

Experimental study of electrocoalescence of water drops in crude oil using near infrared camera

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Abstract:

Results are presented concerning the influence of an applied electric field on the coalescence of a water droplet with a much bigger water drop, both drops being immersed in a crude oil. This original study of electrocoalescence in crude oil was performed through high-speed optical observations, using a near-infrared (NIR) camera, of a droplet falling onto a bottom drop, a bipolar square voltage being applied. For low electrical field strength, the electrostatic pressure at the water/oil interface of the merging droplet partly counteracts the capillary forces, which slows down the drops coalescence process. Above threshold field strength, the electric forces drastically affect the dynamics of drops deformation and merging. At a working temperature $T = 60^{\circ}\text{C}$, partial coalescence was observed, leaving a daughter drop of size increasing with the applied field. At $T = 40^{\circ}\text{C}$, there was an abrupt transition from coalescence to non-coalescence, the top droplet inducing an upward directed nearly conical deformation of the bottom drop. This is attributed to charge exchange between the colliding water drops at each polarity reversal of the applied voltage, thus generating a bouncing-like behaviour at a frequency twice the voltage frequency. The charge exchange occurs through a very thin filament interconnecting the drops during a short time and presumably generated by interface instability. Under strong enough applied field, there was also formation of a very fine mist around the zone of drops quasi contact; this mist is ascribed to the breakup of the transitory very thin ligament bridging the drops. Some explanations and considerations are proposed for the various observations; in particular, it is guessed that the contrast between the transitions to partial coalescence (at 60°C) and to non-coalescence (at 40°C) arises from geometrical differences in the temporary small bridge interconnecting the drops.

1. Introduction

Crude oils coming out of the wells contain some water and the removal of water is a crucial step in oil production and processing for both technical and economical reasons. The process train of a typical oil production facility results in the fluid flow being subjected to several intentional pressure drops, e.g. by choke valves, and unintentional pressure drops by any other flow restrictions. The resulting strong shear forces break up water drops into smaller droplets thus creating a very fine water-in-oil emulsion [1] (droplets diameter down to a few micrometres). The water is usually separated from the oil in large gravity separators where the water drops settle due to their higher density. But the sedimentation speed w_d (determined by balancing the drag force, given by Stokes law, with the buoyancy force) is proportional to D^2 (D : water drop diameter) and takes extremely small values for the small droplets. An effective separation requires to dramatically increase w_d ; this might be achieved using centrifugal separators [2] or decreasing the oil viscosity by heating the emulsions, means that are not attractive from the economical viewpoint.

The most efficient way of increasing the sedimentation speed is to increase the drop size through merging of the droplets. This can be performed by using additives which destabilize the water-in-oil emulsion [3] but, again, such a technique is not attractive from the economical and environmental viewpoints. The coalescence of droplets into larger drops can be promoted by applying an electrical field to the crude oil emulsion [4]. This technique, called electrocoalescence, can be applied in existing process equipment to improve separation performance and increase process flow. The main effect of the electrical field is to induce an attractive force between drops in close vicinity that tends to align them with the electrical field lines and to promote drops merging. In compact electrocoalescers, the combination of turbulence and electric field enhances the droplets coalescence. Schematically, turbulence leads to a high collision rate of water drops in the flow and the electric field then brings about the drops merging [5].

The field induced merging of two drops can be considered a three-stage process : *i*) the two drops becoming closely spaced; *ii*) the drainage of the oil film separating the close drops; *iii*) the merging of the two drops. The first stage, that can be named drops collision, is mainly determined by the properties of the emulsion flow. The second stage, that of drainage of the oil film between the close drops, possibly up to contact [6], is strongly influenced by the electrostatic force between water drops [7] and also depends on the droplets sizes and on the interfacial tension. The influence of the electrostatic forces on drop movement in a dielectric liquid is generally well understood as long as the drops are not too close [8].

The influence of the electrical field on the final coalescence stage is, however, not so well understood, particularly for water drops in crude oils. Ordinary coalescence between two drops, i.e. without any electric field imposed, is triggered when the thin film separating the drops has drained so much that the distance between the drop surfaces becomes small enough for van der Waals forces between the drop surfaces to become significant, pulling the surfaces together to form a water bridge [9,10]. However, for coalescence between two drops with an imposed electric field, the electric charge distributions on the drops interfaces and the subsequent electric forces play the major role. The attraction force between the drops can become very large as they approach one another [5,11,12]; moreover, the electrostatic pressure at the interfaces can significantly affect the drops shape and, even, trigger surface instability [13,14]. It has been guessed that such a surface instability generates a water bridge between the drops that finally results in the drops merging [8]. The observed phenomena, however, are not so simple. Under strong enough applied field strength, for a drop falling on a flat interface, coalescence of only a part of the drop volume (partial coalescence) can occur [15-17]; also, colliding water drops immersed in pure dielectric liquid can be non coalescing and bouncing [17-20]. Keeping in mind that a strong electric field can also disintegrate big enough water drops [21], it is clear that the choice of the electric field strength to be

applied in order to promote electrocoalescence of a water-in-oil emulsion is not straightforward and will depend on many parameters like the initial distribution of droplets diameters, the interfacial tension and also the permittivity, conductivity and viscosity of the oil [22].

In the practical case of emulsions of water in crude oil, the problem is even more complex because crude oils contain numerous surface active compounds (surfactants, resins, asphaltenes, etc.) that can build films at the water-oil interfaces. The chemical, mechanical and electrical properties of such films at the water-oil interface might influence the drops deformation, the interface instability and disruption and, hence, the electrocoalescence process. Therefore it is of primary importance to examine the influence of the electric field on the coalescence of water drops in crude oil. As the basic mechanism of electrocoalescence in water-in-oil emulsions concerns what occurs between pairs of close drops, it is reasonable to investigate the problem for two water drops only.

Up to now, studies of the basic electrocoalescence process were performed in models oils through optical observation, using a high speed video camera, of a small water drop falling on a horizontal water/oil interface or on the top of a bigger water drop, the system being subjected to a vertical field [23,24]. The opacity of most crude oils to visible light makes such studies impossible with the usual optical equipments. Nevertheless, crude oils are somewhat transparent to near infrared light so that the experimental study can be performed using a high speed, near infrared (NIR), video camera. Before describing the used experimental equipment and presenting the results of the investigation, we give a picture of the forces and mechanisms involved in the interaction, under electric field, of two drops of very different size immersed in a dielectric liquid.

2. General considerations

In the simplest case of very pure dielectric liquids (without surfactants), the interfacial tension is constant and a qualitative picture of the coalescence process can be given by taking into account the electrical, capillary and viscous forces. Let us first recall that when a drop of liquid #1 immersed in an immiscible liquid #2 of lower density gently falls on a flat horizontal interface separating the two liquids, the drop stays just above the interface during a rest time, often on the order of seconds, before coalescing with the underlying liquid #1 [26]. In the case considered here of a small water drop falling in a lighter dielectric liquid above a much bigger and immobile water drop – as shown in Fig. 2 – the falling velocity w_d of the drop decreases when it approaches the big drop because of the increased viscous effects in the oil zone squeezed between them and there should be again a rest time. But in the absence of electric field, the situation of the small water drop located just above the top of the big drop is unstable : due to gravity effects, after a time lower than the rest time on a flat interface, the small drop escapes from the apex by rolling over the big drop surface.

The dielectric liquid is assumed to be a very good insulating medium and the water a very good conductor. When subjected to an electric field, water drops are equipotential (zero field inside) and have, at the water/oil interface, distributions of surface charge $\sigma_s = \varepsilon E$ and of electrostatic pressure $p_{es} = \frac{1}{2} \sigma_s E = \frac{1}{2} \varepsilon E^2$ (ε : dielectric liquid permittivity, E : electric field intensity). In the case of a single drop in a uniform field, the σ_s distribution is anti-symmetrical (but the p_{es} distribution is symmetrical) and the drop is equivalent to a pure dipole and the resultant electric force on the drop is zero (this is still valid when taking into account the drop deformation due to the electrostatic pressure). Two far enough drops in a uniform electric field experience a force between each other that can be determined through the interaction of the two equivalent dipoles [5,8,11]. When the two drops spacing is lower than about the diameter of the smallest drop, analytical expressions give the value of interaction forces and the value of enhanced electric field at the facing poles for spheres [8,11]. When the applied electric field is high enough, the interaction force between the two drops aligns the drops centres with the electrical field which results in the falling drop going to the top of the bottom big drop and staying there [23]. With reference to the problem

considered in this paper, Fig. 1-*a* illustrates the fact that the falling water drop has a positive (negative) surface charge density on its bottom part higher than the negative (positive) charge density at its top part (although its net charge is zero); hence the electrostatic pressure is clearly higher at the bottom part of drop surface than at the top part and the resulting force, directed downwards, tends to expel the liquid lying between the drops. The attraction force increases as the drops spacing decreases and the drops get into contact after a finite time [5,27] similarly to what was observed for drops falling on a flat water/oil interface [25]. The rest time of the small drop at the apex of the bottom big drop is a strongly decreasing function of the applied electric field when working with model oils [23].

After a water bridge begins to connect the two drops, coalescence is typically expected to proceed, i.e. the small drop is expected to merge with the big one because the surface energy is thus minimized. However, this simple energy argument ignores the dynamical effects and, practically, in the absence of electric field, total coalescence is not observed in all cases. By studying many couples of liquids in the drop/flat interface system, Charles and Mason observed the occurrence of partial coalescence of drops for the ratio of drop and surrounding liquid viscosities ranging from 0.02 to about 10 [26]. The mechanism responsible for partial coalescence is the following: capillary waves induced by the initial fast changes of the bridge shape converge at the top of the drop that empties and this raises the upper part of the emptying drop that then takes a columnar shape; this transitory column experiences a pinch-off that leads to the formation of a daughter droplet [28]. This partial coalescence process implies vigorous capillary waves; with liquids of high enough viscosity, the viscous dissipation damps the capillary waves and inhibits the partial coalescence.

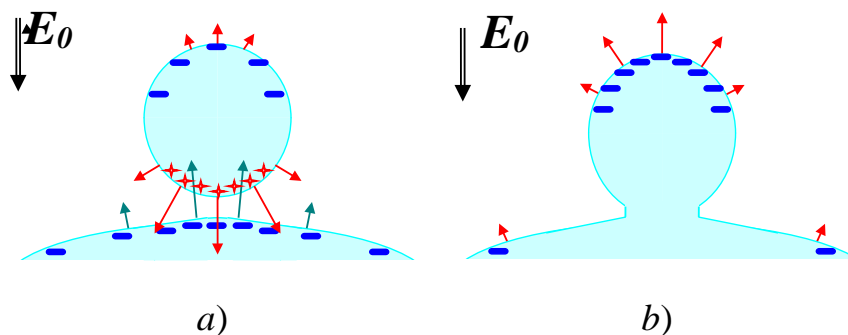


Figure 1. Schematic depiction of the surface charge density and electrostatic pressure at the drops interfaces induced by the applied field E_0 . *a*) water droplet close above a water drop; *b*) shortly after bridging.

The transition from partial to total coalescence depends on the values of the Ohnesorge numbers Oh_1 and Oh_2 characterizing the two fluids; for the drop/flat interface system, partial coalescence occurs when $Oh_1 < Oh_1^* \leq 0.02$ and $Oh_2 < Oh_2^* \leq 0.3$, the transition values Oh_1^* and Oh_2^* being interrelated [28-30]. For two drops of different radii, partial coalescence can also occur provided the radii ratio is smaller than 0.5 [31] (in this case the transition values decrease with the radii ratio). In the context of electrocoalescence implying oils of high enough viscosity, without applied field total coalescence is expected.

Let us consider the case of total coalescence for a small water drop falling on a big one in the absence of electric field. In order to examine what is the influence of an applied DC electric field on the coalescence process, it is important to point out that the applied field E_0 induces a finite potential difference between the two drops related with the attraction force (Fig. 1-a). Once the oil film between the drops has been drained and punctured, the water bridge linking the drops very rapidly causes a charge exchange which brings the two drops to the same electric potential. The modified distributions of surface charge density and electrostatic pressure (Fig. 1-b) induce a resultant electric force F_e which tends to draw upwards the falling drop. The order of magnitude of F_e is : $\pi a^2 \varepsilon (E_{max})^2 / 2$ where a is an effective radius ($a < R$, R being the radius of falling droplet) and E_{max} is the maximum field at the interface apex. The upper part of the droplet can also be considered to be subjected to a capillary force F_{cap} whose order of magnitude is : $2\pi b\gamma$ (γ : interfacial tension, b : minimum radius of the neck joining the drops) and its acceleration is determined by the force difference $F_e - F_{cap}$. For low applied field strength E_0 , the electrical force only partly counteracts the capillary force which tends to smooth out the interface shape; consequently, a slower and slower drops merging is expected as E_0 is increased.

For an applied field such that the electrical force exerted on the top of the connected drop and the resultant capillary force are of same order, the upper part of the droplet tends to remain at the same level and not to flow downward into the big drop. The process is then more or less similar to the partial coalescence occurring with fluids of low viscosity in the absence of electric field : a part of the water of the former falling drop flows into the lower drop, then the remaining part takes a transitory columnar shape that pinches off, leaving a "daughter" drop [16,17]. There is a threshold field for partial coalescence and increasing the field strength results in a daughter drop of increasing radius [16,32,17].

Now this electrically induced partial coalescence is not the only process generated by the applied electric field subsequently to the drops contact and bridging. Another behaviour, non-coalescence – or equivalently bouncing – has been observed for pairs of drops of similar size a long time ago [18] and more recently in microfluidic devices [19,33]. An extensive study with various liquids showed that bouncing is a general phenomenon occurring above a field threshold for pairs of unequal drops and is often associated with marked deformations of the drops [20,17]. However, the necessary conditions for bouncing relating the field strength with drop size, interfacial tension, oil viscosity, etc., have not yet been totally clarified. It has been proposed that for two drops of same size, the non-coalescence behaviour is promoted by cone-like deformations of the approaching parts of the drops with a cone half angle lower than about 60° [34]. But this is only part of the story; a qualitative explanation states that a jet forms at the tip of one of the approaching cones resulting in a liquid bridge between the drops; if this bridge is long enough it breaks up and the exchange of charge between the drops makes them to repel each other [19]. It has been shown experimentally that a thin temporary bridge is created which provides the charge exchange [20,33,34]. Moreover, it has been established unambiguously that the temporary bridge also promotes a vanishingly small mass exchange between the two drops [19]. Finally the recent study by Hamlin et al. [17] showed that the conductivity of the drops plays an important role on the two phenomena of partial coalescence and bouncing and that there exists a first order transition from partial coalescence to bouncing as the applied DC electric field is increased.

To summarize, the application of a strong enough electric field drastically modifies the process of total coalescence between two drops (or a drop and a flat interface) occurring without field and two scenarios have been observed : partial coalescence and bouncing. Several parameters and fluid properties: field strength, interfacial tension, viscosities and conductivities, presumably are involved; but the present knowledge on electrical partial coalescence and non coalescence in pure dielectric liquids is limited and does not allow delineating the domains of occurrence of these

two processes. Last, but not least, applying an AC voltage (or BSV : bipolar square voltage) results in extra phenomena that can alter some of the behaviours expected when using DC fields.

3. Experimental set-up and procedures

The aim of the experiments was to investigate the electrocoalescence of water drops in a crude oil. The experimental arrangement which has been chosen is basically similar to the one used in a previous study of electrocoalescence in a simple oil [23]. It consists of a water droplet falling on a much bigger water drop, the study being carried out through optical observation of the drops collision and merging.

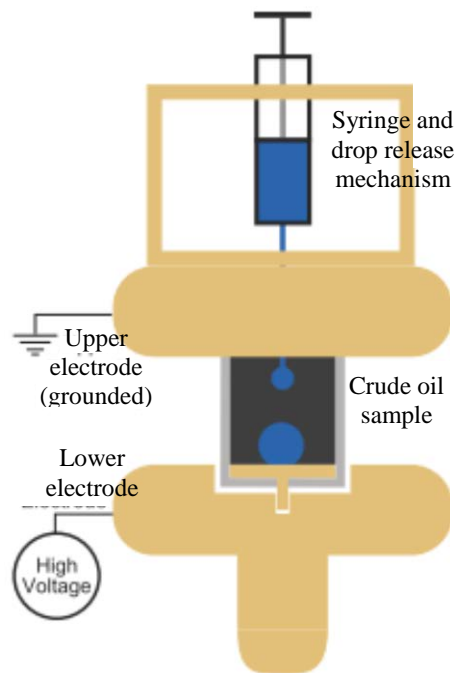


Figure 2. Experimental setup showing a stationary drop on the lower electrode and a droplet still attached to the needle tip.

A. Experimental setup and visualization

A schematic view of the test cell is shown in Fig. 2. In order to reduce the amount of crude oil used in the experiments, the crude oil sample was contained in a small glass cuvette (20 mm × 20 mm and height 15 mm or 20 mm) sandwiched between two horizontal electrodes. A bottom water drop ($\varnothing \cong 4$ mm) was lying on the lower electrode and anchored on it by an indentation in the metal surface. The falling droplet was formed at the tip of a glass needle by a syringe with a screw plunger. The cuvette was placed in a test cell made of Macor, a ceramic material that is electrically and thermally insulating; this cell having quartz windows for optical observations was filled with a circulating dielectric liquid (MIDEL 7131) to control the operating temperature.

Crude oils are opaque for visible light but nearly transparent in the near-infrared range (between 1000 and 2300 nm [35]). The cameras used for recording the coalescence process were, initially, a Phoenix NIR camera with a DTS backend manufactured by FLIR Systems and, later, a Xenics Cheetah CL camera. The first had an InGaAs image sensor with a resolution of 256 by 320

pixels, 14 bit dynamic range and sensitivity to light with wavelength from 900 nm to 1700 nm. The maximum recording rate was 345 frames per second for a full frame (higher frame rates can be achieved at lower resolutions). The second camera has the same sensor type but with a resolution of 640 by 512 pixels and a maximum rate of 1730 frames per second at full resolution. The light source used in the experiments was a quartz-halogen lamp with peak emissive power at 800 nm and a slowly falling tail into the IR-region. Appropriate long-range microscope lenses were used to obtain the adequate image magnification. Water absorbs light in the near infrared range; but the major phenomenon leading to clear pictures of water drops in crude oil is the geometrical effect of the light being bent away from the optical axis, preventing most of the light hitting the drop from reaching the camera.

B. Fluid properties

The crude oil used in the current experiments is from the Grane field in the North Sea. The water drops were made from MilliQ- filtered water with 3.5 wt% NaCl added. The main properties of the two fluids are given in Table I. The interfacial tension γ depends on the accumulation of surfactants on the drops surface; therefore, it is a decreasing function of time as can be seen on Fig. 3 giving the γ value versus time for the same Grane crude oil and brine [36]. The decrease of interfacial tension from its initial value (at $t = 0$) down to a saturation value is of limited extent ($\leq 20\%$ as inferred from Fig. 3). As it is well known that the saturation value of γ has a very weak dependence on temperature [36], we concluded that the γ value characterizing the water droplets in crude oil was nearly the same for the two working temperatures : $\gamma \cong 25$ mN/m. The dielectric constant ϵ_r of the crude oil was about 2.5 and its conductivity was $\sigma_{oil} \cong 2 \cdot 10^{-8}$ S/m at $T = 40^\circ\text{C}$ and $\sigma_{oil} \cong 10^{-7}$ S/m at $T = 60^\circ\text{C}$. The charge relaxation time constant $\tau_{oil} = \epsilon_{oil}/\sigma_{oil}$ of the crude oil is thus $\tau_{oil} \cong 1$ ms at 40°C and $\tau_{oil} \cong 200$ μs at 60°C . The conductivity of the salted water was about 5 S/m and its permittivity $\epsilon_{water} \cong 7 \cdot 10^{-10}$ F/m. The Bond number $Bo = (\rho_{water} - \rho_{oil})gR^2/\gamma$ (g : gravitational acceleration) gives an indication of the relative influence of gravity and interfacial tension. For the current system, a water drop with a diameter of 0.7 mm has a Bond number $Bo \cong 0.02$, which indicates that, to a very good approximation, the falling drop is spherical.

Table I Some fluids properties at three temperatures.

Temperature T ($^\circ\text{C}$)	20	40	60
Crude oil density ρ_{oil} (kg/m^3)	924	912	899
Water density ρ_{water} (kg/m^3)	1024	1017	1007
Crude oil viscosity μ_{oil} (mPa/s)	208	62.5	25
Water viscosity μ_{water} (mPa/s)	1.07	0.672	0.446
Interfacial tension γ (mN/m) (after 10 min)	$\cong 25$	$\cong 25$	$\cong 25$
Crude oil conductivity σ_{oil} (S/m)		$2 \cdot 10^{-8}$	10^{-7}
Water conductivity σ_{water} (S/m)		5	7

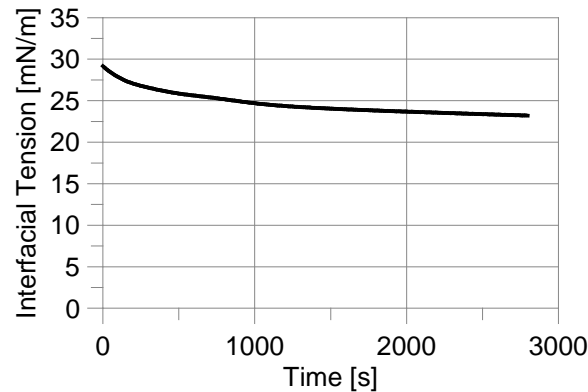


Figure 3. Measured values of the interfacial tension between Grane crude oil and brine (water with 3.5 wt% NaCl) at 60°C (reconstructed from [36]).

C. Experimental procedures

The bottle with crude oil was heated in a water bath and shaken vigorously by hand before a sample of crude oil was extracted to fill the glass cuvette. The stationary water drop was put on the lower electrode just before the crude oil was added to the cuvette. The stationary drop was not replaced during the experiments, thus the aging time of the stationary drop was long, ranging from a few hours up to a few days. The falling drop was formed at the tip of a glass needle using a syringe with a screw plunger. The glass needle was made in-house by heating and drawing glass tubing in a special apparatus. The outer surface of the tip of the glass needle was made hydrophilic by silanization with SL2 Sigmacote from Sigma-Aldrich. After silanization, the tip of the glass needle was cut off, giving a small area of non-hydrophilic glass at the very tip of the needle. The drop of typical diameter $\varnothing \cong 700 \mu\text{m}$ was kept attached to the glass needle for 10 minutes to allow accumulation of surfactants on the oil/water interface. The drop was then released by rapidly moving the glass needle and syringe upwards using an electromagnetic actuator.

For calibration of the images captured by the camera a tungsten wire of diameter 300 μm was inserted into the oil-filled glass tank. The wire was brought into focus and a movie was then recorded. The focus of the camera lens was not changed after this (the camera was moved back and forth to bring the experimental plane into focus). A large lens aperture was used during the experiments to keep the depth of field as narrow as possible and maximize the amount of light admitted to the image sensor.

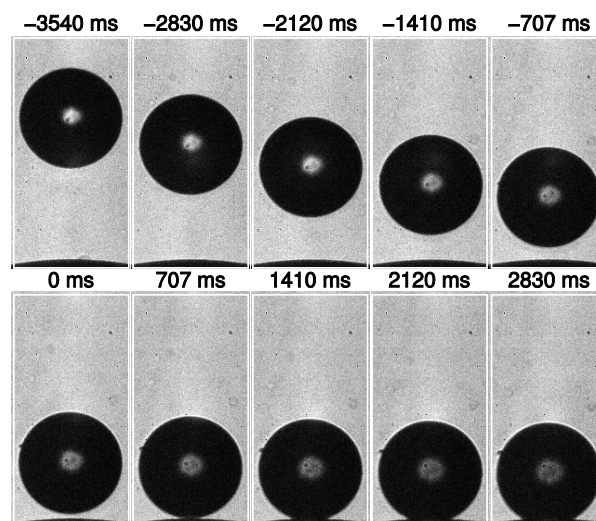
Experiments were performed using a bipolar square voltage (BSV) with a frequency of 1000 Hz given by a signal generator and amplified by a TREK 20/20B-H high voltage amplifier. The amplifier delivers a maximum voltage of 20 kV and a maximum current of 20 mA, with a bandwidth of 20 kHz (the voltage applied to the lower electrode was measured with a Tektronix high voltage probe connected to an oscilloscope). Using a BSV waveform gives a phase-independent electrostatic force on non-charged drops; this is equivalent to applying a DC field, but with the advantage of a capacitive voltage distribution in the electrode gap. The high voltage was applied to the lower electrode after the drop was released, to ensure that the drop was not charged while hanging at the glass needle tip. Experiments were conducted at two temperatures, 40 °C and 60 °C. At higher temperatures than 60 °C, thermal convection in the crude oil made it difficult to make the small falling drop hitting the stationary drop at its apex.

4. Results and analysis

A. Drop falling in crude oil ($E = 0$)

Several experiments without applying an electrical field were performed as a reference case. After release, the falling water droplet rapidly reached terminal velocity w_{term} but started to slow down when the drops separation became lower than about one droplet diameter (see Fig. 4). The slowing is due to the enhanced viscous dissipation associated with the flow of the oil between the drops. Systematic measurement of the terminal velocity w_{term} far enough above the stationary drop was performed varying the drop diameter. The measured values are presented in Fig. 5 in terms of the non-dimensional velocity parameter $w^* = w_{term}/w_{ref}$, w_{ref} being the theoretical velocity based on the Stokes formula for the friction force of rigid spheres $F_v = 6\pi R \mu_{oil} w_{term}$. For falling drops with totally immobile interface we should have $w^* = (9/2) \mu_{oil} w_{term} / (R^2 g \Delta\rho) = 1$ whereas for drops with fully mobile interface the Hadamard-Rybczynski expression $F_v = 6\pi R \mu_{oil} (2\mu_{oil} + 3\mu_{water}) / (3\mu_{oil} + 3\mu_{water}) w_{term}$ leads to $w^* = (3\mu_{oil} + 3\mu_{water}) / (2\mu_{oil} + 3\mu_{water}) \cong 1.49$ in the present conditions ($w^* = 1.492$ at $T = 40^\circ\text{C}$ and $w^* = 1.487$ at $T = 60^\circ\text{C}$). Fig. 5 indicates that the viscous force on the drop is close to the value given by the Stokes formula characteristic of a solid body. This suggests that the falling drops have an immobile interface.

It is clear that we are not dealing with fluid spheres having fully mobile interface ; such a behaviour is obtained only when using refined techniques for highly purifying the liquids [37]. In the studied crude oil, there are many surface active compounds, including asphaltenes, which can make the interface partially or totally immobile. In his review paper Loth [38] recalls that the contaminants influence depends on their concentrations, on their properties and on the composition of the two phases (water is particularly susceptible to the influence of surfactants). He states that fully mobile interface of a falling drop requires vanishingly small contaminant concentration c ($c < 10^{-7} \text{ g/l}$) whereas with $c > 10^{-2} \text{ g/l}$, the interface is totally immobile [38]. In the Grane crude oil, the concentrations of asphaltenes and resins are about 30 g/l and 300 g/l respectively [39]. These high concentrations make sure that the water/oil interfaces are immobile.



a)

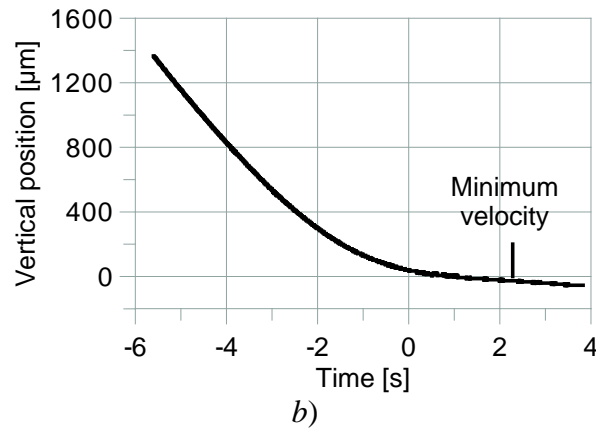


Figure 4. Water drop (diameter $D = 740 \mu\text{m}$) falling in Grane crude oil above a stationary bigger drop in the absence of electric field ($E = 0$) at temperature $T = 40^\circ\text{C}$. *a*) selected images from a video recording; *b*) vertical position of falling drop.

There was no detectable deformation of the falling drop. The falling velocity was reduced to almost zero when the falling drop approached very closely the bottom drop. Without the focusing effect of the electrical force it was quite difficult to make the falling drop hit the apex of the stationary drop and only a small percentage of falling drops were colliding the big drop at its apex. In the case of collision at the apex of the stationary drop, the small drop stayed immobile for some time and then rolled off on the lower big drop. No coalescence was observed in the absence of electric field.

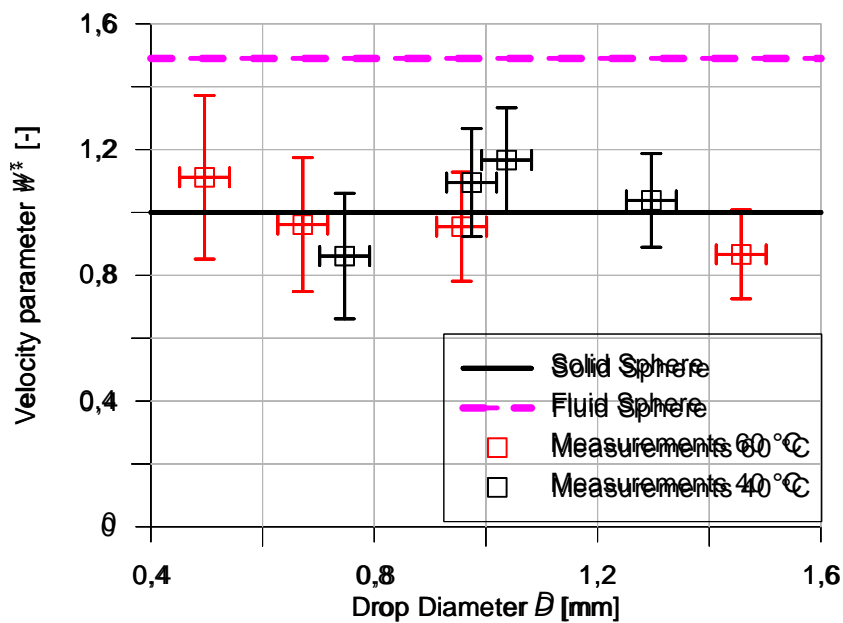


Figure 5. Velocity parameter w^* for water drops falling in Grane crude oil at two different temperatures.

B. Field influence on the motion of falling drop

The voltage used is bipolar square and passes from $+V$ to $-V$ or from $-V$ to $+V$ at each half period. In each drop (without contact) the reversal of the surface charge distribution at the interface requires a time on the order of the relaxation time of the salted water: $\tau_{water} = \epsilon_{water}/\sigma_{water} \sim 10^{-10}$ s which is totally negligible. Therefore the electrostatic pressure, quadratic in E , can be considered constant and equal to the one provided by a DC voltage.

When applying an electric field, it was easier to obtain head-on collisions between the two drops, because the field induced force between the drops tends to align their centres with the field. Indeed, denoting by θ the angle between the line of drop centres and the vertical axis (direction of applied field), following Davis [11] the components of the electric force F_e for an uncharged drop is :

$$(F_e)_r = 4\pi\epsilon_{oil} R^2 E_0^2 (F_1 \cos^2\theta + F_2 \sin^2\theta) \quad (1)$$

$$(F_e)_\theta = 4\pi\epsilon_{oil} R^2 E_0^2 F_8 \sin 2\theta \quad (2)$$

where the coefficients F_1 , F_2 and F_8 have complex expressions determined by an exact resolution of the problem [11] and depend on the two geometrical parameters s/R and R/R_b , s being the spacing between the drops and R_b the radius of the bottom drop (F_1 diverges when $s \rightarrow 0$). Determining the trajectory of a slightly off-axis falling droplet is outside the scope of this paper. Let us just remark that from the few numerical values given in [11], for $s/R < 1$ $F_8 \sim -0.5$ in our case ($R_b/R \cong 7$); then the component $(F_e)_\theta \sim -4\pi\epsilon_{oil} R^2 E_0^2 \theta$ draws the falling drop towards the vertical axis $\theta = 0$ and for a strong enough field the point of impact is presumably very close to the bottom drop apex characterised by $\theta = 0$.

During drop falling, the electrical force added to the buoyancy force but its influence was noticeable only when the spacing s between drops was lower than the falling drop diameter D . The electrical force then increased when the drops separation decreased, behaviour analogous to the one of viscous dissipation due to film-thinning. For low applied field, the electric force partly counteracted the effect of film thinning dissipation and the droplet deceleration was less marked (Fig. 6). For strong enough applied field, the electrical force played the major role and the falling drop accelerated when approaching the stationary drop (Fig. 6). For the studied system, the constant falling velocity observed under the nominal electric field $E_0 = 750$ V/cm for the two temperatures (see Fig. 6) strongly suggests that the electrostatic attraction force balanced the film thinning force in these conditions. Such a nearly constant velocity of the falling drop until impact was also observed in a model oil for a similar applied field strength [23].

These observations can be grossly accounted for. Firstly, for two conducting drops of radii R and R_b at a small spacing s and subjected to a potential difference ΔV , the attraction force takes the asymptotic expression in the case $s/R \ll 1$ [5] :

$$F_e \cong \frac{\pi \epsilon_{oil} (\Delta V)^2}{1 + R/R_b} \frac{R}{s} \quad (3)$$

Secondly, for spheres with immobile interface, the lubrication force F_{lu} due to the squeezing of the oil between the drops has the following asymptotic expression, again for $s/R \ll 1$ [6] :

$$F_{lu} \cong \frac{6\pi \mu_{oil} R w}{(1 + R/R_b)^2} \frac{R}{s} \quad (4)$$

where w is the velocity of the falling drop. The striking point here is that, if the potential difference ΔV between the drops is constant, the forces F_e and F_{lu} have the same variation law as a function of the spacing s . Now for two close drops of same radius R aligned with the field E_0 , ΔV scales with

the product $2R^*E_0$ and has a weak logarithmic dependence on R/s [13]; hence ΔV here can be assumed constant to a first approximation. Further rough approximations allow to estimate the velocity of the falling drop : *i*) the viscous effect on the falling drop is assumed to be the sum of Stokes and lubrication forces; *ii*) similarly for the motive force we add the gravitational F_g and the electric F_e forces. The inertial effects being negligible, we have $F_v + F_{lu} = F_g + F_e$ which leads to :

$$w \cong \frac{2 R^2 g \Delta\rho}{9 \mu_{oil}} \frac{\left(1 + \frac{G}{(1+R/R_b)^2} \frac{R}{s}\right)}{\left(1 + \frac{1}{(1+R/R_b)^2} \frac{R}{s}\right)} \quad \text{with} \quad G = \frac{3 \varepsilon_{oil} (\Delta V)^2 (1+R/R_b)}{4 R^3 g \Delta\rho} \quad (5)$$

The parameter G is proportional to the ratio of an electrostatic pressure and the pressure arising from a water column of height R in the oil. As the drop falls, the spacing s decreases down to zero; the falling velocity w decreases monotonically when $G < 1$ and increases monotonically when $G > 1$. The case $G = 1$ corresponds to the constant terminal falling velocity w_{term} .

This rough analysis qualitatively accounts for the results shown in Fig. 6, in particular for the constant velocities obtained with a nominal field of 750 V/cm. Let us remark that these constant velocities imply an approximately constant value of ΔV in the last zone of the drop fall ($s < R$) that, moreover, is independent of the temperature and, therefore of the oil viscosity. From $G = 1$ we deduce $\Delta V \cong 50$ V in the present conditions; as ΔV here scales with the product R^*E , this gives $E \sim 1.4$ kV/cm for the typical value of the field above the bottom drop; this value appears to be quite consistent with the nominal field $E_0 = 0.75$ kV/cm in consideration of the curvature of the bottom drop protruding over the metallic electrode (see Fig. 2).

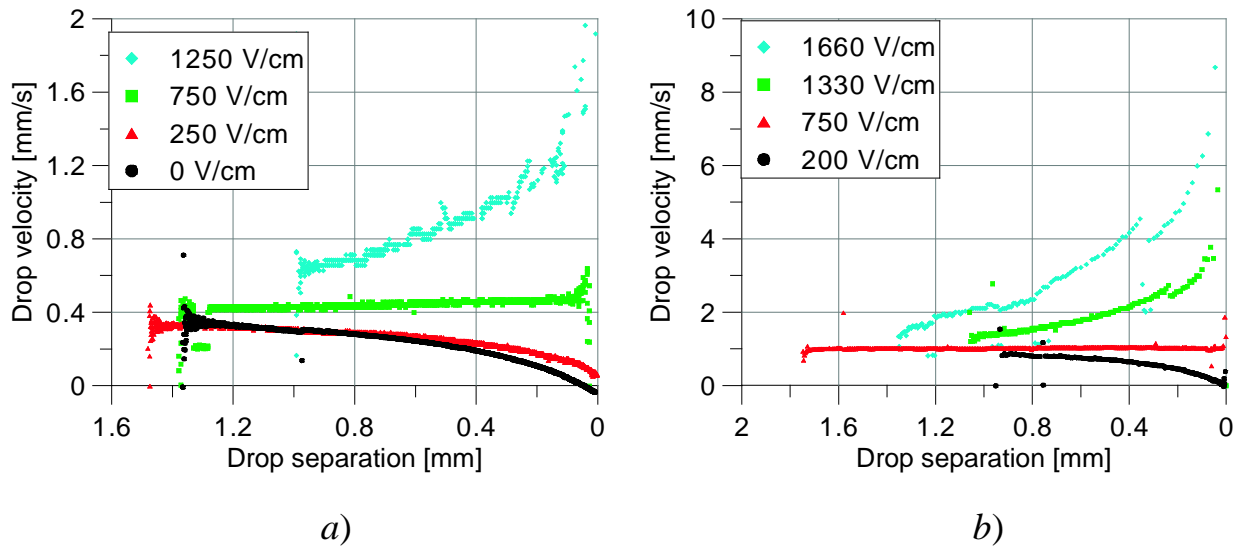
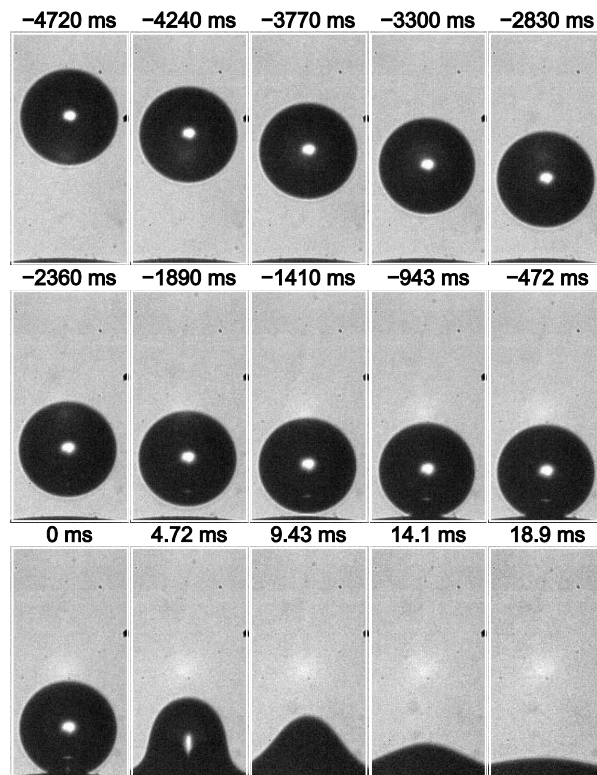


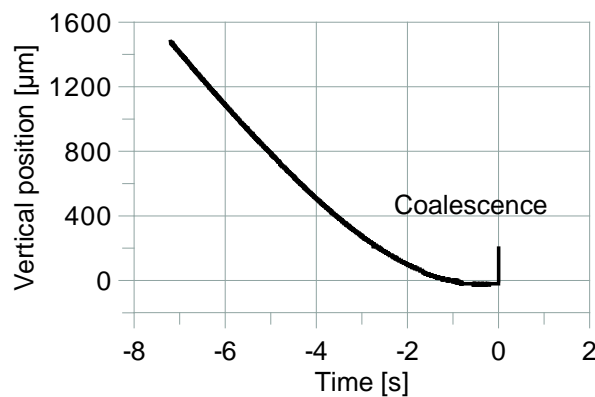
Figure 6. Velocity of water drops of diameter $D \cong 0.7$ mm falling in Grane crude oil; the velocity was derived from the drop position as a function of time determined from the recorded movies. *a*) $T = 40^\circ\text{C}$ and *b*) $T = 60^\circ\text{C}$.

C. Field influence on coalescence

Under small enough nominal field strength ($E_0 < \sim 1000$ V/cm), there was no significant deformation of the falling drops and of the upper part of the stationary drop (Fig. 7). After impact, the droplet remained resting on the top of the lower drop for a while before coalescing (it is well known that the rest time strongly decreases with the field strength [23,25]). As shown in Fig. 7, the coalescence was total as it would have been presumably in the absence of applied electric field if it would have been possible to observe it. Such a total coalescence was expected because the Ohnesorge number of the oil phase takes values $Oh_2 \cong 0.7$ at $T = 40^\circ\text{C}$ and $Oh_2 \cong 0.28$ at $T = 60^\circ\text{C}$ and the Ohnesorge number of water was $Oh_1 \cong 0.01$ so that the corresponding points in the Oh_2 vs Oh_1 plot are in the zone of total coalescence for a planar interface (see Fig. 5 in [30]).



a)



b)

Figure 7. Drop coalescence in Grane crude oil : $T = 40^\circ\text{C}$, $D = 710$ μm , $E_0 = 250$ V/cm; a) selected images from the recorded movie; b) vertical position of falling drop.

After the exchange of charge between the connected drops, the whole interface was equipotential and the new surface charge distribution at the water/oil interface changed the local electrostatic pressure and gave rise to an upward resultant force F_e for the upper part of the droplet. The movies taken with the NIR camera give interesting information on merging dynamics. Fig. 8 illustrates the field induced slowing down of the merging process. Clearly the electric force F_e on the upper part of the coalescing droplet partly counteracted the effect of capillary force so that the lowering of this upper part was slower than without field. The pictures of Fig. 8 moreover reveal that the shape of the retracting interface is more acute for $E_0 = 750$ V/cm than for $E_0 = 250$ V/cm; this is due to the electrostatic pressure $\epsilon E^2/2$ acting mainly on the interface zone near the apex where the electric field is strongest. The slowing down of coalescence was observed to be quite similar at $T = 60^\circ\text{C}$, the only difference being a lower coalescence time which can be ascribed to the lower viscosity of the crude oil ($\mu_{oil} = 25$ mPa.s instead of 62.5 mPa.s at $T = 40^\circ\text{C}$).

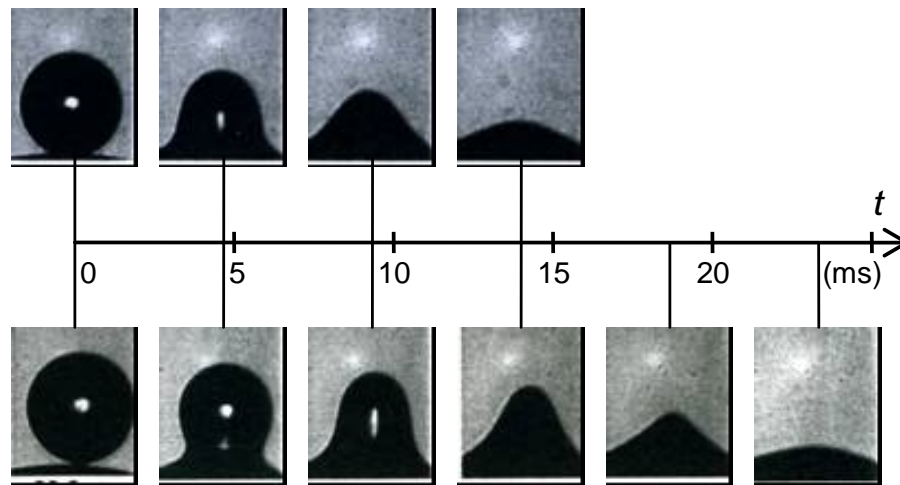


Figure 8. Images from 2 movies of drop coalescence in Grane crude oil at 40°C taken at the same instants after bridging. Top images : $E_0 = 250$ V/cm, falling droplet diameter $D = 710$ μm . Bottom images : $E_0 = 750$ V/cm, $D = 750$ μm

Fig. 9 shows the variations of the apex level of the falling – and then coalescing – drop as a function of time. Just after the drops connected, the apex level slowly decreased due to the difference $F_{cap} - F_e$ between capillary and electric forces. A constant $F_{cap} - F_e$ force applying to a constant volume of water would lead to a parabolic shape of the time variation of the apex height; this qualitatively accounts for the shape of the curves in Fig. 9 during the first milliseconds. The quadratic dependence of electrostatic pressure on E_0 results in a very small influence for the low applied field value $E_0 = 250$ V/cm so that, to a first approximation, the corresponding curves in Fig. 9 can be taken as references for the coalescence process which would exist without field influence (as afore mentioned, coalescence without applied field could not be observed). Fig. 9 unambiguously shows that the lowering of the interface apex was slower and slower as the applied field was increased. The coalescence time t_{coal} can be defined as the time interval between the drops bridging and the instant at which the apex level has decreased by 80%. Fig. 9 shows that for the two working temperatures, the time for total coalescence is an increasing function of E_0 up to a threshold field $(E_0)_{th}$ which depends on the working temperature.

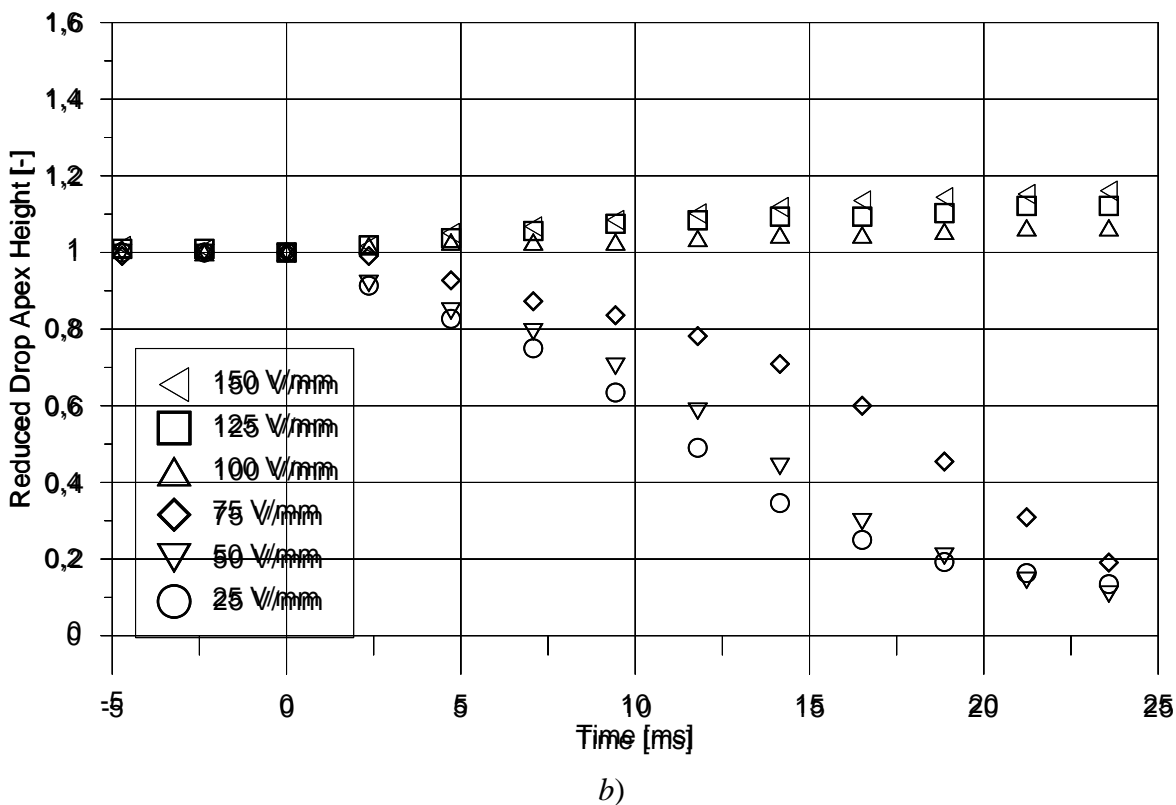
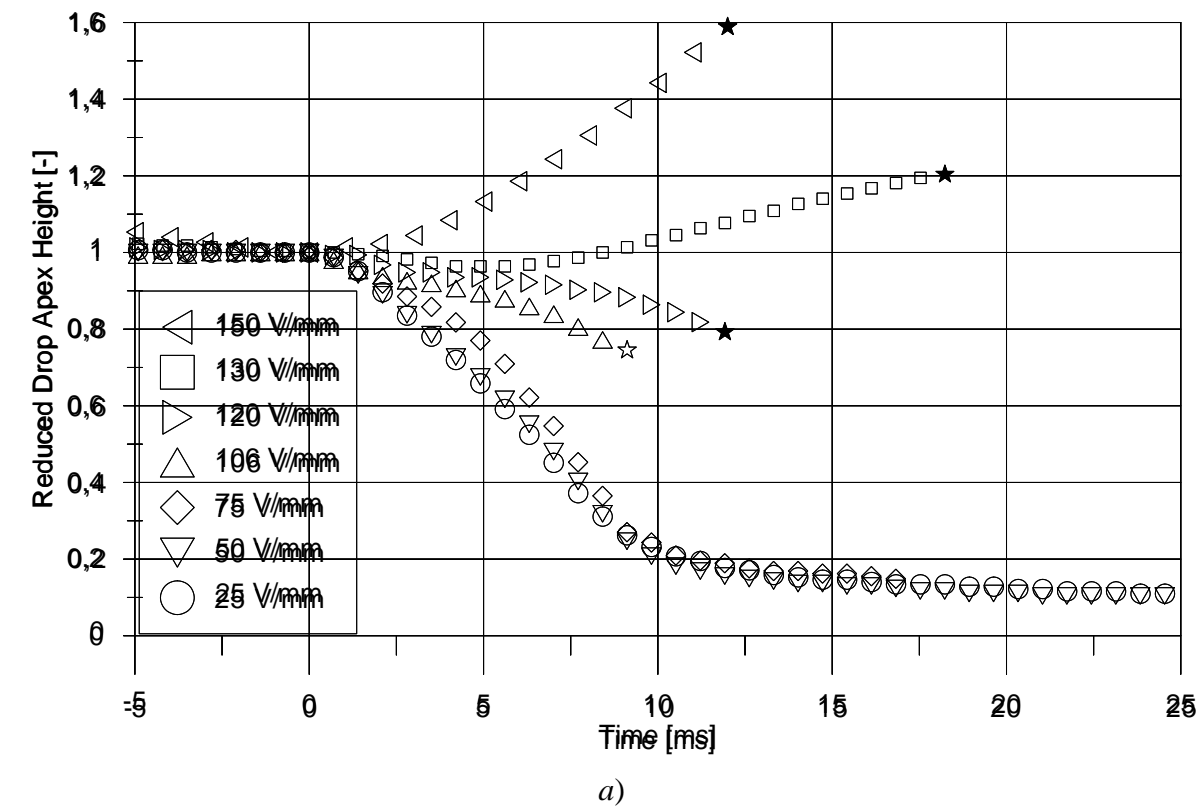


Figure 9. Progress of the apex level of the falling drop during the electrically induced coalescence for different drops and working conditions. An asterisk indicates detachment of a daughter drop (the water bridge is broken). a) $T = 60^{\circ}\text{C}$; b) $T = 40^{\circ}\text{C}$.

From the results shown in Fig. 9, the value t_{coal} of the coalescence time without electric field can be taken equal to that obtained under the small applied field $E_0 = 250$ V/cm. The coalescence time is most often scaled with the inertio-capillary time $t_i = (\rho R^3/\gamma)^{1/2}$ but, as the Ohnesorge number $Oh_2 = \mu_{oil}/(\rho_{oil} \gamma R)^{1/2}$ is on the order of 1, the viscous stresses also influence the coalescence dynamics (see Fig. 9). The results given in Table II are grossly similar to previous ones [29]. No definite conclusion, however, can be derived from the comparison of the present results with those obtained by Aryafar and Kavehpour on silicone oils [29]. Firstly, our experimental conditions are not identical, these authors working with a planar interface; secondly, their results exhibit rather large dispersions and, even, some kind of inconsistency. Otherwise, from Fig. 9, there appears a marked difference between the two temperatures : at 40°C t_{coal} increases by about 30% under $E_0 = 750$ V/cm whereas at 60°C t_{coal} to a first approximation does not depend on the applied field strength, at least up to 750 V/cm. This difference might be due to the existence of a viscoelastic film at the water/oil interface.

Table II Crude oil Ohnesorge number Oh_2 , coalescence time t_{coal} and inertial time scale t_i at the two studied temperatures and comparison with results of a previous study for similar Oh values [29].

T (°C)	Oh_2	t_{coal} (ms)	t_i (ms)	t_{coal}/t_i	$(t_{coal}/t_i)_{Aryafar}$
40	0.7	18	$\cong 1.6$	11	4 to 6
60	0.28	8	$\cong 2$	4	4 to 8

D. Partial coalescence at 60°C

Under electric field strength $E_0 > (E_0)_{th}$ ($(E_0)_{th} \sim 900$ V/cm), due to the electrostatic pressure becoming non negligible compared with the capillary pressure $p_{cap} = 2\gamma/R$, the falling drop was slightly elongated in the field direction. When it approached the bottom drop, its deformation became asymmetrical (see Figs. 10 to 14). Moreover, the top surface of the stationary drop was raised and exhibited a deformation more marked than on the falling drop because of the higher radius of curvature of the stationary drop and the subsequent lower capillary pressure p_{cap} and higher ratio p_{es}/p_{cap} . (this is clearly visible on Figs. 12 to 14).

In the experiments carried out at temperature $T = 60^\circ\text{C}$ for which the crude oil viscosity was moderate ($\mu_{oil} = 25$ mPa.s), under a nominal field $E_0 > (E_0)_{th} \cong 0.9$ kV/cm, partial coalescence was observed (Figs. 10, 11 and 12) and looks very similar to the one observed with pure dielectric liquids [16,17,32]. This is not surprising because the electrostatic pressure induced by the bipolar square applied voltage is very similar to the one with DC voltage as long as the daughter droplet has not detached from the lower drop. We observed that only the lower part of the falling drop tended to flow into the big drop and that the more or less columnar intermediate zone pinched off, leaving a daughter drop and a string of fine droplets created by break-up of the water ligament (Figs. 10, 11 and 12). Clearly, as found with pure dielectric liquids, the daughter droplet has a size increasing with E_0 . Due to the limited field strength that could be applied (for $E_0 > 1.8$ kV/cm the big stationary drop was destabilized and emitted a jet), a sudden transition from partial coalescence to bouncing as found by Hamlin et al. [17] was not observed in our case.

The threshold field of partial coalescence $(E_0)_{th}$ observed at $T = 60^\circ\text{C}$ can be scaled by stating that the electrical and capillary forces are of the same order. In order to obtain an estimate of the

electrical force F_e that the field exerts on the top portion of the protruding interface, let us first recall that the mean applied field E_I experienced by the falling droplet is not the nominal field E_0 but approximately the maximum field E_I at the top of the bottom drop alone; this field is between $3 E_0$ for a half-sphere and $4.208 E_0$ for a sphere resting on a planar electrode [40] and a value of $E_I \cong 4 E_0$ can be used in order of magnitude calculations without causing too large an error. Secondly, after electrical contact, the upward electrical force on the droplet at the top of the much bigger bottom drop should be close to the one on a conducting sphere resting on a plate and subjected to a uniform field E_I , i.e. $F_e \sim 5.476 \pi R^2 \varepsilon E_I^2$ [40]. The resultant capillary force on the remainder of the emptying droplet is given by $F_{cap} = 2\pi b \gamma$ where b is the minimum radius of the neck between the two drops; due to the interface shape evolution, the value of b varies with time (see Figs. 11 and 12) and a typical value is $b = R/2$. Balancing the two forces leads to the following expression for the threshold field :

$$(E_0)_{th} \cong \frac{1}{4} (E_I)_{th} \sim 0.107 \left(\frac{\gamma}{\varepsilon R} \right)^{1/2} \quad (6)$$

At working temperature $T = 60^\circ\text{C}$, this rough estimate leads to $(E_0)_{th} \sim 1.9 \text{ kV/cm}$, a value which is compatible with the experimental results. More specifically, from the curves in Fig. 9-a, it appears that the level of the interface apex exhibits an initial decreasing behaviour when the nominal field E_0 is lower than 1.5 kV/cm . This means that, during the first phase of the partial coalescence process, the capillary force on the upper part of the coalescing droplet is greater than the electric force it experiences; therefore the above estimate is an upper bound for $(E_0)_{th}$.

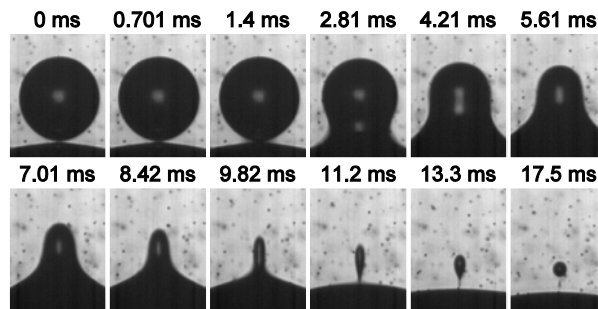


Figure 10. Selected images from a movie showing drop partial coalescence in Grane crude oil at $T = 60^\circ\text{C}$, $D = 742 \mu\text{m}$, $E_0 = 900 \text{ V/cm}$.

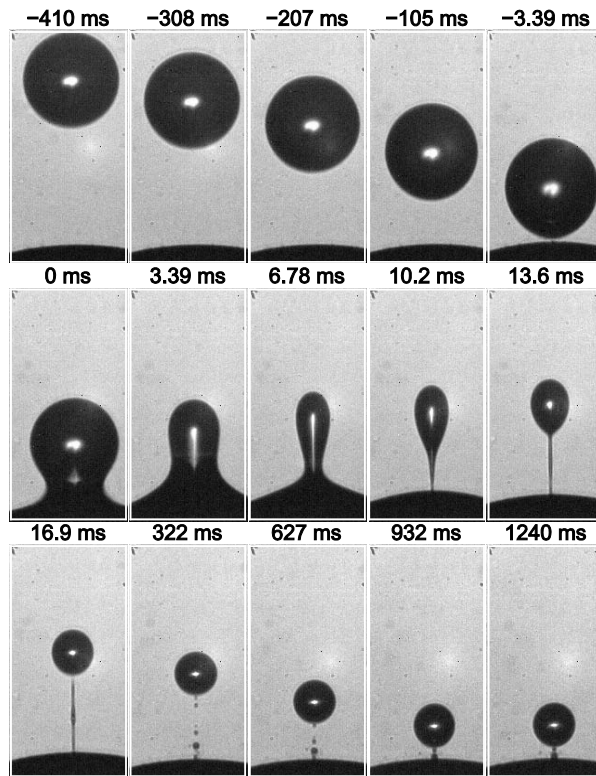
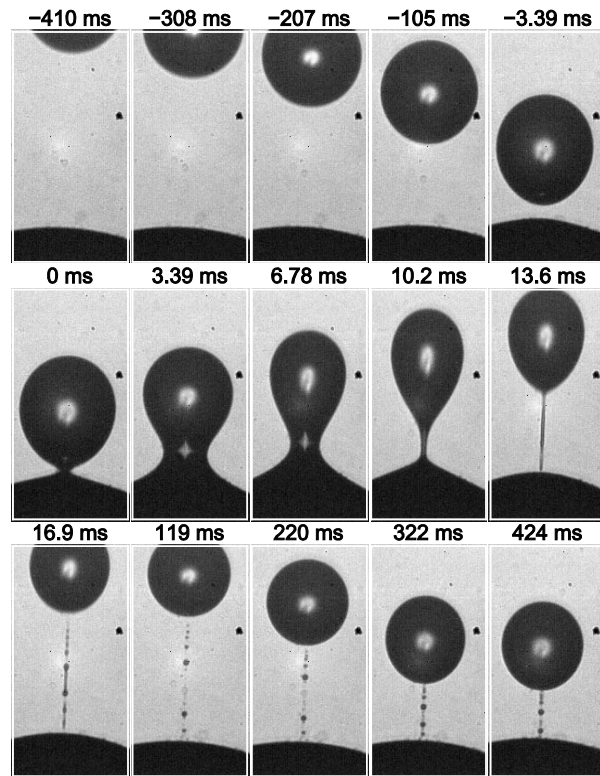
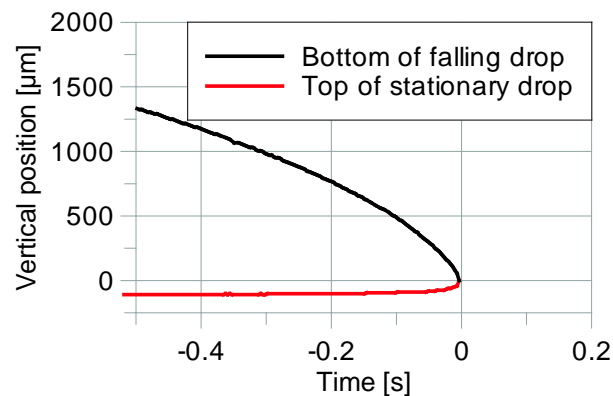


Figure 11. Selected images from a movie showing drop partial coalescence in Grane crude oil at $T = 60^\circ\text{C}$: $D = 710 \mu\text{m}$, $E_0 = 1330 \text{ V/cm}$.

Fig. 9-a provides more accurate information : for a nominal field of moderate strength (E_0 from 1 to 1.2 kV/cm), after the drops bridging there was a very limited lowering of the interface apex before break-up of the ligament. For $E_0 = 1.3 \text{ kV/cm}$, there was first a slow lowering due to an initial electric force F_e lower than the capillary force F_{cap} on the upper part of the coalescing drop; but, as the protruding part partially emptied, the curvature at the apex increased (see Fig. 11) leading to the increase of the field and, hence, of the electrostatic force then becoming higher than the capillary force and inducing the rise of the upper part of the protrusion.



a)



b)

Figure 12. Drop partial coalescence in Grane crude oil at $T = 60^\circ\text{C}$: $D = 730 \mu\text{m}$, $E_0 = 1660 \text{ V/cm}$; a) selected images from movie; b) vertical position of facing interfaces.

Another point worth emphasizing: when breaking up, the ligament should leave the daughter droplet charged. With a DC field, this droplet should move up due to the electrical field action on its charge. In the BSV field conditions the daughter droplet might oscillate, but the used voltage frequency (1 kHz) made such an oscillation totally undetectable. Moreover, as the crude oil conductivity σ_{oil} was high, the charge relaxation time $\tau_{oil} = \epsilon_{oil}/\sigma_{oil}$ was low ($\tau_{oil} \cong 2 \cdot 10^{-4} \text{ s}$) and the droplet charge relaxed totally in about 1 ms. The daughter droplet which then experienced the gravity force and was electrically slightly attracted by the bottom drop, slowly fell down (see pictures taken at 16.9 and 322 ms in Figs. 11 and 12) but did not coalesce during the 2 or 3 seconds of video recording after partial coalescence; the falling was stopped after some time and, presumably, the string of remaining fine droplets between the bottom drop and the daughter droplet played a role in maintaining the droplet at a finite spacing.

E. Non coalescence at 40°C

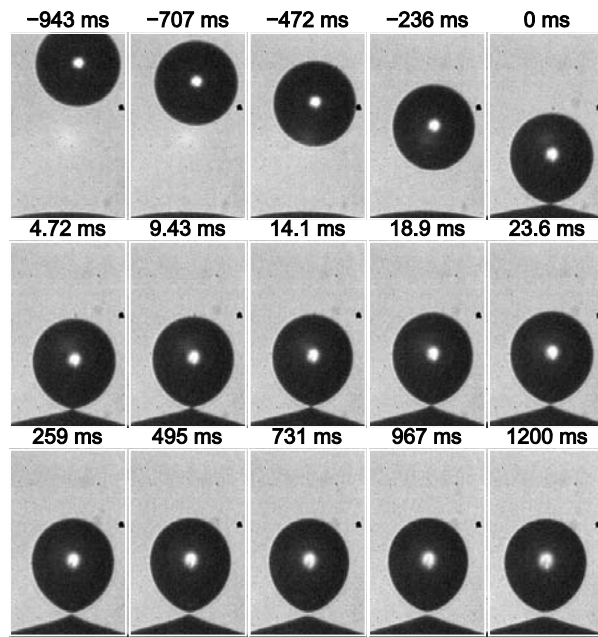
In crude oil at 40°C and under high electric field ($E_0 > \sim 0.8$ kV/cm), just before collision the falling drop was also asymmetrically elongated in the field direction (Figs. 13 and 14), its shape being similar to the one observed at 60°C (Figs. 11 and 12). As already mentioned, the top surface of the big stationary drop was rising towards the falling drop, but now with a nearly conical tip (see Figs. 13 and 14). Beyond the similarities for the two studied temperatures in the approaching phase of the drops, there is a surprising difference in the phenomena following the drops bridging. In contradistinction with what occurred at higher temperature, there was no partial coalescence at 40°C, the drop of apparently constant volume staying at a short distance above the bottom drop (Figs. 13 and 14). At this temperature $T = 40^\circ\text{C}$, there is an abrupt change from coalescence to non coalescence at a critical field on the order of 1 kV/cm. This non coalescence behaviour appears to be similar to the so-called bouncing between drops under high enough field [17-20,33], which is accompanied by a charge exchange between the drops.

Interesting information is gained by examining the behaviour of the two drops after their first contact and exchange of electric charge. Under a DC applied field, the two bouncing drops repel each other and move away from each other due to the electrophoretic force exerted on their charge [19,20,17]. Under a BSV field as used here, a different behaviour can be expected. Indeed, the level of the upper drop was found to be rising during a time of about 30 ms (see Figs. 13-b and 14-b) and the facing interface of the lower drop rose as well while keeping its nearly conical shape; moreover, the spacing between the close interfaces remained approximately constant and very small (Figs. 13-b and 14-b). This indicates that the upper drop was subjected to a mean upward electrical force compensating for the gravitational force F_g and the attraction force due to the stationary drop. As a bipolar square voltage was applied, the only possible explanation for this behaviour is that the mean electrical force during each half-period was directed upward, i.e. an upward (downward) field was acting on positive (negative) surface charge at the top of the droplet. Therefore, the observed repel-like behaviour implies that, at each half period of the applied voltage, a charge was transferred between the lower drop (lying on the bottom electrode) and the upper droplet.

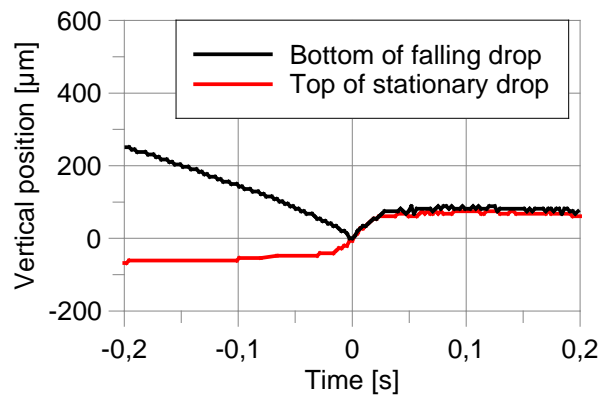
In the drops bouncing studies, it was shown that a short and thin columnar water bridge exists during a time which can be very short ($\ll 10^{-3}$ s – see [20,33,34]) or long (~ 1 s – see [41]) depending on the various parameters (in particular drop radius and oil viscosity). In the present case, it can be conjectured that, at every polarity reversal of the applied voltage, a water bridge (presumably due to a jet issuing from the bottom drop apex) connected the two drops for a short time and provided the way for transferring charge between them. In this case the characteristic time of charge transfer τ_{charge} was no longer the relaxation time τ_{water} (τ_{charge} depends on the geometry of the water bridge). In order to estimate τ_{charge} let us assume a cylindrical bridge of radius r and length λ ; the current through the bridge is $I = \pi r^2 j$ where j is the current density : $j = \sigma_{water} E$, E being the field in the bridge. This current has to change the droplet charge from $+Q$ to $-Q$ or vice versa; hence we have $\tau_{charge} \sim 2Q/I$. Once fully charged, the droplet is at the potential of the bottom drop; assuming that τ_{charge} is lower than the life time of the bridge, the value of Q is close to the one of a conducting sphere on a planar electrode [40] : $Q \cong (2\pi^2/3) \pi R^2 \epsilon_{oil} E_1 \cong 6.58 \pi R^2 \epsilon_{oil} E_1$ E_1 being the mean field above the bottom drop in the absence of a droplet. At the applied voltage reversal, due to the existing charge Q and the reversed applied field E_0 , the droplet is initially brought to a potential $\Delta V \sim 2 R E_1$; just after a bridge connects the two drops, the field in the bridge is $E \cong \Delta V / \lambda$; this gives :

$$\tau_{charge} \sim 6.6 \frac{\epsilon_{oil}}{\sigma_{water}} \frac{R\lambda}{r^2} \quad (7)$$

For a thin water bridge of length at most 10 times its diameter ($\lambda/r \leq 20$), with a radius $r \geq 1 \mu\text{m}$, we would have $\tau_{charge} < \sim 2 \cdot 10^{-7} \text{ s}$; in this case a bridge life time of a few microseconds is long enough to result in a full charging of the droplet.

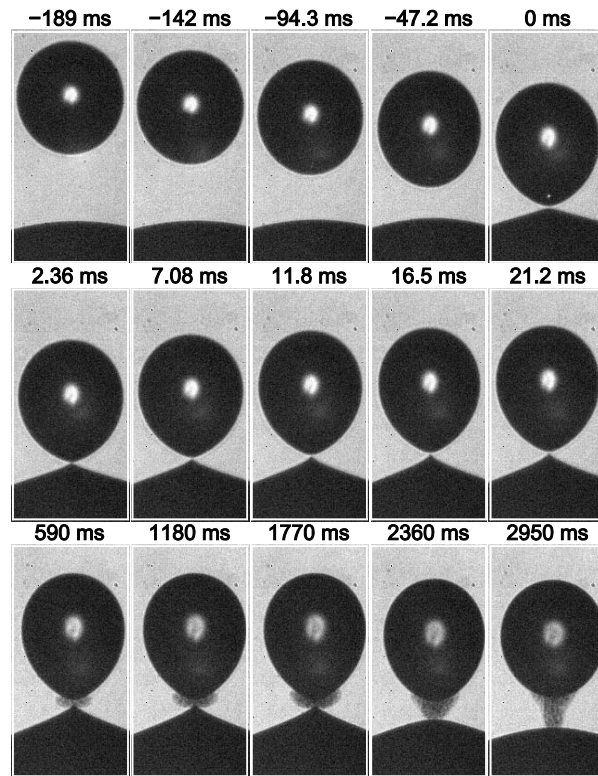


a)

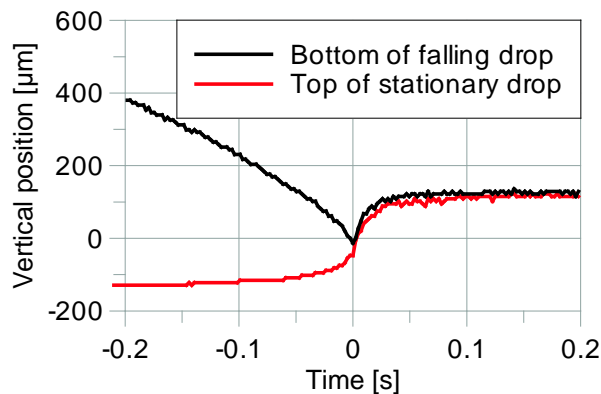


b)

Figure 13. Non coalescence of the droplet in Grane crude oil at $T = 40^\circ\text{C}$: $D = 730 \mu\text{m}$, $E_0 = 1250 \text{ V/cm}$; a) selected images from movie; b) vertical position of facing interfaces.



a)



b)

Figure 14. Non coalescence of the droplet in Grane crude oil at $T = 40^{\circ}\text{C}$: $D = 800 \mu\text{m}$, $E_0 = 1500 \text{ V/cm}$; a) selected images from movie; b) vertical position of facing interfaces.

Fig. 14-a exhibits a totally unexpected phenomenon : after a few tenths of second, a ring of extremely fine droplets was observed at the bottom of the upper drop; the zone of mist was slowly swelling with time and, after about two seconds, there was a definite change, the top interface of the stationary drop becoming smoother and the cloud of fine droplets taking a more columnar shape between the facing interfaces of the drops (see picture at 2360 ms in Fig. 14). Presumably the generation of the fine droplets arises from the breakup of the temporary bridge connecting the two very closely spaced drops at each voltage reversal.

5. Discussion

The above presented results concerning the effect of an applied field on the coalescence of water drops immersed in a crude oil and falling on a much bigger immobile water drop are not qualitatively very different from what has been observed for drops of conducting liquid immersed in a simple dielectric liquid. Nevertheless, the change of scenario above a threshold electric field – transition to partial coalescence or to non-coalescence – by only changing the working temperature is unexpected. Working with charged water droplets of low enough conductivity falling in model oils, Hamlin et al. [17] showed that by increasing the applied DC field, the post-impact behaviour passes from total to partial coalescence and, above a second field threshold, to non-coalescence, the droplet then bouncing away from the bottom meniscus. These authors also investigated the influence of water conductivity σ_{water} and showed that, for a given applied field, there is a change from partial coalescence to bouncing above a field dependent threshold of σ_{water} . But what is observed in the present case of Grane crude oil is the converse : by raising the temperature, the water conductivity is increased but we pass from non-coalescence to partial coalescence. Moreover the rule that could be deduced from the results of Hamlin et al. does not apply here because the conductivity of the salted water we used was very much higher than the transition conductivities found in [17].

The two scenarios we observed concerning the field influence are not specific to the studied crude oil. In the case of simple liquid systems (where possibly some classical surfactants, but not asphaltenes or resins, are present), the two scenarios have also been observed. For drops of similar or clearly different size, by increasing the applied field, the behaviour passes from coalescence to bouncing in many cases [19,20,33,34] but partial coalescence also occurs, at least for drops falling on a planar interface [16,17,32]. The difference presumably does not arise from the different viscosity values of the crude oil at 40°C and 60°C : the field induced bouncing of drops of conducting liquid has been observed in various simple oils of viscosity ranging from 10 to 1000 mPa.s [20,33].

What is the physical property or phenomenon responsible for the scenario changing with temperature ? The fact that the threshold fields are nearly the same for both temperatures may appear fortuitous because the hypothesis of balance of electrical and capillary forces, leading to the estimate (6), does not apply at 40°C, the capillary force due to the transitory thin bridge being negligible. The similar values of the threshold fields may arise from an initially similar phenomenon whose further evolution drastically differs. We suspect that the transition to bouncing or partial coalescence is determined by the step of temporary bridge connecting the drops. As already argued [19,20,34], the initial shape of the bridge plays an important role on its further evolution. If the ratio λ/r of bridge typical length λ and radius r is small enough, the capillary forces result in a quick swelling of the bridge [19,20,34] and lead to a thick neck that can subsequently pinch off (partial coalescence scenario). If the ratio λ/r is large enough, conversely, the pressure in the thin bridge is higher than in the two adjacent drops and it tends to expel the water outside the bridge which breaks up rapidly (bouncing scenario).

We thus guess that at 40°C the bridge connecting the two drops is longer than at 60°C. This guess can be inferred from the observation of the drops shape in Figs. 11 to 14. Just before bridging, the droplet and the bottom drop interfaces tend to become conical but the deformation is more acute at 40°C than at 60°C; therefore the electric field near the cone tips very likely is stronger at 40°C than at 60°C for a same drops spacing; this should lead to the ejection of a thin jet for a larger drops spacing at 40°C than at 60°C, thus resulting in a longer bridge. The generation of very fine droplets at 40°C (Fig. 14a) for a strong enough applied field also supports the hypothesis of transitory existence of a bridge with large enough λ/r ratio; such a thin thread then breaks up at least at two locations (close to the two drops) which leaves at least one very small satellite droplet.

If the above guess is true, the question that remains is to understand why the deformation of the interfaces appears to be smaller at 60°C than at 40°C despite the fact that the interfacial tension is similar. Such a difference with the behaviour observed in model oils might be due to the complex properties of water drops in crude oils, in particular, those resulting from the existence of a very thin film between water and oil built by the asphaltene compounds and resins present in the crude oils at important concentrations. A recent study in a dilute crude oil showed the existence of such a very thin film characterized by a viscoelastic behaviour for the water drops [42]. A tentative explanation is that, at 60°C, a noticeable interfacial film imparts a surface elasticity that can alter the deformation of the interfaces. This film would be responsible for a smoother deformation of the interfaces before bridging at 60°C than at 40°C; also, it would explain that the influence of the electric forces on the dynamics of full coalescence is much smaller at 60°C than at 40°C as can be viewed by comparing the curves for $E_0 \leq 750$ V/cm in Figs. 9a and 9b.

In any case, there is a very limited understanding concerning the electrically induced instability of the facing interfaces that leads to the generation of the bridge; moreover, knowledge about the interfacial films between water and crude oil is at the very beginning. A study of these two questions is a prerequisite before attempting to delineate the domains of occurrence of the transitions to partial coalescence or to non-coalescence.

6. Conclusions

Experiments based on the use of a NIR video camera have been performed to study the effect of electric field on the coalescence of water droplets falling, in a crude oil, onto a big water drop. The investigation reported in this paper provides new results concerning electro-coalescence in crude oil which should be of interest for the application of compact electrocoalescers in petroleum industry. The electric field induces forces that affect both the water drop motion in crude oil and the coalescence process. Up to a threshold field, the surface charge density redistributed just after bridging of the two drops and charge exchange slows down the coalescence process.

Above this threshold field, the coalescence was not total and two different transitions were found : from full coalescence to partial coalescence at 60°C and to non-coalescence at 40°C. Each of these two scenarios has been observed in the case of model oils without resins and asphaltenes but the fluids properties determining either the partial coalescence or the bouncing behaviour are not yet well characterized. The peculiarity of the studied crude oil is that there was a scenario change by changing the working temperature (which, of course, affects many properties of the system).

With the bipolar square wave shape of the field that was applied, at the lowest investigated temperature (40°C) and above the field threshold, there was non-coalescence of the falling droplet which tended to rise while attracting and raising the conical part of the deformed bottom drop This demonstrates the exchange of charge through a transitory and short lived bridge between the drops and, therefore, a bouncing at every reversal of the applied field. Moreover, at high enough applied field, the generation of a mist of very fine droplets around the impact zone indicates that satellite droplets were created by the breakup of the thin thread when its length to radius ratio was high enough.

For model oils, the transition to partial coalescence or bouncing is not really characterized in terms of applied field, interfacial tension, properties of the oil like permittivity and viscosity, water conductivity, presence of surface active compounds, etc... The generation of the thread connecting the two drops and its initial shape appear to play a crucial role : a small length to radius ratio λ/r of the bridge should lead to partial coalescence whereas a large one should promote the so-called electrical bouncing. Now, to our knowledge, there is no theoretical approach delineating the two scenarios on the basis of the evolution of the bridge; in any case, the dynamical analysis to develop is far from being a straightforward problem and requires much effort.

In our case of a water drop falling in a crude oil, the problem is further more intricate because of the presence of various surface active compounds that modify the static interfacial tension and might influence the dynamic evolution of the interface subjected to a shear stress. The presence of asphaltenes and resins is of primary importance because these compounds can build an interfacial film promoting a visco-elastic behaviour of the drops. The present study suggests the existence of such a film at 60°C but its absence or its negligible influence at 40°C. Therefore the understanding of the phenomenon of electrocoalescence in crude oils requires first a better knowledge of the interfacial film whose rheological properties are far from being fully clarified.

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