second law of motion it represents a driving force causing channel expansion. Driving force to progress the expansion of channel is surface tension force trying to drive droplet content into bulk phase. Negative acceleration implies the driving force is reducing fast and in fact the maximum driving force exists at the start of the process. On the other hand, focusing attention on two other speeds i.e. equator reduction and peak falling shows there is no change in these two points till 0.7 msec and only after this time do they start to vary. This observation means in this time period there is mass flow into the droplet; the source for this mass flow can be supplied from bulk phase under the droplet. By considering the trend of equator reduction and peak falling speeds,

following a short time of stagnation till 0.7 ms, there is first an increasing and then decreasing trend till these two speeds reach zero value. There are two important points here: firstly, the value of these two speeds is considerably lower than expansion speed. The expansion speed is related to the pumping process but the peak falling can qualitatively describe the necking process; implying relative predominance and importance of the pumping process in comparison with necking process at the beginning of the partial coalescence process. After 10.2 ms of the start of the partial coalescence phenomenon both speeds become negative, meaning that the speeds are in inverse trend. From this point the necking process overcomes pumping process which results in detaching the secondary droplet from primary droplet coalescing with interface. In Fig. 13, the expansion speed has been shown for three different electric field strengths:

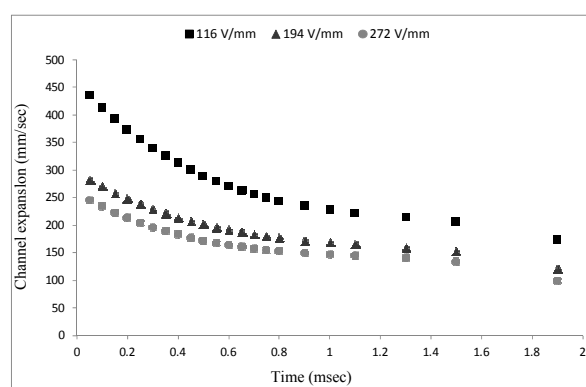


Figure 13. Channel expansion speed of a drop with diameter $984 \pm 2 \mu\text{m}$ for three different electric field strengths.

It can be seen by increasing the strength of the electric field the channel expansion speed reduces, this result is consistent with the results observed in Fig. 8, which shows by

increasing the electric field strength the volume of the secondary droplets increases as well; after contact between primary droplet and negatively charged interface whole droplet obtains negative charge; the more electric field strength the more attractive force on the negative droplet in the electric field; this attractive force decreases the mass flow from droplet to the homophase meaning the weakness of the pumping process in comparison with the necking process. In brief, by increasing the electric field the importance of the necking process increases and this causes a higher volume of secondary droplets to detach.

5. Conclusions

In the coalescence process of an aqueous droplet with a planar interface in the presence of an electric field two patterns of coalescence are observed: full coalescence and partial coalescence. In the latter case, the process itself produces secondary droplets: one main droplet and a number of much finer ones. The factors affecting the formation of secondary droplets are electric field strength and interfacial tension. Normalizing the total volume of the secondary droplets formed with respect to the initial drop volume, it is observed that the ratio of volumes is relatively independent of the primary drop size for pure system containing no surfactants for the electric field strengths in the range 116-256 V/mm; only at high field strengths some dependency on droplet size is observed. The volume of the secondary droplets clearly increases in the presence of water and oil soluble surfactants under electric field strength of 232 V/mm. The effect of the electric field stress is intensified

in the presence of surfactants due to lower interfacial tension. Two key processes are involved in the formation of the secondary droplets, necking and pumping. A mutual interaction between these two processes can determine total volume of secondary droplets.

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