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Electrostatic phase separation: A review

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ABSTRACT

The current understanding and developments in the electrostatic phase separation are reviewed. The literature covers predominantly two immiscible and inter-dispersed liquids following the last review on the topic some 15 years. Electrocoalescence kinetics and governing parameters, such as the applied field, liquid properties, drop shape and flow, are considered. The unfavorable effects, such as chain formation and partial coalescence, are discussed in detail. Moreover, the prospects of microfluidics platforms, non-uniform fields, coalescence on the dielectric surfaces to enhance the electrocoalescence rate are also considered. In addition to the electrocoalescence in water-in-oil emulsions the research in oil-in-oil coalescence is also discussed. Finally the studies in electrocoalescer development and commercial devices are also surveyed.

The analysis of the literature reveals that the use of pulsed DC and AC electric fields is preferred over constant DC fields for efficient coalescence; but the selection of the optimum field frequency a priori is still not possible and requires further research. Some recent studies have helped to clarify important aspects of the process such as partial coalescence and drop-drop non-coalescence. On the other hand, some key phenomena such as thin film breakup and chain formation are still unclear. Some designs of inline electrocoalescers have recently been proposed; however with limited success: the inadequate knowledge of the underlying physics still prevents this technology from leaving the realm of empiricism and fully developing in one based on rigorous scientific methodology.

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Contents

1. Introduction	178
2. Electrocoalescence dynamics	179
2.1. Electrostatic force of attraction between drops	179
2.2. Shapes of coalescing drops	180
2.3. Damping forces	181
2.4. Thin film breakup	182
3. Critical conditions for electrocoalescence	182
4. Effect of turbulence and shear flow	183

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5. Chain formation.....	183
6. Partial coalescence.....	184
7. Effects of operating parameters and fluid properties.....	186
8. Electrocoalescence on dielectric surfaces.....	187
9. Effects of drop size and polydispersity.....	187
10. Non-uniformity of the applied field.....	188
11. Electrocoalescence in leaky dielectric emulsions.....	188
12. Electrocoalescence in microfluidics devices.....	188
13. Available technologies.....	189
14. Conclusions.....	191
Acknowledgement.....	192
References.....	192

Nomenclature

a, b	drop radii (m)
A	Hamaker constant (J)
Be, Be_{crit}	dimensionless number and its critical value
Ca_E	electrical capillary number
C_d	drag coefficient
d	minimum distance between leading edges of drops (m)
d_0	initial separation between leading poles (m)
d_{crit}	critical separation between leading poles (m)
d_d	drop diameter (m)
E	electric field (V/m)
E_0	applied electric field (V/m)
E_c	critical field for drop breakup (V/m)
E_{crit}	critical field of coalescence (V/m)
$E_{crit,int}$	critical field for stability of flat interface (V/m)
f	frequency of applied field (Hz)
f_0	optimum frequency (Hz)
f_{th}	threshold frequency (Hz)
F_D	drag force (N)
F_e	electrostatic force (N)
F_r, F_θ	radial and angular components of electrostatic force (N)
F_v	film-thinning force (N)
g	gravitational constant (m/s ²)
I_p	parameter for acceleration of drop in electric field
κ_1, κ_2	coefficients
K_1, K_2	coefficients
L	distance between drop and ground electrode (m)
M	potential energy of dipole moment (J)
Oh	Ohnesorge number
P	electric dipole moment (C m)
Q	charge (C)
Re_d	drop Reynolds number
s	separation between drop centers (m)
s_0	initial separation between drop centers (m)
s_{crit}	critical separation of coalescence (m)
t_c	critical thickness of film rupture (m)
V	electric potential (V)
We	Weber number
WO	dimensionless number
ΔV	potential difference between leading poles (V)
ΔV_{crit}	critical potential difference (V)

Greek symbols

$\Delta\rho$	density difference of dispersed and medium phase (kg/m ³)
$\varepsilon_d, \varepsilon_m$	permittivities of dispersed and medium phase, respectively (F/m)
γ	interfacial tension of drop-medium interface (N/m)
γ'	shear rate (1/s)
κ	coefficient
λ	viscosity ratio μ_d/μ_m
v	velocity of drop approach (m/s)
μ_d, μ_m	viscosities of dispersed and medium phase, respectively (Pa.s)
ρ_d, ρ_m	densities of dispersed and medium phase, respectively (kg/m ³)
σ_d, σ_m	conductivities of dispersed and medium phase, respectively (S/m)
θ	angle made by a line joining centers of drops with direction of an applied electric field

1. Introduction

Dispersions of one fluid in another immiscible fluid can be found in many natural as well as synthetic products, such as milk, petroleum, food products, drugs, paints, etc. The separation of the two phases becomes necessary to recover or purify the product, e.g. water separation from crude oil, phase separation in solvent extraction, glycerol separation from the bio-diesel (Abeynaïke et al., 2012), etc.

Crude oil naturally contains a significant amount of water. The water is in dispersed form in tiny droplets of average size less than 50 μm (Atten, 1993). The crude oil from an oil well may contain a large fraction of water depending on the age of the well and it has to be reduced to less than 0.3% before further processing (Atten, 1993; Less and Vilagines, 2012). The removal of water from the crude oil is crucial to avoid corrosion of the process equipment, possible catalyst-poisoning and extra transportation cost owing to increased volume as well as increased viscosity. The water separation from the crude oil can be self-induced or influenced by the external forces. In an emulsion, droplets can also coalesce while moving due to gravity or buoyancy. Such kind of conventional methods of demulsification of crude oil are effective only when the size of the droplets is large. Due to the very small drop-size and the stable interface, gravitational settling takes too long, i.e. hours and sometimes days to separate water from crude oil;

moreover, the separation cannot be to the desired extent. In an emulsion, phase separation speed can be governed by the probability of drop–drop contact. Therefore, the phase separation process can be enhanced by stimulating the relative motion between the droplets, using external forces such as mechanical, thermal, electrostatic and chemical or combination of some of these means (Eow et al., 2002; Klasson et al., 2005; Sun et al., 1999; Wu et al., 2003).

Inspired by the invention of an electrostatic precipitator by Frederick Cottrell in early 20th century (Cottrell and Speed, 1911), the use of electrostatic forces for the phase separation led to a flurry of publications in the first third of that century (Speed, 1919; Muth, 1927). The technique involving the use of the electric field in phase separation of liquid phases, commonly known as electrocoalescence, has been developed and extensively investigated due to its fast, clean and efficient coalescence capabilities (Eow et al., 2001). Apart from water separation from oil, electrocoalescence can also be employed in phase separation operations such as solvent extraction and dispersion as well as to fractionate mixed oils (Scott and Wham, 1989). In industrial applications such as crude oil demulsification, electric field is commonly used to help the small water droplets to come closer to each other and eventually merge into each other or with an interface. The larger drops can then be easily settled by gravity, resulting in a final product that contains water below a prescribed level. The application of electric field increases the coalescence rate as well as enhancing the migration speed of the droplets towards the electrodes, facilitating phase separation.

Research into electrocoalescence has resulted in many findings, which have helped to make the phase separation faster. Several studies, identifying the challenges in the electrocoalescence process and the methods to resolve them, have been carried out (Pearce, 1953; Zhang et al., 2011; Rayat and Feyzi, 2012; Noik et al., 2002; Fjeldly et al., 2008; Midtgard, 2009). The factors directly affecting the rate of electrocoalescence are many; namely the chain formation, fluid motion, type (DC or AC) and frequency of the applied field, partial coalescence, electrical and physical properties of fluids, etc. The physical and electrical properties of the crude oil from different reservoirs can be different; also they can vary with the age of an oil well (Berg et al., 2010). Since these properties govern the stability of the emulsion, identifying the optimum operating parameters is difficult. It follows that designing a universal electrocoalescer which can handle crude oil from variety of oil reservoirs is still very challenging.

2. Electrocoalescence dynamics

2.1. Electrostatic force of attraction between drops

The mechanism of coalescence of two drops in the presence of an electric field involves three distinct steps. The applied electric field polarizes an individual drop and each drop acts as a dipole with induced positive and negative charges at two polar ends. The dipole aligns in the direction of applied electric field. The first step of electrocoalescence involves the interaction of two drops due to attraction between opposite polarity poles. In the large separation limits, the different forces acting on a drop can be electrostatic force, drag force and gravitational force (if drops are coarse). When two drops move towards each other, at small separation there exists an interstitial film of the medium fluid between the leading

faces of the drops. The second step of electrocoalescence involves the squeezing of fluid at the plateau border of the thin film. As drops come closer, the thickness of the film reduces further. In the third step, when the film becomes very thin, the coupled action of electrostatic and molecular forces break the film allowing the two drops to merge together.

If drops carry inherent charges, ‘migratory coalescence’ results due to the electrophoresis (Williams and Bailey, 1986). In addition to driving drops closer, an electric field also enhances the thin film breakup. In one of the first studies of electrocoalescence, Berg et al. (1963) found that for two anchored drops, the coalescence rate was proportional to the strength of the applied electric field (E_0) when E_0 was small; whereas if E_0 was high, the rate was found to be proportional to E_0^2 . Apart from the magnitude of the applied electric field, there are many other parameters which can influence the force of attraction between the coalescing drops. They are the inter-drop separation, size of drops, shape distortion and fluid properties such as conductivity, permittivity, viscosity, interfacial tension, etc. The subsequent studies in electrocoalescence were focused on these parameters, their optimization and consequently making the process faster.

The radial (F_r) and angular (F_θ) components of the electrostatic force of attraction between two conducting spherical drops of radii a and b in a dielectric medium (as in Fig. 1) are given as (Waterman, 1965; Atten, 1993),

$$F_r = -12\pi\epsilon_m b^3 E_0^2 \frac{a^3}{(d+a+b)^4} (3\cos^2\theta - 1), \quad (1)$$

$$F_\theta = -12\pi\epsilon_m b^3 E_0^2 \frac{a^3}{(d+a+b)^4} \sin 2\theta. \quad (2)$$

Eqs. (1) and (2) can be rewritten for the uniform sized drops ($a=b$) aligned in the direction of the applied electric field ($\theta=0$) as (Waterman, 1965; Atten, 1993),

$$F_e = -24\pi\epsilon_m a^2 E_0^2 \frac{a^4}{(d+a+b)^4}. \quad (3)$$

The force F_e (which is proportional to E_0^2) is dielectrophoretic in nature. It is a short range force and Eq. (3) becomes invalid when the separation becomes such that $d/a \ll 1$. In that case Dipole-Induced-Dipole (DID) model gives more accurate estimate of the electrostatic force of attraction (Yu and Jones, 2000; Siu et al., 2001). DID model is based on the assumption that when two dipoles are in close proximity, a point-dipole induces its multiple reflections. The radial (F_r) and angular (F_θ) components of electrostatic force, using DID model, can be expressed in a more compact form as follows (Lundgaard et al., 2006),

$$F_r = -12\pi\epsilon_m b^3 E_0^2 \frac{a^3}{(d+a+b)^4} (3K_1 \cos^2\theta - 1), \quad (4)$$

$$F_\theta = -12\pi\epsilon_m b^3 E_0^2 \frac{a^3}{(d+a+b)^4} K_2 \sin 2\theta. \quad (5)$$

The coefficients K_1 and K_2 are expressed as,

$$K_1 = 1 + \frac{a^3 s^5}{(s^2 - b^2)^4} + \frac{b^3 s^5}{(s^2 - a^2)^4} + \frac{3a^3 b^3 (3s^2 - a^2 - b^2)}{(s^2 - a^2 - b^2)^4}, \quad (6)$$

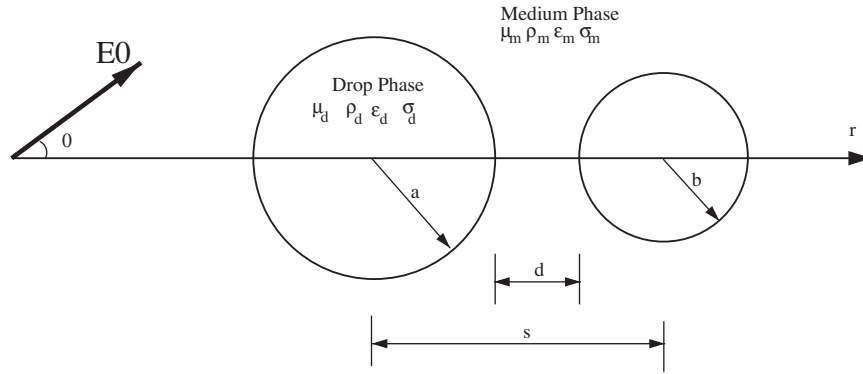


Fig. 1 – Interaction of two drops in an electric field.

and

$$K_2 = 1 + \frac{a^3 s^3}{(s^2 - b^2)^3} + \frac{b^3 s^3}{2(s^2 - a^2)^3} + \frac{3a^3 b^3}{(s^2 - a^2 - b^2)^3}. \quad (7)$$

When the drop–drop separation is large ($d > a, b$), the coefficients K_1 and K_2 are equal to 1 and Eqs. (4) and (5) reduce to Eqs. (1) and (2), respectively.

The effect of separation on drop shape and in turn on the rate of electrocoalescence has been the topic of many studies (Latham and Roxburgh, 1966; Atten, 2005; Atten et al., 2006; Raisin et al., 2008). Since the electrostatic force between neighboring drops is short range, for two drops to attract and coalesce, they must be within an ambit which is determined by a combination of the applied field, drop size and fluid properties. In an emulsion, the inter-drop separation is determined by the content and the size of the dispersed phase (Panchenkov and Vinogradov, 1970). The electrostatic interaction of two falling drops in a quiescent oil was studied by Pedersen et al. (2004) using numerical simulations and experiments. Their results suggested that the point dipole model can be used for accurate estimation of electric forces when $d > a$. At the separations $d < a$, the actual electric field between inner poles and the effect of film drainage should be considered.

The estimation of the actual electric potential difference (ΔV) between leading poles of the two approaching drops in an external electric field (E_0) has been a crucial factor in electrocoalescence calculations. However, the present models to estimate electrostatic interaction force between two drops are based on externally applied electric field. The relationship between ΔV and E_0 suggested by Davis (1964) for closely spaced drops ($0.001 < d/a < 0.1$) is mathematically hard to use. The magnitude of the induced charge at the interface of a coalescing drop, due to the applied field, can be estimated as,

$$\pm Q = \kappa \pi a^2 \epsilon_m E_0. \quad (8)$$

Assuming the value of coefficient $\kappa = 5$ for two uniform sized drops aligned in the direction of field for separation $0.001 < d/a < 0.1$, Atten and Aitken (2010) derived a simpler relationship between ΔV and E_0 as,

$$\Delta V \cong \frac{\kappa}{2} \frac{2aE_0}{\log \left[1.78 \frac{a}{s} \right]}. \quad (9)$$

Eq. (9) is in good agreement with Davis's (1964) expression in the separation range of $0.001 < d/a < 0.1$.

A theoretical study of two colliding spheres was carried out by Friesen and Levine (1992), who developed a method

to calculate the interaction energy and the force between two charged, conducting spheres in a uniform electric field. The deformation and aggregation of droplets in an emulsion alter its viscoelastic properties such as yield properties (Mason et al., 1996). Detailed analysis of the motion and interaction of two drops, falling in a quiescent medium, parallel and perpendicular to the applied external field was done experimentally as well as numerically by Chiesa et al. (2005). The study was more focused on flow circulations, dipoles and surface tension gradients. The analysis indicated that the expressions for drag, buoyancy and film-drainage, applicable for the rigid particles do not give the accurate results for the two-drop interaction due to the internal flows as well as variation in the surface tension in the presence of a electric field (Chiesa et al., 2005). The simultaneous effects of the hydrodynamics and electric stresses on a pair of drops were studied by Raisin et al. (2011a). They suggested an experimental setup for generation of a pair of uncharged drops, putting them in the Poiseuille flow and applying an electric field to study the electrohydrodynamic interactions.

2.2. Shapes of coalescing drops

An immediate effect a drop shows upon application of an electric field is shape deformation. Two closely placed drops in an electric field exhibit deformation when the electrocapillary number (or electrical Weber number), $Ca_E = \epsilon_m a E_0^2 / \gamma$ is large. At small Ca_E , the deformation can be observed only at the leading poles of the coalescing drops. Such deformation is the result of high electric field between leading edges of the two drops and thus high charge density. The Taylor's factor ($E_0 \sqrt{(2a\epsilon_m/\gamma)}$) for the stability in an electric field (Taylor, 1964) which is 0.648 for a single drop is lower for a drop in a pair (Latham and Roxburgh, 1966; Brazier-Smith, 1971) and depends on the inter-drop separation. A drop can also show fore and aft asymmetric deformation when it is near to the electrode surface (Imano and Beroual, 2006). The presence of drops in the proximity of electrodes not only induces deformation, but also shields the inner drops in the emulsion.

Adamiak (1999) investigated the deformation of two perfectly conducting, uniform size drops in a uniform electric field numerically. He used the Finite Element Method (FEM) to solve the shape change equations while the electric field distribution over the drop interface was solved by using the Boundary Element Method (BEM). At large separations ($s > 20a$), drops do not interact with each other and they deform like a single drop in the electric field. Raisin et al. (2008) studied the deformation and coalescence of two anchored conducting drops in an electric field. Their results with asymptotic and

numerical methods indicate that for closely held uniform size drops, coalescence occurs when leading faces deform such that $d/d_0 \approx 0.5$. Bjorklund (2009) used the coupled Level-Set method and Ghost-Fluid method for the numerical investigation of two drops in an electric field. In a uniform field, when equal size drops align in vertical direction with the electrodes, drops can interact only when they are within a critical separation. If drops are far apart, they attract to the electrodes due to the mirror charges across the electrode (Bjorklund, 2009). Imano and Beroual (2006) reported similar observations for a single as well as multiple drops resting on a dielectric surface in AC electric field. They carried out theoretical and experimental investigation of influence of an electrode on shapes and coalescence of drops. A drop nearer to an electrode stretch asymmetrically due to unbalance of electric forces. The contact time of two freely suspended as well as anchored drops in an electric field was studied by Raisin et al. (2011b). They found that the contact time was inversely proportional to the initial maximum electrostatic pressure. They also studied the deformation of water–air and water–oil interfaces in electric field using Finite Element Arbitrary Lagrangian Eulerian (FE-ALE) method with moving meshes in COMSOL Multiphysics (Raisin et al., 2011c). In a flowing emulsion under a uniform electric field the coalescence of drops depends on deformation of facing surfaces, their motion and the drainage of the oil film between drops. The time of contact for very small and closely placed droplets with high viscosity ratio ($\lambda = \mu_d/\mu_m$) is larger compared to deformable larger drops.

2.3. Damping forces

When two approaching drops are at a fairly large separation distance the drag force (F_D) opposes the motion. For a rigid sphere in Stokes regime F_D can be given as (Davis et al., 1989),

$$F_D = 6\pi\mu_m a v. \quad (10)$$

However, in the case of a drop motion in viscous medium the actual drag force experienced by drop is lower than that given by Eq. (10) on account of the circulations on both sides of the interface. Hadamard–Rybczynski equation gives an accurate estimate of the drag force on drop, which is written as,

$$F_D = 4\pi\mu_m a c v, \quad (11)$$

where $c = (3\lambda + 2)/(2(\lambda + 1))$

The resistance to the motion of the coalescing drops at the large separation distances ($d > a$) is predominantly due to drag force whereas at $d \ll a$, it is governed by the film thinning force. Film thinning force (F_f) is the force due to drainage of the interstitial liquid film between about to coalesce drops. If the distance between the leading edges is very small relative to drop radii and the flow is within the Stokes regime, the expression for the resistive film thinning force can be written as (Davis et al., 1989; Chiesa et al., 2006),

$$F_f = -6\pi \frac{\nu\mu_m}{d} \left(\frac{ab}{a+b} \right)^2 f, \quad (12)$$

In Eq. (12), $f = 1$ if drops are treated as rigid spheres. The force F_f resists the drainage of fluid from the film trapped between the two drops. Different expressions of F_f have been

proposed for the drops (Vinogradova, 1995; Barnocky and Davis, 1989).

Eq. (12) indicates that apart from drop radius and separation (d), the medium phase viscosity (μ_m) plays a major role in the film drainage stage of drop–drop coalescence. Chiesa et al. (2006) reported that in the absence of an electric field, the film thinning force (F_f) increases with decreasing viscosity of the medium fluid. Increase in F_f can be attributed to the increase in relative velocity of drops on the decreasing medium viscosity. On applying an electric field, the dipolar force between leading edges of drops opposes the film thinning force. The viscosity effect diminishes as drops approach each other and completely vanishes at onset of the coalescence (Chiesa et al., 2006).

The presence of surfactants at the interface, accompanied by drop elongation, causes the interfacial tension gradient leading to Marangoni stresses. This inhibits the generation of internal circulations. Levan (1981) took in to account the effects of induced circulations and interfacial tension gradient to get a revised expression for drag coefficient. The presence of surfactants strongly alters the drag force by the formation of stagnant caps (Hamlin and Ristenpart, 2012). Depending on the adsorption and desorption rate of the surfactant molecules, at very low and very high concentrations a drop obeys Hadamard–Rybczynski model. However at intermediate concentrations Stokes expression gives better estimates for drag force.

The deformability of the coalescing drops plays a crucial role in the film drainage stage of the coalescence. With a numerical study of the coalescence of two drops in a flowing emulsion in uniform electric field, Raisin et al. (2011c) concluded that the rate of electrocoalescence depended on the deformation of the leading surfaces, drop motion and the drainage of the oil film between the drops. The time of contact for closely spaced very small droplets with high viscosity ratio is longer compared to deformable larger drops. The externally induced fluid flow also determines the rate of film thinning. Giljarhus and Munkejord (2011) used FEM to solve the head-on collisions of two drops in the flowing medium and predicted that as the flow capillary number ($Ca = \mu_m \gamma' a / \gamma$) increases, drop deforms more and the contact area becomes larger. It takes a longer time to drain the film, and thus the coalescence time is longer. However, the coalescence time decreases with an increase in the electrocapillary number (Ca_E). Similar observations were reported by Dong et al. (2002). Their experimental observations suggest that decreasing interfacial tension in the absence of electric field can resist the coalescence as large deformations (due to high Ca) inhibit the film thinning. However, in the presence of electric field the increased deformation on decreasing interfacial tension assists the coalescence.

Depending on the Ohnesorge number, $Oh = \mu_d / \sqrt{(\rho_d \gamma a)}$, coalescence has two dynamical regimes: viscous or Stokes regime, which always dominates at sufficiently early times when the neck radius is microscopically small, and an inertial regime that occurs at later times if viscous effects become negligible. Paulsen et al. (2012) from their experimental and numerical results, showed that an additional regime of coalescence dynamics exists called inertially limited viscous regime. Both, inertia and viscous forces play a role in this regime. The inertia associated with each drop moving as a rigid object prevents the system from being in the Stokes flow regime. The coalescence dynamics cannot be in the Stokes flow regime until the surface tension force around the neck is large enough

to rigidly translate two initially stationary drops towards each other.

The rate of drop–drop approach can be estimated using expressions for electrostatic force of attraction and drag force. [Atten \(1993\)](#) proposed the use of the point dipole approximation (Eq. (3)) and Stokes expression (Eq. (10)) to estimate the time of drop–drop approach (t') when the initial inter-drop separation is large ($d_0 \geq a$). His expression for t' for uniform sized rigid drops is given as

$$t' = \frac{8}{15} \frac{\mu_m}{\varepsilon_m E_0^2} \left[\left(\frac{s_0}{2a} \right)^5 - 1 \right]. \quad (13)$$

Considering the flow circulations in and around the drop and validity of point dipole approximation only at separations $d_0 \geq a$, Eq. (11) can be used along with Eq. (3) to calculate the time of drop motion between separations $s = s_0$ and $s = 3a$. The resulting expression for time (t_1) for uniform size drops is,

$$t_1 = \frac{81}{20} \frac{\mu_m c}{\varepsilon_m E_0^2} \left[\left(\frac{s_0}{3a} \right)^5 - 1 \right] \quad (14)$$

At the lower drop–drop separations ($s < a$), the resistance to squeezing of medium fluid in thin film plays a vital role. When the viscosity ratio $\lambda \ll 1$, the thinning of the film induces convection rings inside the drops. Film thinning between undeformed drops and electrostatic pressure distribution ($=\varepsilon_m E^2/2$) near the drop interface result in the viscous force proportional to the product $\mu_m a v$ ([Atten, 2012](#)). Thus, the time of film drainage until contact of two drops is given as ([Raisin et al., 2010; Raisin, 2011; Atten, 2012](#)),

$$t_2 \approx \frac{1}{B} \frac{\mu_m}{\varepsilon_m E_0^2} \left(\frac{s_0}{a} \right)^{1.7}, \quad (15)$$

where B is a nondimensional constant. Time for drop–drop contact (t') in an electric field can be more accurately estimated as ($t_1 + t_2$) than by using Eq. (13).

2.4. Thin film breakup

The approaching drops contain a film of the medium fluid between their leading faces. As drops move closer, the thickness of the film continuously reduces by squeezing at the plateau border. On further reduction in the film thickness below 1000 Å, molecular forces start playing role. The attractive van der Waals force helps to reduce the thickness while the double-layer repulsion tries to push drops apart. The film attains a metastable state when the plateau border suction, van der Waals attraction and double-layer repulsion do balance each other. Instability can set in due to the thermal/mechanical shocks or the presence of impurities at the interface which results in to the breakup of the film separating the two drops.

When two drops are within a critical distance from each other, the microscopically thin film separating them can rupture rapidly, followed by the drop–drop coalescence. A critical thickness of film rupture is given by the expression ([Chesters, 1991](#)), $t_c = (Aa/8\pi\gamma)^{1/3}$, where A is the Hamaker constant. A number of mechanisms for the film rupture and subsequent merging of two drops have been proposed in the literature. One of the hypotheses suggests that the film breaks when the electric field across the film attains the dielectric breakdown strength of the medium phase ([Pearce, 1953](#)). Similarly,

[Sartor \(1954\)](#) and [Allan and Mason \(1962\)](#) suggested the spark discharge as a cause of the film breakup. These hypotheses of film breakup were proved wrong by [Priest et al. \(2006\)](#) by their study of selective coalescence of drops in a microfluidics channel. From the analysis of the experimental data it was proved that, not the dielectric breakdown but the electric field-induced dynamical instability of the water–oil interface drives the coalescence. Another hypothesis ([Fordedal et al., 1996](#)) states that when the applied electric field is high, the ions in the dispersed phase are pulled through the interface. The ruptured interface leads the coalescence of drops. According to [Berg et al.'s \(1963\)](#) hypothesis, the coalescence involves continuous making, breaking and rearrangement of the intermolecular bonds over the two interfaces in contact. The oppositely charged drops attract each other; but whether they coalesce or retreat back is dependent on the cone angle they make on contact. The expression for the critical cone angle at drop contact, which is a function of the electrocapillary number, was derived by [Bird et al. \(2009\)](#). Their surface energy model predicted the critical cone angle at drop contact as 30.8° which was close to the experimentally observed value. According to [Jung and Kang \(2009\)](#) the strength of the electrical force and the surface tension force determine the coalescence. When the electrical force is weaker than the surface tension force, drops can coalesce; whereas stronger electrical force may result in the retreat of drops after their contact. However, present theories fail to convincingly explain the mechanism of thin film breakup.

3. Critical conditions for electrocoalescence

The threshold conditions for a drop-pair in electric field, above which the shape distortion and coalescence can occur, have been suggested in literature. [Latham and Roxburgh \(1966\)](#) obtained an expression for the critical applied field for two closely spaced drops as $E_{\text{crit}} \propto d_0^{1.3}$ and the critical separation as $d_{\text{crit}} \approx 0.63d_0$. Due to the motion of the coalescing drops with time, there is no static solution for the deformed drops at very small separations. [Latham–Roxburgh's \(1966\)](#) assumption of full elongation of a drop was refuted by [Taylor \(1968\)](#) with an argument that the neighboring drops in a pair do not deform as a whole but only the nearest polar surfaces show deformation. Taylor, thus, did the instability study with two anchored drops in an electric field. With the assumptions of very small separation ($d_0 \ll a$) and the field being the potential difference ΔV divided by the center-to-center distance, Taylor could get the static solution for the small separations ([Taylor, 1968](#)). Taylor's analysis resulted into the critical parameters as, $d_{\text{crit}} \approx 0.5d_0$ and critical potential difference as, $\Delta V_{\text{crit}} \approx 0.38d_0 \sqrt{(\gamma/\varepsilon_m a)}$. The same relation for d_{crit} was obtained by [Atten and Aitken \(2007\)](#) using mathematical analysis and by [Reboud et al. \(2008\)](#) using numerical method and experiments. [Brazier-Smith \(1971\)](#) suggested that for every value of the separation ratio (d_0/a), there exists a critical value of $E \sqrt{(a/\gamma)}$, above which drops cannot remain stable. [Brazier-Smith et al. \(1971\)](#) numerically investigated the possibilities of the coalescence and possible shapes during interaction of two drops in an electric field. They concluded that for the separation ratio $d_0/a < 1.2$, drops deform, readily attract and eventually coalesce, whereas for $d_0/a > 1.2$, the facing surfaces deform, assume conical shape with an angle equal to the static Taylor cone angle and finally give rise to the jets of fine droplets. Such disintegration of a drop in the presence of other drops hampers the rate

of electrocoalescence. [Atten et al. \(2006\)](#) obtained expressions for the critical separation and the critical electric field similar to d_{crit} and ΔV_{crit} by Taylor. They also suggested that for a pair of drops in an electric field there exist a critical field for deformation for every separation value, given as,

$$\left(\frac{d_0}{a}\right)_{\text{crit}} = 8 \left(E_0 \sqrt{\frac{\epsilon_{ma}}{\gamma}} \right)^{1.22} \quad (16)$$

Based on Taylor's assumption of deformation only at the inner poles, [Atten \(2005\)](#) extended his work ([Atten et al., 2006](#)) to unequal size drops with $d_0/a \ll 1$. He theoretically investigated the critical conditions for the drop deformation and the coalescence of two drops (equal as well as unequal size) in electric field. The electrocoalescence occurs if, after the deformation, the facing poles come closer than 45% of their initial separation ([Atten, 2005](#)). Atten proposed a dimensionless number,

$$Be \cong \frac{\epsilon_m}{2} \left(\frac{\Delta V}{d_0} \right)^2 \frac{a}{\gamma}, \quad (17)$$

which is a ratio of the electrostatic and capillary forces. The critical conditions, Be_{crit} and d_{crit} , are logarithmic functions of the initial separation to drop-radius ratio, d_0/a . With the assumption of the quasistatic deformation of an approaching drop in a freely suspended as well as anchored drop-pair, [Atten and Aitken \(2010\)](#) obtained an analytical expression for the interfacial deformation. This analysis suggested that the shape of the approaching faces is slightly dependent on the separation ratio d_0/a ; however, the interfacial tension γ plays a major role in drop deformation. The numerical solution for two anchored drops by [Raisin et al. \(2008\)](#) suggested that a small pressure gradient exists inside the dispersed phase (near the inner face) due to high electrostatic stresses. This pressure gradient is responsible for deformation at inner poles of the coalescing drops.

In a coalescing emulsion, the orientation of two neighboring drops with respect to direction of applied field also affects the possibility of coalescence ([Friesen and Levine, 1992](#)). In an electric field, for two drops to attract each other, the potential energy of dipole moment ($M = P \cdot E$) must be negative, which can only be satisfied when $(3\cos^2\theta - 1) > 0$. Two drops cannot interact if $\arccos(1/\sqrt{3}) < \theta < \arccos(-1/\sqrt{3})$. For a drop-pair in electric field, coalescence can be possible when the angle made by the line joining the centers of the two drops with the direction of the applied field is such that, $54.71^\circ > \theta > 125.19^\circ$ ([Atten, 1993](#); [Eow and Ghadiri, 2003](#)).

4. Effect of turbulence and shear flow

An effective design of a water–oil separator strongly depends on the water fraction in the emulsion and the size of the droplets. The electrostatic force of attraction between opposite polarity poles of two dipoles is a short range force. In a dilute emulsion, inter-drop separation is large and therefore drops need to be moved closer to expedite the electrocoalescence. Inducing the shear flow or turbulence in the electrocoalescing emulsion increases the probability of drop–drop contact.

Other ways of increasing the probability of inter-drop contact is by applying shear ([Atten, 1993](#); [Urdahl et al., 1996, 2001](#)) or by generating flow by purging gas ([Thoroddsen and Takehara, 2000](#)). In an emulsion, the application of shear forces

or turbulence also resists the chain formation ([Chen et al., 1994](#)). However, very intense shear can induce drop breakup ([Galinat et al., 2005](#); [Mhatre and Thoakar, 2014](#)). [Fernandez \(2009\)](#) reported a coupled effect of shear and electrostatics for emulsions with less conducting dispersed phase. The effects of viscosity as well as electrical properties on the coalescence rate of two drops as well as in emulsions were studied numerically. Their direct numerical simulation results suggest that shear force causes tilting and breakup of chains. The commercial AC electrocoalescers before 1960s were designed to maintain the turbulent flow conditions in order to resist the chain formation. However, the modern electrocoalescers are designed to prevail laminar flows so as to facilitate the simultaneous coalescence and gravity settling of dispersed phase ([Noik et al., 2006](#)). The simultaneous effect of fluid flow and electric field can speed up the electrocoalescence. [Bailes and Kuipa \(2001\)](#) used sparged air bubbles to enhance the drop–drop interaction in a pulsed DC electric field. The rate of electrocoalescence was found to be high at the optimum frequency and increased with increasing the air flow rate up to a critical flow rate. However, excessive air flow in an emulsion can result in unfavorable effects as it does not allow drops to be in contact for enough time. [Urdahl et al. \(2001\)](#) reviewed correlations to estimate the maximum stable droplet diameter in laminar and turbulent flows. It was also showed that different mechanisms can contribute to the electrostatic coalescence, such as Brownian motion, sedimentation, laminar shear, turbulent shear or turbulent inertia.

[Melheim and Chiesa \(2006\)](#) used numerical simulations to show that turbulence enhances the electro-coalescence rate over a wide range of water cuts in the oil. The simultaneous effects of the electric and shear forces on the coalescence of two drops as well as emulsions consisting of less conducting drops in the more conducting medium were studied by [Fernandez \(2009\)](#) using direct numerical simulations.

5. Chain formation

Chain formation has been considered as one of the major retarding factors in electrocoalescence ([Fig. 2](#)). The stable chains not only reduce the rate of coalescence but also can extend and bridge the electrodes, leading to short circuit. Chain formation in electrocoalescing emulsions has been attributed to the presence of impurities which alters the properties of an interface. In a crude oil the presence of components such as asphaltenes and resins stabilize the drop interface ([Hannisdal et al., 2006](#)) and hinder the film thinning, leading to chain formation ([Taylor, 1988](#); [Mohammed et al., 1993](#)). The presence of asphaltenes is also considered responsible for increasing conductivity of the crude oil ([Lesaint et al., 2010](#)).

In one of the early scientific works in electrocoalescence, [Pearce \(1953\)](#) studied a water-in-oil emulsion under the action of uniform and concentric electric field and recorded some interesting observations. In DC electric fields droplet chains form in the direction of maximum field and they do not always start or end at the electrodes. Pearce argued that neither electrophoretic and dielectrophoretic force nor dipole interactions were responsible for the chain formation. In all these cases droplets would have moved to the electrodes; which was not observed. The reason, was attributed to the potential difference between two drops in contact. The film separating drops in a chain breaks when the field between interfaces in

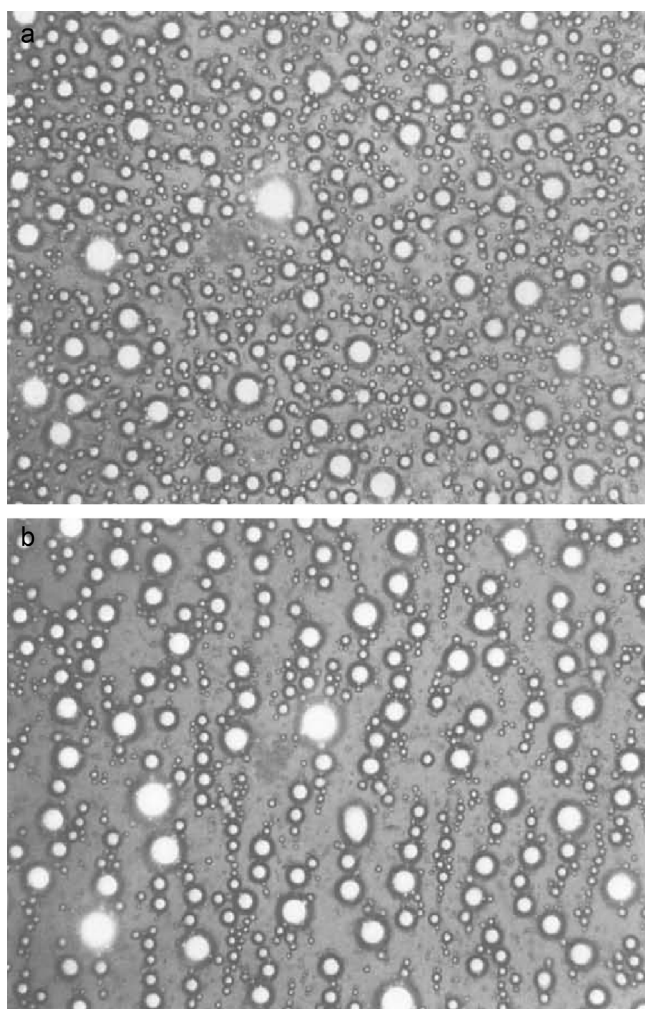


Fig. 2 – Chain formation in 20% water-in-Buchan crude oil emulsion at 50 Hz and 1.6 kV. Photomicrographs showing emulsion at; (a) 0 s and (b) 22 s (after Chen et al., 1994).

contact attains dielectric breakdown limit. In another work, Bezemer and Croes (1955) used a similar experimental system to Pearce's experiment (Pearce, 1953) but with larger electrodes separation and very dilute emulsions. Contrary to Pearce's observation, they found that droplets move in the direction of the maximum field strength. They also reported other new observations: large droplets move faster than the smaller ones, velocity of the droplets increase on increasing the electric field, droplets collect near the high field electrode and form radial projections, etc. These observations confirmed the role of dielectrophoresis as well as electrophoresis in the electrocoalescence and chain formation. Later Galvin (1986) claimed that the electrophoresis does not play any role in the coalescence as two drops move in opposite directions in order to contact each other. However, the presence of surfactants or other impurities can impart charge at the water–oil interface. Howe and Pearce (1955) used similar electrode systems as in references Pearce (1953) and Bezemer and Croes (1955) to electrocoalesce water in very viscous tar and observed that the carbon particles in the tar inhibited the water droplets from coalescing.

Chain formation is an undesirable effect in electrocoalescence and has been addressed in a number of studies. Nevertheless, a convincing mechanism for chain formation is not available in the literature. However, there exist studies suggesting methods to avoid the chain formation. Bailes

and Larkai (1981) first used the pulsed DC electric fields for coalescence of a water-in-oil emulsion. They found that for the pulsed DC electric field in frequency range between 0.5 and 60 Hz, 8 Hz gave optimum coalescence rate. Based on Bailes and Larkai's (1981) experimental setup, Midtgard (2009, 2012) subsequently proposed the electrostatic field theory as well as the circuit analysis theory and suggested the optimum pulsed field parameters which did not result in the chain formation. Application of the pulsed DC fields has been proposed as a solution to avoid the chain formation (Mohammed et al., 1993; Eow et al., 2002). In the presence of a pulsed DC field, coalescence can be observed only during rising and falling edges while no coalescence occurs in the middle of the pulse width (Taylor, 1996). Like an alternate current, a pulsed DC field exhibits an optimum frequency and it is dependent on the properties of the crude oil and electrodes aspects.

The AC electric field has an advantage over the DC field because of its abilities to suppress hydrodynamic flows in an emulsion. Moreover, the AC field above a critical frequency does not result in the chain formation and suppresses the subsequent short-circuiting of the electrodes (Chen et al., 1994). High frequency AC field has been suggested as a solution to avoid the chain formation. Chen et al. (1994) studied the chain formation and coalescence in AC electric field both experimentally as well as using molecular dynamics simulations. They also investigated the effect of stabilizing reagents in crude oil, such as asphaltenes and resins, on the rate of coalescence. They attributed the chain formation to the induced dipoles in the electric field and noted that the high frequency AC field resisted the chain formation in the electrocoalescing emulsions.

In the AC electric field, finding an optimum frequency (f_0) of coalescence is an important aspect of investigation (Zhang et al., 2011). In the electrocoalescence literature there is no unanimity on the value of such an optimum frequency. Apparently f_0 is affected by many factors, including physical and electrical properties, percentage of dispersed phase, droplets size, polydispersity, etc. Ingebrigtsen et al. (2005) observed that for a water-in-oil emulsion, at a low frequency $f < 100$ Hz the electrophoretic force induced fluid motion. The resulting advective flows reduced the coalescence efficiency. Contrary to previous observations, Holto et al. (2009) asserted that the coalescence at low frequency does not allow chain formation. At higher frequencies, electrophoretic movement of a charged drop covers a short distance during half cycle, thereby preventing large-scale motion and lowering the frequency of contact with neighboring drops (Holto et al., 2009).

6. Partial coalescence

When a drop, moving through another immiscible fluid, hits the interface of its homo-phase it can either merge completely with the bulk fluid or partially to form smaller drop(s). The latter phenomenon, called partial coalescence, can be observed both in the absence of external forces (Charles and Mason, 1960a, 1960b; Honey and Kavehpour, 2006; Blanchette and Bigioni, 2006) or in the presence of strong field for large drops. The resulting drops are termed secondary droplets. Sometimes it also exhibits the cascade of the hopping secondary drops at the interface and the simultaneous formation of finer daughter droplets (Charles and Mason, 1960b; Honey and Kavehpour, 2006; Thoroddsen and Takehara, 2000). Ohnesorge number (Oh) has been used as the criterion for the occurrence of partial coalescence (Aryafar and Kavehpour,

2006). Ohnesorge number, $Oh = \mu_d \sqrt{a \rho_d \gamma}$, is the ratio of the viscous to the inertial and surface tension forces. In the absence of external forces, a system with large Oh , exhibits complete coalescence (Aryafar and Kavehpour, 2009; Ray et al., 2010). Higher value of the viscous force (Stokes flow regime, $Oh \gg 1$) retards the secondary drop formation (Aryafar and Kavehpour, 2006). The electrocoalescence of an aqueous drop or organic drop with its homophase was studied by Dong et al. (2002). They reported that the coalescence time decreased with increasing the applied electric field for aqueous drops; however, in the case of organic drops, the coalescence time is dependent on polarity of the field. They attributed the polarity dependence of organic drop-interface coalescence to the electric double layer at interface in aqueous phase.

Partial coalescence in an electrocoalescing emulsion is considered as one of the most undesired effects. The presence of the high electric field expedites the drop-interface coalescence but it can also induce partial coalescence. Minute droplets are generated as a result of elongation and breakup of the neck between two coalescing drops or between a drop and an interface which leads to partial coalescence. The secondary droplets are much smaller than the primary droplets of an emulsion. The separation of such minute droplets from an emulsion makes the electrocoalescence process more difficult. Although partial coalescence in the absence of electric field has been studied extensively using experimental and theoretical techniques; there are very few studies available in literature on the phenomenon in the presence of the electric field (Allan and Mason, 1961; Aryafar and Kavehpour, 2007; Mousavichoubeh et al., 2011a, 2011b).

The partial coalescence of a drop at a flat interface in the presence of electric field was first reported by Allan and Mason (1961). Aryafar and Kavehpour (2007) reported that the film between a drop and an interface can become more perturbed in the presence of a DC electric field. For a system with constant Oh , the volume of secondary drop in partial coalescence can be more in the presence of electric field compared to that in the absence of electric field. Interestingly at bulk phase high viscosities, drops show complete coalescence in the absence of electric field but applying a strong field results in emergence of secondary drops (Aryafar and Kavehpour, 2007). Like in drop-drop electrocoalescence, initial time of approach between drop and the interface is a function of electrostatic force of attraction. After the film between the drop and interface attains a critical thickness, the film-drainage becomes rate limiting (Lukyanets and Kavehpour, 2008). Lukyanets and Kavehpour (2008) concluded that the drop-interface coalescence can be collectively affected by the magnitude as well as frequency of the applied electric field. The high electric field strength at the point of coalescence can induce local instability at the interface. The competition between the electrostatic stresses and gravitational force at the interface results in to a column extending towards the drop. The critical field of interfacial instability is given as (Taylor and McEwan, 1965),

$$E_{\text{crit, int}} \approx (\Delta \rho g \gamma)^{1/4} \sqrt{\frac{2}{\epsilon_m}}. \quad (18)$$

In drop-interface coalescence studies, the initial drop-velocity and release height above an interface significantly affect the partial coalescence (Aryafar and Kavehpour, 2009). These effects need to be nullified in order to understand the electrohydrodynamics of the drop-interface coalescence. That

can be achieved by applying the electric field at the onset of coalescence. Aryafar and Kavehpour (2009) with their experimental work showed that in the Stokes flow regime if the applied electric field is high, a secondary droplet undergoes breakup with a jet at its top surface. The jet eventually breaks in the surrounding dielectric medium as a stream of fine droplets. Strikingly, the jet exhibits whipping instability as the ternary droplets are highly charged. The time of coalescence remains unaffected by varying the applied electric field; however, a coalescing drop can stretch more if E_0 is high (Aryafar and Kavehpour, 2009). Same group experimentally demonstrated that the high strength DC electric field can give a semistable jet emerged from the trailing end of the drop, coalescing into interface (Aryafar and Kavehpour, 2010). Such a jet can give the whipping electrospray depending on the strength of the field. The drop-interface interaction can be analogized with a metal sphere held near a flat liquid-liquid interface (Atten et al., 2005; Reboud et al., 2008; Atten et al., 2008). In such a system the interface rises at the axis of symmetry and the height of the liquid column is governed by the applied potential and the separation between sphere and the interface (Reboud et al., 2008).

Mousavichoubeh et al. (2011a) studied the partial coalescence and expressed the phenomenon in terms of electrical clamping mechanism (Ghadiri et al., 2006). The electric current within a narrow path between nearest faces of a drop and an interface causes a compressive force squeezing the medium fluid out and making microscopic film unstable. A hole formed in the film acts as a conduit through which the drop fluid is pumped into the bulk fluid due to the surface tension force (Mousavichoubeh et al., 2011a; Honey and Kavehpour, 2006). At the same time the drop after contacting the interface acquires the same polarity charge and start experiencing Coulombic repulsion. The strength of the Coulombic repulsion and rate of the fluid pumping decides the occurrence of partial coalescence. In partial coalescence, when the applied electric field is high, the necking occurs faster and the volume of the resultant secondary drop can be larger. Also, the length of a tail emerging from a secondary drop can be longer and it can further break into finer progeny droplets (Fig. 3q and r). It was also observed that the volume of the secondary droplet was proportional to size of the primary drop as well as the distance between the interface and the point of drop-injection.

In another study Mousavichoubeh et al. (2011b) investigated the effect of the interfacial tension γ and applied electric field on the volume of the secondary droplet. Since the presence of surface active impurities in the crude oil stabilizes the water-oil interface, investigating of effect of γ in partial coalescence is important. The presence of the surfactants lowers the interfacial tension, resulting in the tip-streaming on both polar ends. This gives rise to very tiny secondary droplets of order $1 \mu\text{m}$. Furthermore, the lower value of γ reduces the critical electric field (E_{crit}), above which the secondary drops become unstable. Mousavichoubeh et al. (2011b) obtained a new dimensionless number, WO, by coupling the Weber number ($We = 2a\epsilon_m E_0^2 / \gamma$) and Ohnesorge number (Oh) to express the tendency of partial coalescence. The number WO is defined as,

$$WO = We \times Oh = 2\epsilon_m E_0^2 \mu_d \frac{\sqrt{a}}{\gamma^{1.5} \sqrt{\rho_d}}. \quad (19)$$

It was observed that the volume of the secondary droplets increased with the WO number for a wide range of interfacial tensions and electric fields.

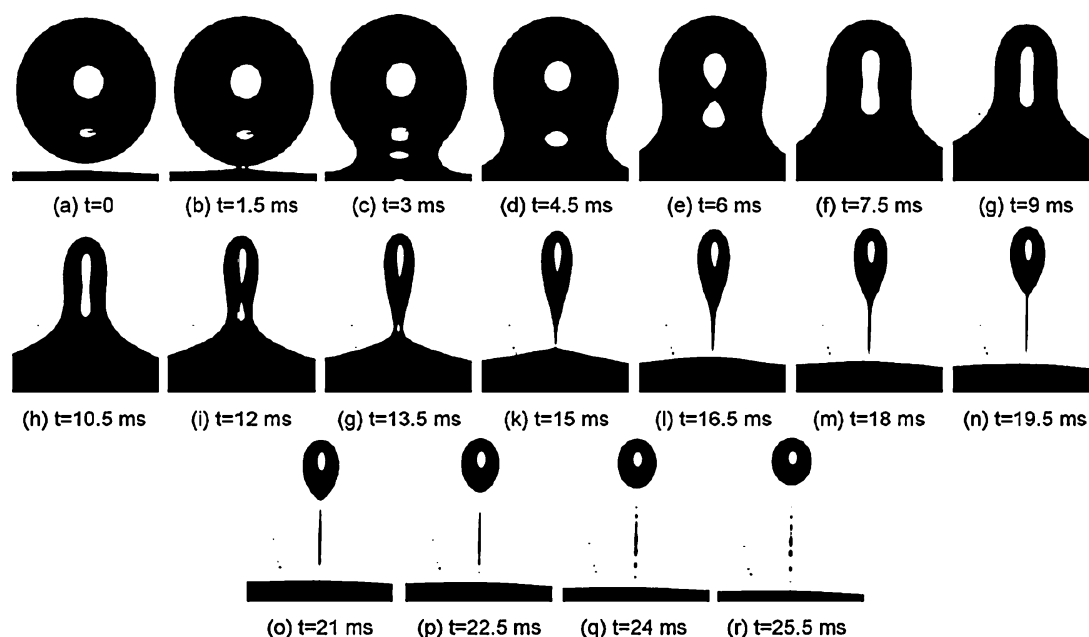


Fig. 3 – Partial coalescence of a droplet of size $1196 \pm 4 \mu\text{m}$ in the presence of electric field 181 V/mm (after Mousavichoubeh et al. (2011a)).

When a drop with inherent charges comes into the contact with an interface of the same fluid but with opposite polarity, the two should readily coalesce. However, two charged interfaces with opposite polarities attract to each other but do not always coalesce. Ristenpart et al. (2009) reported that under a very high electric field two oppositely charged bodies repel after contact. Hamlin et al. (2012) with their experimental results concluded that in addition to the applied electric field, the drop-interface coalescence is also governed by ionic conductivity. There exists a critical ionic conductivity, below which partial coalescence can be observed, whereas above it, the drop bounces against the interface. Surprisingly, ionic conductivity does not determine the charge and size of the secondary droplets. Hamlin et al. (2012) also gave an explanation of the generation and charging of the secondary drops. In the partial coalescence an interface does not impart charge to the secondary drop but the charge comes from the induced charge on the primary drop prior to coalescence. On contacting the interface, the leading edge (which has charge of polarity opposite to that of interface) loses its charge by convection, whereas, the top edge of drop which has residual dipolar charges now acts as a charged entity and experiences electrophoretic pull in the opposite direction leading to formation of secondary droplet. Atten (2012) proposed a similar reason for the non-coalescence behavior of two drops in a high electric field.

7. Effects of operating parameters and fluid properties

The majority of the early studies in electrocoalescence were motivated by the use in crude oil demulsification. Since the crude oils from different oil fields are black in color, visualization and microscopy of the process is difficult. As a result, most of the experiments used emulsions in mineral oils such as paraffin oil, Nynas Nytro 10 \times , Model oil, etc. Near infra-red (NIR) microscopes have also been employed in experiments with crude oils to overcome low refractive index problem (Lesaint et al., 2009).

The different operating parameters that govern the rate of electrocoalescence are the magnitude, waveform and frequency of the applied electric field, coalescer geometry, electrode configuration, flow or turbulence, etc. (Hano et al., 1988; Lee et al., 2001). Also, the emulsion properties such as interfacial tension, density difference, viscosities, conductivities and permittivities of fluids, drop size, polydispersity, percentage of dispersed phase play significant roles in electrocoalescence (Lee et al., 2001; Al-Sabagh et al., 2011; Hosseini and Shahavi, 2012; Zhang et al., 2012).

The effect of various parameters on coalescence efficiency in continuous process in an AC electric field was studied by Kim et al. (2002). Apart from the electric field strength, the effect of parameters such as frequency of applied field, demulsifier concentration, temperature, and contact time, on the separation rate was studied. They attributed the increase in the coalescence rate with temperature to the reduction in the viscosity of the continuous phase.

The DC electric field is effective in promoting coalescence since the driving force is maintained continuously high. However, with high water content the risk of electric breakdown is high. Short-circuiting can be avoided by insulating the electrodes. However, in the presence of a DC electric field, all ions and charge carriers quickly move to the insulating barrier. As a result the voltage drop between the metallic electrodes takes place mainly across the solid insulation. Pulsed DC or high frequency AC fields are therefore used to avoid this effect. In these cases, the frequency of the applied field should be high enough so that the duration of one half period of the voltage is short compared to the time-scale of charge distribution in the drop (Lundgaard et al., 2002, 2006). DC electric field is considered suitable for emulsions containing tiny and sparsely spaced dispersed phase; whereas AC field benefits in handling the emulsions with high water content as it resists the short-circuiting of an electric field (Noik et al., 2006). The combined AC/DC field allows handling the emulsions with high water content at the high coalescence rate.

Electrorheology of water-in-model oil emulsion under the action of AC electric field was studied by Lesaint et al. (2009). Along with viscosity effect, they also investigated the influence of other parameters including frequency, time of field application, temperature, etc. The viscosity of the emulsion decreases with time of field application as well as with temperature (within the range of 20–60 °C). The efficiency of coalescence is found increasing with frequency in the range of 50–5000 Hz. Type of the waveform of the applied AC field can also be a significant factor affecting the rate of coalescence. Among three waveforms used, Lesaint et al. (2009) observed that the coalescence efficiency was higher with square waves than with the sinusoidal and triangular waves. The waveform with more area of the form results in more coalescence. However, contrary results were reported by Mousavi et al. (2014) who found that the triangular waves were more effective in drop-interface coalescence than the sinusoidal and triangular waves. They observed that the volume ratio of secondary and primary drops was lower with triangular waves than that with sinusoidal and triangular waves. In the presence of dissolved salts in water, the rate of coalescence in AC field is characterized by a sharp threshold frequency (f_{th}) (Szymborski et al., 2011). At $f > f_{th}$ there is no coalescence, whereas at $f < f_{th}$ coalescence depends on magnitude as well as frequency of the applied field. Lee et al. (2001) investigated the performance of an electrocoalescer under AC and pulsed DC fields. They concluded that the AC field results in the better coalescence rate than the pulsed DC which was contrary to the earlier findings (Waterman, 1965; Hsu and Li, 1985; Bailes and Larkai, 1981; Wakeman, 1986; Figueroa and Wagner, 1997). The different parameters they examined were conductivity and viscosity of emulsions, electrical field (strength, wave forms and frequencies) and emulsion feed rate keeping the particle size distribution constant. They also reported the actual power consumption to demulsify crude oil under AC and pulsed DC fields in the frequency range 60–600 Hz. No optimum frequency was observed in this frequency range; however the coalescence rate was found increased on increasing the frequency (Lee et al., 2001).

In electrostatic demulsification the coalescence rate depends on stability of the water–oil interface. This is predominantly governed by interfacial tension and in turn by the presence of surfactants, impurities, etc. The characterization of crude oil samples for such impurities and their effects on emulsion stability have been studied using different techniques (Sjoblom et al., 1990; Sjoblom et al., 1992; Mingyuan et al., 1992; Fordedal et al., 1996). The presence of impurities in the crude oil alters the electrical as well as physical properties like viscosity, interfacial tension, elasticity, etc. (Berg et al., 2010). In the batch electrocoalescence, the concentration of impurities in crude oil can increase as the coalescence progresses and emulsion can eventually be more stable (Noik et al., 2002).

Asphaltenes are a major contributor to the conductivity of the crude oil. However, not the amount of asphaltenes but its aggregation state decides the conductivity of crude oil (Lesaint et al., 2010). From the conductivity (σ_m) versus viscosity (μ_m) behavior of diluted crude oil samples, Lesaint et al. (2010) concluded that $\sigma_m \propto 1/\mu_m$, as higher viscosity resists the mobility of the charge carriers. Fjeldly et al. (2008) investigated the electrocoalescence in heavy and medium crude oil as well as multistage separation of three phase emulsion in an industrial unit. High percentage of water in crude oil leads to short circuiting the electrodes. Use of coated electrodes not only helps

to overcome this problem but also greatly enhances the water separation and improves the produced water quality (Fjeldly et al., 2008). From the analysis of industrial and pilot plant data for the different crude oil samples, Suemar et al. (2012) concluded that the separation efficiency is dependent upon applied field as well as on the time of residence in the coalescer.

The effect of physical and electrical properties on electrocoalescence was numerically investigated by Lin et al. (2012) using a coupled phase field and leaky dielectric model. Their analysis showed that, when the outer fluid is more viscous than the drop phase, the time scale for coalescence is longer as it is harder to drain the microscopic film between the drops. On reducing the interfacial tension, the bigger drops formed after the coalescence can be susceptible to further breakup. The deformed faces of the coalescing drops in the absence of electric field resist the film drainage. Since the lesser interfacial tension (γ) causes more deformation, in the absence of electric field, decreasing γ slows down the coalescence. However, in electrocoalescence, increasing γ enhances coalescence rate (Dong et al., 2002). Recently, Rayat and Feyzi (2012) used Barker–Henderson perturbation theory to propose a thermodynamic model for the prediction of critical conditions for breaking an emulsion. They also studied the role of deformation on the demulsification and the different parameters, such as temperature, size, interfacial tension, viscosity, Gibbs elasticity, etc., affecting the deformation of drops.

8. Electrocoalescence on dielectric surfaces

Distortion and coalescence of water drops on insulation surfaces in high electric power applications is considered troublesome. The enhanced field at the poles of the deformed drops leads to the ignition of corona discharges which can adversely affect the insulation quality (Ndoumbe et al., 2012). Moreover, drops in vicinity can coalesce and the resulting bigger drop can bridge the electrodes. Such kind of behavior of water drops resting on the dielectric surface was investigated by Ndoumbe et al. (2012). The coalescence of sessile drops on insulation surface under the action of DC electric field was found to be dependent on the applied electric field, volume of drops and position of drop in the electric field. The coalescence rate on hydrophobic insulator surface increases with the field strength as well as volume of drops. Orientation of drops in the electric field determines the probability of coalescence. If the line joining centers of two drops make an approximate angle 45° with the direction of the applied field, the chances of drop–drop attraction are highest. However, if the angle is around 90°, drops hardly respond to the electric field.

9. Effects of drop size and polydispersity

An emulsion with coarser drops breaks faster (Pearce, 1953; Bezemer and Croes, 1955; Eow et al., 2003; Ingebrigtsen et al., 2005). Moreover, the presences of few big drops also enhance the demulsification as minute drops are easily sucked into the interface of a large drop. The critical field (E_{crit}) for a single conductor drop above which it becomes unstable (Taylor, 1964) is,

$$E_{crit} = 0.648 \sqrt{\frac{\gamma}{2a\epsilon_m}} \quad (20)$$

After the merging of two drops, if the resultant drop has size above the critical limit, it can become unstable and break

(Eow et al., 2003). The instability can be asymmetric due to the inhomogeneity of field in the emulsion (Holto et al., 2009). Such instabilities make the water separation more complicated as they introduce very finer droplets than the original drops in the emulsion. The deformation of drops affects the coalescence rate. This effect is observed more in emulsions with smaller droplets than emulsions of coarser drops (Rayat and Feyzi, 2012) although the deformation can be more in the latter case.

Two dissimilar size drops in an electric field do not coalesce at the same speed as the equal size drops. The electrostatic forces (Eqs. (1) and (2)) as well as mechanical forces (Eqs. (10)–(12)) are functions of the drop size. Emulsions are essentially polydispersed in nature. Therefore, in the electrocoalescer design, the observations for the coalescence of uniform size drops cannot necessarily be true for actual emulsions.

10. Non-uniformity of the applied field

Electric fields employed in the earlier electrocoalescence studies were predominantly of uniform kind. The effectiveness of the non-uniform fields in electrocoalescence has not been explored very much. A concentric non-uniform electric field was first used for emulsion breaking by Pearce (1953). In another study, Eow and Ghadiri (2003) suggested five electrode designs for coalescence of flowing drops and investigated the effect of direction of the applied field on two drops coalescence. They also studied the effect of the field and frequency on oscillations of a moving drop in their electrode systems (Eow and Ghadiri, 2003). Noik et al. (2002), working at pilot plant-scale, designed a compact coalescer with two annular cylinders as electrodes and having a centrifugal flow of emulsion. They studied effects of electric field strength as well as flow on the rate of electrocoalescence and found that the shear forces or turbulence due to the hydrodynamic conditions can induce droplet break-up or can disturb the dipole–dipole interaction. Hosseini and Shahavi (2012) used concentric cylindrical electrodes to separate tiny sunflower oil droplets from water.

11. Electrocoalescence in leaky dielectric emulsions

The use of oil-in-oil emulsions is getting prominence in many industrial applications particularly in emerging areas like polymer blends and electrorheological (ER) fluids (Block and Kelly, 1988; Zukoski, 1993; Aida et al., 2010). In polymer blends, two or more immiscible polymers are mixed together to get a resulting product with improved properties. The size control of the dispersed phase plays a vital role in defining the qualities of the final product. Electrocoalescence in polymer blends can be used to control the drop size and dispersity of the dispersed polymer. ER fluid is an electro-magneto responsive fluid which exhibits shear thinning or thickening of suspension under the influence of electric field (Block and Kelly, 1988; Zukoski, 1993). These fluids have the potential to be developed for active control devices such as dampers, shock absorbers, clutches, brakes, etc. The polydispersity and spatial distribution of the particles govern the rheological properties of the suspension in significant manner. Application of the electric field helps to effectively control the dispersity, segregation and merging of dispersed phase.

The electrocoalescence of leaky dielectric (LD) fluids, i.e. those with very low but finite electrical conductivity, such as vegetable and mineral oils, organic solvents and perfectly dielectric (PD) systems was first addressed by Baygent et al. (1998) using the Boundary Element Method (BEM). The shapes, motion and flows in and around the interface were studied for the equal size drops in a uniform electric field. In a PD system, the drop velocity approaches ($\propto 1/s^4$) when the deformation is small; whereas for a LD drop pair, the velocity scales as $(s/a)^{-2}$ for larger inter-drop separation. Attraction or repulsion of LD drops is decided by electrical properties such as conductivity and permittivity. When $(\sigma_d/\sigma_m) < (\epsilon_d/\epsilon_m)$, drops attract to each other; while they repel when $(\sigma_d/\sigma_m) > (\epsilon_d/\epsilon_m)$. Adamiak (2001) conducted a similar study with the Finite Difference Method and the BEM, with primary focus on interdependence of deformation and drop position in PD and perfectly conducting (PC) systems. He observed that the point-point model did not give the accurate estimate of electrostatic force when inter-drop separation was small; whereas, sphere-sphere model was accurate at low E_0 and large s . Lin et al. (2012) used phase field model to investigate the effects of physical and electrical properties on the shapes of the coalescing drops in LD systems. The shape of the electrocoalescing drops is governed by permittivities of fluids. A LD drop in a pair assumes oblate shape when $(\epsilon_d/\epsilon_m) > 1$ and deformation increases on increasing the ratio, ϵ_d/ϵ_m . When the medium phase viscosity (μ_m) is low, inertia assists the film drainage while higher magnitude of μ_m results in the non-trivial shapes of the inner faces.

Klingenberg et al. (1991) studied the rheological properties of the electrorheological (ER) suspensions using experiments and molecular dynamics simulations. The rheological response depends on the concentration as well as dielectric constants of the fluids. “Hierarchical Model” proposed by Aida et al. (2010) to derive the coalescence rate in immiscible polymer blends suggested that the rate increased with the electric field and the volume fraction of the dispersed phase.

12. Electrocoalescence in microfluidics devices

Coupling of electrostatics with microfluidics is used advantageously in technological applications (Zhao and Yang, 2013; Chabert et al., 2005a, 2005b) where the system; (a) can be operated with electricity, (b) has good control on operation, (c) no moving parts and therefore less maintenance, (d) high throughput screening in applications such as, premature chromosome condensation (PCC), (e) fluid velocities are independent of channel size and (f) can be fully automated. Electrocoalescence in microfluidics setups has been applied predominantly in bioassay as well as in microreactors where reactants in small amounts can be contacted with full control. In bio-applications such as bio-particles separation, microfluidics-based electrocoalescence helps to avoid the use of harsh chemicals and multiple washings (Chokkalingam et al., 2014). Different functions such as drop generation, motion, trapping, charging, sorting, coalescence and discharge can be performed on a single miniaturized platform which can be fabricated using lithographic techniques. Although there are other methods of drops fusion in microfluidics devices, the control on drop movements and their selective fusion can be achieved employing electric fields (Jose and Cubaud, 2012). A review article by Gu et al. (2011) discussed the breakup and coalescence in microfluidic devices applying the electric field.

They detailed different types of microchannel designs suitable for the passive merging (merging of unstabilized drops in the absence of external forces) as well as for the active merging (merging of stabilized drops by applying external forces). Electric fields are predominantly used in the active merging inside the microfluidic channels (Gu et al., 2011).

Electrodes can be embedded inside the microfluidics channels in such a way that neither dispersed phase nor carrier phase come in direct contact with electrodes. This aspect lowers the chances of cross-contamination in biological applications. Electric field helps drops to coalesce in a microfluidics channel where drop size is comparable to the channel size. The microfluidic channels are mostly fabricated in polydimethylsiloxane (PDMS) which is highly hydrophobic. In the microreactor applications the carrier oil phase wets the channel surfaces and prevents the contact of the dispersed phase with the channel walls (Link et al., 2006).

Chabert et al. (2005a) studied drop–drop coalescence in stationary phase as well as in flowing phase in microfluidics channel. They observed that in DC electric field, drops did not coalesce; instead they migrated to the electrode due to the acquired charge. Under an AC electric field, drops showed coalescence; however, velocity of their approach was constant due to the proximity of the electrodes. In an AC electric field drops in a channel exhibited shape oscillations. Above a critical frequency, a drop did not oscillate but it exhibited DC field-like elongation in the direction of the applied field. Chabert et al. (2005a) plotted critical magnitude versus frequency of the applied electric field to get the region of coalescence. Lower limit of the region is independent of frequency but the upper limit varies with frequency. The same group proposed a microfluidics system for continuous flow high-throughput polymerase chain reaction (PCR) (Chabert et al., 2005b). In the microreactor application the electric field can be used to charge the drops of reactants to opposite polarity. The applied electric field not only facilitate the precise control of motion of charged drops but also ease their coalescence in the presence of stabilizing impurities (Link et al., 2006).

Similar to the electrocoalescence in free space (Atten, 2012), in microfluidics channel, when the applied field is very high, the long and narrow bridge between drops can break and drops repel violently. The breakup of the bridge results in minute satellite droplet. The charge-exchange between drops induces the cycles of attraction and repulsion. Wang et al. (2010) used DC, non-uniform electric field to selectively trap and fuse the drops on microfluidics platform. The magnitude of applied voltage needed for trapping and coalescence increases on increasing the flow rate of carrier fluid. However, a carefully designed microfluidics channel can help to enhance coalescence even at low electric field (Niu et al., 2009).

13. Available technologies

The state of the art of electrostatic demulsification was earlier reviewed by Eow and Ghadiri (2002a,b). They also developed a novel and compact electrocoalescer to handle the flowing emulsions (Eow et al., 2002) (shown in Fig. 4). The separation of aqueous phase was based on drop–drop and drop-interface coalescence. The water layer that accumulated at the bottom of the device not only acted as an interface for the drop-interface coalescence but also acted as the ground electrode. Their results with the pulsed DC field indicated that for every

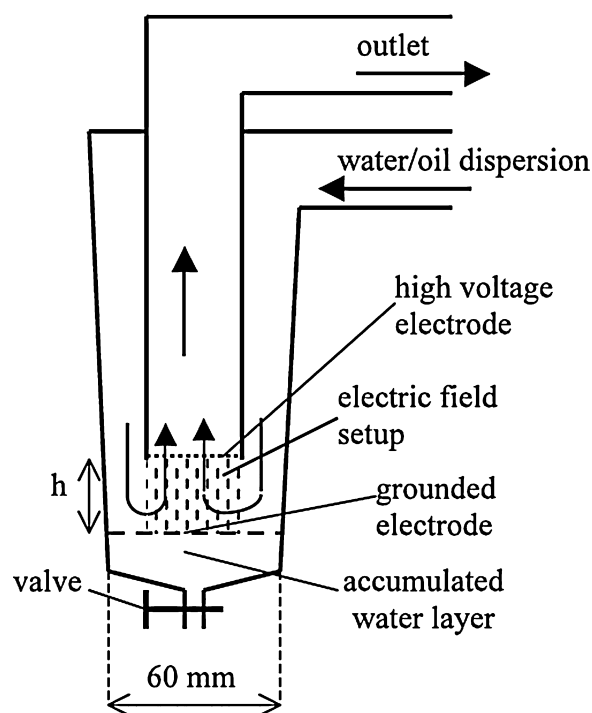


Fig. 4 – Schematic of electrocoalescer by Eow et al. (2002).

magnitude of applied potential, there exists an optimum frequency which gives maximum separation efficiency. They also found that separation efficiency increased with increasing the applied field and size of droplets. However, above certain applied field, the efficiency started diminishing due to short-circuiting and further breakup of drops. Using a force balance on a drop in an electric field, Eow et al. (2002) used a parameter I_p to describe the acceleration due to the applied electric field. I_p is defined as,

$$I_p = 1 + \frac{g \frac{(\rho_d - \rho_m)}{\rho_d} + \frac{9}{4} \frac{\epsilon_m d_d^3 E_0^2}{\rho_d L^4} - \frac{18\nu\mu_m}{\rho_d d_d^2}}{\frac{18\nu\mu_m}{\rho_d d_d^2}}, \quad (21)$$

where L is distance between drop and the ground electrode, d_d is diameter of drop, ν is velocity of the drop relative to the medium fluid, and ρ_d and ρ_m are the densities of drop phase and medium phase, respectively. I_p increases with applied electric field as well as the size of the dispersed phase.

Although not commercialized, additional novel electrocoalescers were designed by Eow and Ghadiri (2002a) and Eow (2002). The first design was based on a standard gravitational separator, commercially available to separate water drops from the oil, which was modified and equipped with electrodes (Fig. 4). This design has combined effects of the gravitational and electrically-induced forces to enhance the separation of dispersed drops by drop–drop as well as drop–interface coalescence. The accumulated aqueous phase at the bottom of the separator acts as a ground electrode and it also facilitates the drop–interface coalescence.

Larger throughput and smaller inlet drop sizes can be handled with Eow and Ghadiri's (2002) second design (Fig. 5b) which combines the effect of electrostatic forces and centrifugal motion of the liquid. It is actually a commercially available water separator for diesel but equipped with electrodes for application of an electric field. The water-in-oil dispersion enters tangentially in the upper part of the cylindrical section, forcing the denser aqueous drops to move to the cylinder wall

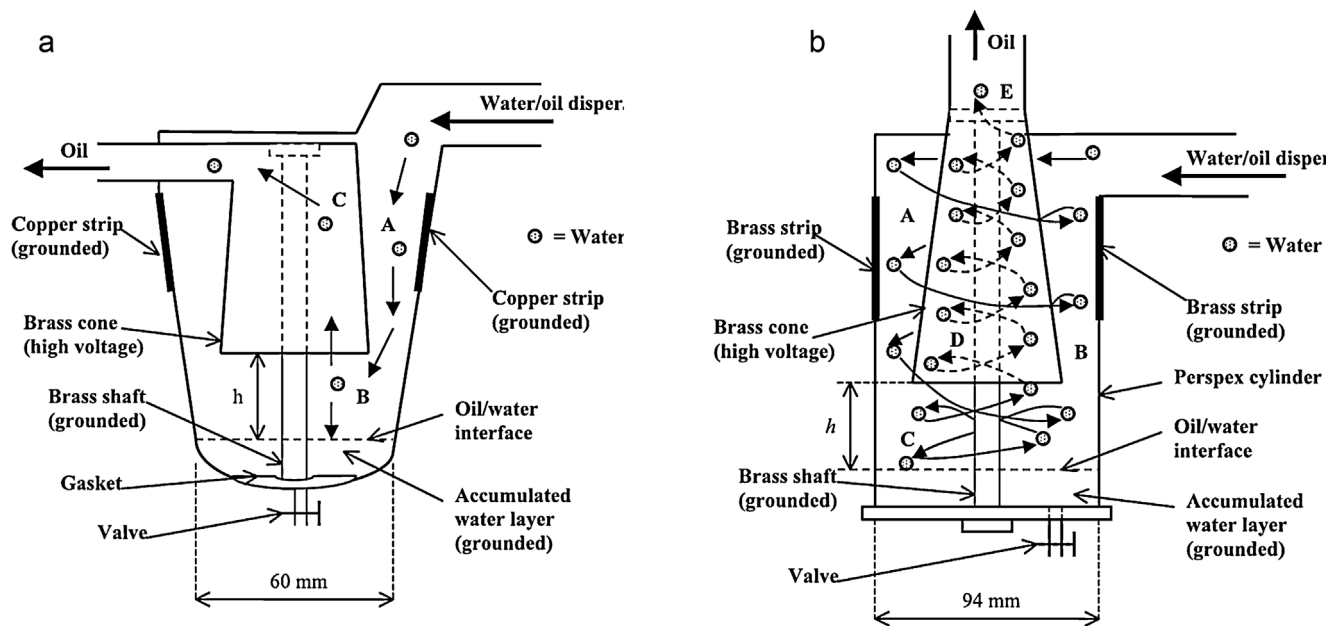


Fig. 5 – Electrocoalescers by Eow and Ghadiri (2002a) and Eow (2002). (a) Schematic diagram and movement of an aqueous drop in the gravitational electrocoalescer-separator. (b) Movement of aqueous drops in the centrifugal electrocoalescer-separator.

where they coalesce under the influence of the electric field. Coalescence also takes place at the water–oil interface at the bottom of the separator. In both designs, the height of the accumulated water layer at the bottom of the separator can be controlled by a valve to optimize the separation efficiency. In both devices, an optimum frequency was observed to exist with pulsed DC electric field.

Trapy and Noik (2007), Noik and Trapy (2004) proposed an electrostatic separator for effluents containing phases of different densities and electrical conductivities. The proposed device contains cylindrical electrodes placed along a common axis and a helical channel situated downstream of the coalescence section which separates the dispersed and medium phases.

In the early days, electrocoalescence devices in the crude oil industry were bulky; divided into a ‘treating space’ for the droplet growth and a ‘settling zone’ for phase separation (Urdahl et al., 2001). Due to the compelling need of fast and efficient water separation, new concepts were developed and various technologies are now available in the market. The survey of Less and Vilagines (2012) was focused on commercial technologies developed in recent years. All the recent technologies use insulated electrodes to prevent the short-circuiting during electrocoalescence.

Vessel Internal Electrostatic Coalescer (VIEC™) technology developed by Hamworthy (a Wartsila company) is claimed to be the first electrocoalescence technology which can be used in the inlet separators (WVIEC <http://www.wartsila.com/en/oil-separation/oil/viec>; Viec <http://www.hamworthy.com/products-systems/oil-gas/oil-separation-systems/viec-separation-technology/viec>). The benefits of VIEC™ include, it can process high viscosity oils, reduces chemical consumption, it can handle light as well as heavy crude oils. The electrodes used in VIEC are fully insulated therefore it can withstand 100% water or gas without short-circuiting. Aibel Vessel Internal Electrostatic Coalescer was tested by Less et al. (2008) and found to reduce the separation time to a quarter. Combination of chemical and electrostatic treatments showed significant improvement in the separation.

The electrodes used are isolated by molding in epoxy and therefore can be used to demulsify crude oils of different kind without short-circuiting.

Three electrocoalescence products are commercialized by Hamworthy: the Vessel Internal Electrostatic Coalescer (VIEC™), the High Temperature VIEC (HT VIEC™) and the Low Water Content Coalescer (VIEC LV™). The VIEC™ technology

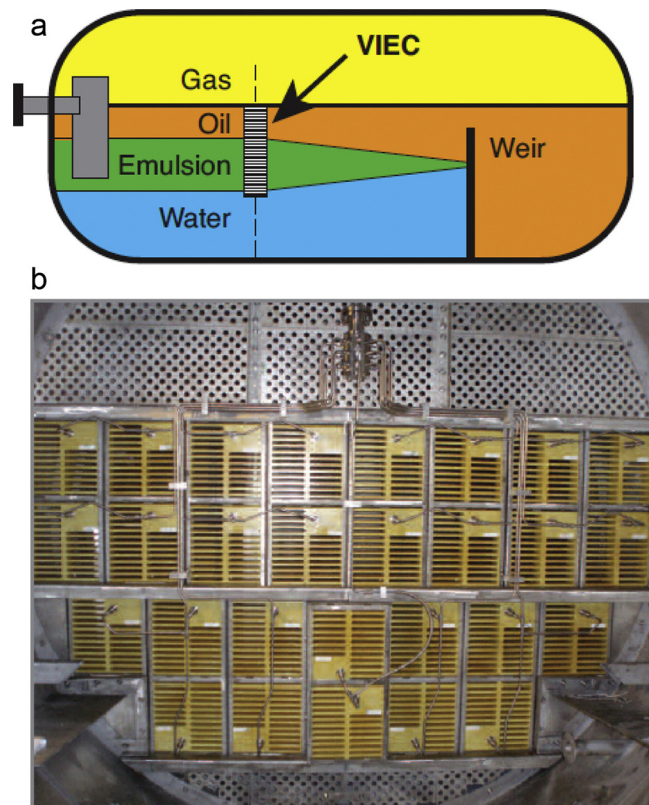


Fig. 6 – VIEC™ Technology (image source: <http://www.hamworthy.com>). (a) Schematic of VIEC™ electrocoalescence system. (b) Cross-sectional view of VIEC™ electrocoalescer.

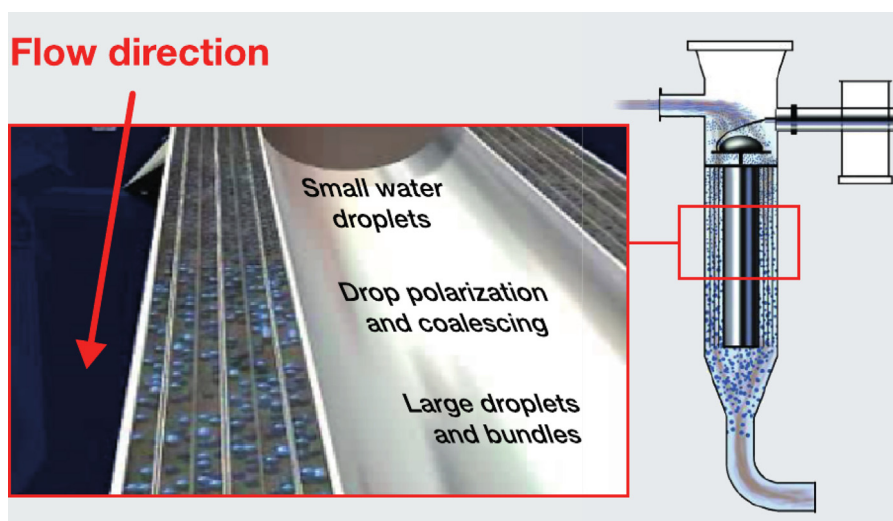


Fig. 7 – CEC™ electrocoalescers by Aker Solutions (Image source: <http://www.akersolutions.com>).

(Fig. 6) integrates a conventional upstream separation vessel and is built as a wall of the coalescer modules sitting vertically across the cross-section of the vessel. The HT VIEC™ has a similar structure but it is suitable for high temperature (150 °C) and high pressure (150 bar) operations. The VIEC LV™ technology makes use of dielectrophoretic force and is designed to operate downstream the VIEC™ or HT VIEC™ to meet the crude oil specification quality.

A compact electrocoalescer developed by FMC Technologies can be fitted into pipeline upstream of separator and claimed to give high separation efficiency with less power consumption. Vetco Aibel developed Low Water Content Coalescer (LOWACC) to be used downstream of VIEC. LOWACC technology enhances the heavy oil separation, improves produced water quality and it is suitable for subsea operations (Fjeldly et al., 2006).

Other commercial technologies involve Statoil patented Compact Electrostatic Coalescer (CEC™) technology (Fig. 7). Natco's Dual Polarity® electrocoalescer uses simultaneous AC and DC electric field. Recently, Natco patented an upgraded Dual Frequency® technology which uses a high base frequency optimized to limit the time of voltage decay.

Starting with the choice of the electric current, DC field is considered favorable when drops are sparsely distributed in an emulsion; whereas, AC fields are more suitable for emulsions with high percentage of dispersed phase. However, the optimum frequency of the applied AC field needs to be identified, which is governed by the emulsion properties. If the electrodes are insulated, the optimal frequency also depends on the insulation properties. A drop in an electric field undergoes disintegration when the electrocapillary number Ca_E exceeds a critical value (Ha and Yang, 1999; Mhatre and Thaokar, 2014; Karyappa et al., 2014), so large Ca_E should be avoided. The magnitude of the applied electric field in electrocoalescence should not exceed a critical value which may lead to the drop breakup and adversely affect the coalescence rate. Partial coalescence can be overcome by avoiding excessive electric field strength and using pulsed DC fields at sufficiently high frequency (Mousavi et al., 2014). Also the chain formation could be averted by the use of pulsed DC electric fields.

The probability of drop–drop contact can be increased by gas purging, shearing, agitation, etc., or internally induced electrohydrodynamic flows. The use of non-uniform electric

fields could generate electrohydrodynamic flows and also results in the dielectrophoretic segregation of the droplets. However, care needs to be taken when applying the strong flows as the shear stresses stimulate the drop breakup (Mhatre and Thaokar, 2014). Some relatively simple correlations to estimate the maximum stable droplet diameter under laminar and turbulent conditions are discussed by Urdahl et al., 2001.

Considering the various design elements for an efficient electro-coalescence, requirements for a new compact design are:

- reducing migration of droplets;
- promoting multiple electrocoalescence;
- immediate pushing away of the large droplets, mitigating short-circuiting.

14. Conclusions

Different aspects of electrostatic phase separation have been critically reviewed. Electrocoalescence of two drops involves three major stages; drop–drop approach, film drainage and thin film breakup. Accelerating every stage can make the coalescence faster. The characteristics of the applied field such as strength, frequency, and kind of field directly determine the electrostatic force of attraction. Other factors involved are electrical and physical properties of fluids, turbulence, water content, polydispersity, etc. There also exist some phenomena which adversely affect the coalescence process; examples are chain formation, partial coalescence, electrohydrodynamic drop breakup. Current developments in electrocoalescence research addressing all the above mentioned aspects are reviewed.

There are no dramatic findings in the field of electrocoalescence in the last 13 years after last review by Eow et al. (2001). However, new areas of application such as microfluidics have been developed. Research in oil-in-oil electrocoalescence is useful in applications involving polymer blends and electro-rheological fluids. New finding in partial coalescence could help to understand the phenomenon and mitigate its effects. Investigations concerning drop–drop interaction and critical conditions of coalescence will certainly help in enhancing the rate and in the design of more efficient electrocoalescers.

However, the present research in electrocoalescence lacks certain aspects. Resolving different unanswered questions and exploring new techniques can definitely help in making electrocoalescence faster, more efficient and in making compact electrocoalescers. One of such unanswered problems is the chain formation in an electrocoalescing emulsion. Although there are quite a few investigations on the chain formation, the mechanisms proposed so far are not satisfactory and convincing. There are no universal measures to avoid the chain formation. Applying AC or pulsed DC fields has been advised but finding the optimum frequency for variety of crude oils needs to be addressed.

There are very few studies investigating the prospects of non-uniformity of the applied electric field. Non-uniform fields help the dispersed phase to segregate in particular region of the field depending on polarizabilities of the fluids involved. This fact has been extensively used in bio-particle research. In electrocoalescence non-uniform electric field can be used to enhance the drop-drop approach. Furthermore, the flow induced due to dielectrophoresis resists the chain formation. However, if the flow convection in electrocoalescing emulsions are excessive, non-uniform field can adversely affect the coalescence rate. Optimization of the timescale of the fluid flow and that of coalescence can be the key in effective use of non-uniform electric fields.

The timescale of the drop-drop approach and film-drainage is much larger than that of the film breakup. There is substantial work in the literature on these two topics, although there is room for improvement by clever design of the electrode system to enhance these rates. Moreover, the understanding of the film instability and investigating possible ways to enhance it can significantly contribute to the coalescence rate. A deep insight in to the mechanism of film breakup can also be helpful to resolve the chain formation. The current understanding of the film-rupture mechanism is poor.

Dissolved impurities in crude oil are major factors in deciding electrocoalescence rate. Identifying different impurities, their extents in crude oils from different reservoirs and fluid properties altered by them is a difficult task. As mentioned by Berg et al. (2010), since each component of crude oil cannot be analyzed individually, different crude oils can be categorized into different chemical classes based on contribution to different physical and chemical properties. Understanding the effect of impurities on the interfacial stability of drops, drop approach and film drainage could help the design of a universal electrocoalescer and in tuning the operating parameters of electrocoalescence of the varying crude oils.

The understanding of microfluidics devices may help optimizing large scale electrocoalescence for example in crude oil demulsification. Electrocoalescence on the surface of an electrode or on the dielectric surfaces has not been explored in any study. Although there are some studies dealing with drop-interface coalescence, the idea of using Electro-Wetting on Dielectric (EWOD)-like applications in large scale electrocoalescence is completely absent in the literature.

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