

## Investigation of Electrical Drop-Drop Coalescence of the Simulated Water Droplets Falling in Sunflower Oil

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### 1. Abstract

In many industries, dispersion of water droplets in oil or oil droplets in water is not desirable and they must be separated. Thus, Non-Uniform Electric field (NUE) is used as an ecological method for separating such a dispersed phase. In this paper, performance of water droplets in sunflower oil was investigated under NUE. Measurements were obtained utilizing a batch of cylindrical demulsifier under a high-voltage system. The cylinder was filled with sunflower oil. Top of this cylinder had a tiny hole allowing a needle to be inserted for introducing water drops by a syringe falling in oil to create drop-drop coalescence under NUE. The effect of three different waveform types (ramp-ac, sinusoidal, and pulse-ac) was also investigated. During the experiment, three coalescence patterns were observed using various voltages and frequencies: complete coalescence, incomplete coalescence (left secondary droplet, which is not needed), and no coalescence. Using speed camera, results of the experiment also showed that ramp waveform was the best type of waveform for making a complete coalescence and demulsification process on the synthesized emulsion (88% of separation efficiency) and 60 Hz and 150 V/mm were optimum frequency and amplitude, correspondingly. Diameter of the water droplets was in approximate range of 650 - 1200  $\mu\text{m}$ .

### 2. Introduction

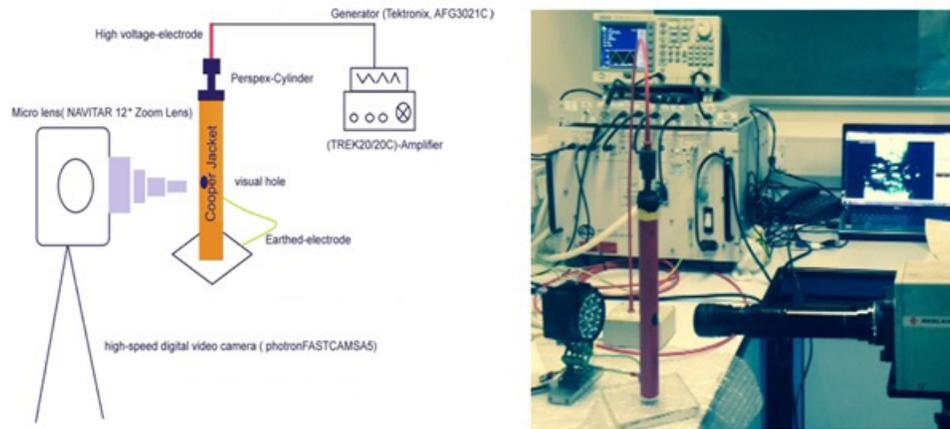
The knowledge on coalescence behavior is essential for separation of oily wastewater or water demulsification in crude oil. Coalescence behavior of water droplets in water-oil interface has been investigated previously under Non-Uniform Electric field (NUE)

using four waveform types (ramp-ac, square-ac, sinusoidal, and pulse-ac) [1]. Among various forms of electric fields, this research focuses on NUE. Electro-coalescence of aqueous droplets in oils is used extensively in oil industry to separate water from oil [2]. Separation methods utilizing high electric fields have been used extensively in petroleum industries [3]. On the one hand, NUE is economical and environmentally friendly and on the other hand, simplicity of the apparatus and process of demulsification makes the related technique suitable for emulsion destruction. Besides electric field, other methods, such as membrane, filtration, etc., are used for separation processes [4] but NUE has been recognized as a good demulsification method due to its beneficial aspects.

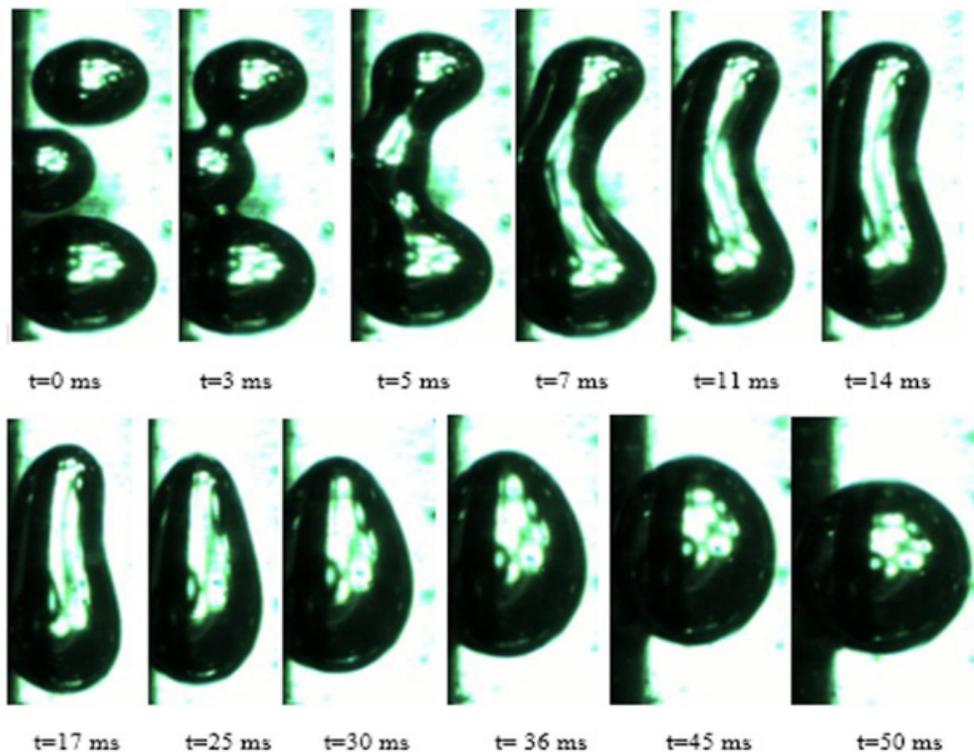
Electrical separation of an emulsion and the effects of temperature, time, voltage, and different kinds of electrical currents have been investigated so far [5]. In specific demulsifier used in this study, one electrode was settled inside cylindrical emulsion container (high-voltage), and the other was a copper formic cylinder jacket (which was grounded as the second electrode) (Figure 1). Accordingly, field was created radially around the immersed electrode by establishing electrical current between two electrodes. In general, two types of droplet coalescence may occur: i) complete coalescence and ii) incomplete coalescence (producing small droplets), removal of which becomes more challenging and consequently, undesirable [6]. According to this experimental study, all the droplets had motions while falling and approaching each other. Besides, the droplets also merged to each other even at a great distance (Figure 2); despite another previous experimental study, upper droplet was approaching stationary lower droplet at a normal angle [7]. When

two water droplets were approaching each other, a film of oil phase was formed between them; consequently, this liquid oil film had to be drained to complete coalescence process [8]. Therefore, in this experiment, for achieving drop-drop coalescence, two or three

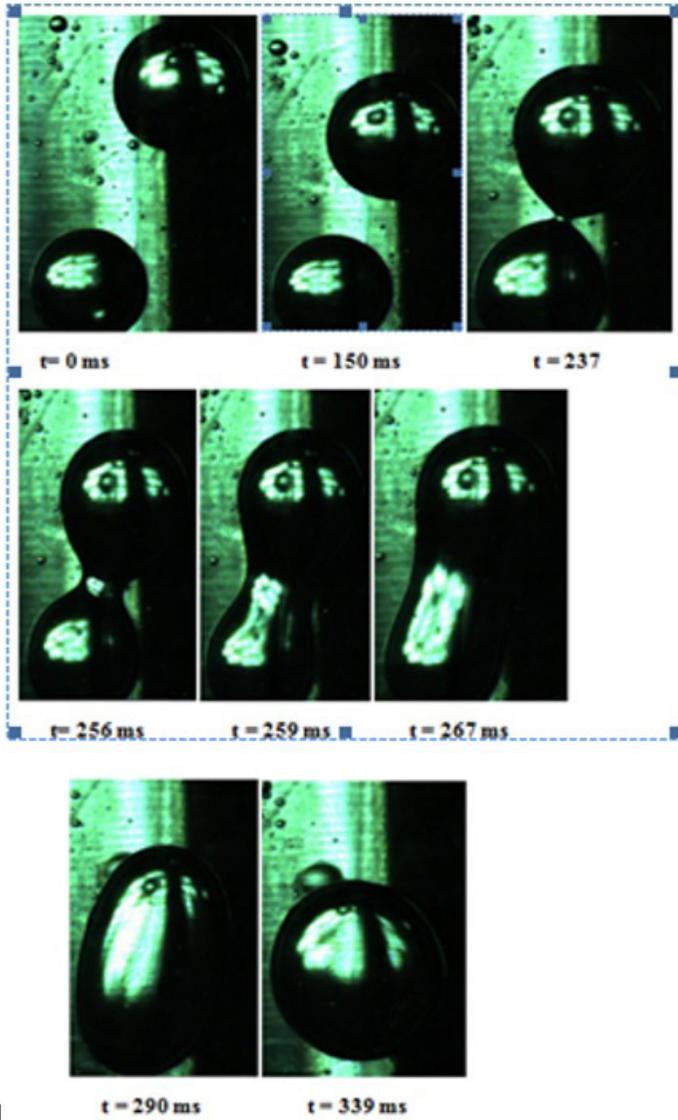
droplets approach each other with different angles (Figure 3). For this purpose, NUE with higher field strength could be applied for the systems with more water content, preventing field breakdown rather than uniform electric field [9].



**Figure1:** A schematic diagram of a non-uniform electrical separation instrument



**Figure 2:** Coalescence of three water droplets under NUE using ramp-ac waveform type and amplitude and frequency of 150 V/mm and 60 Hz, respectively (time: ms=millisecond). Diameter of the water droplets was in approximate range of 650 - 790  $\mu\text{m}$ .



**Figure 3:** A complete coalescence of two drops utilizing ramp-ac waveform type and generating amplitude of 150 V/mm and 60 Hz frequency. Diameter of the water droplets was in approximate range of 650 - 750  $\mu\text{m}$ .

## 2.1. Theory of NUE (Dielectrophoresis)

When a dielectric particle is suspended in a spatially NUE, the applied field's interaction and the induced dipole generate a force on the particle. This force is called as dielectrophoresis (DEP) [10]. DEP-force equation (FDEP) for two coaxial cylindrical containers (co-center) is estimated as follows [11]:

$$F_{\text{DEP}} = 2\pi r^3 \varepsilon_c \text{Re}[K] \nabla |E|^2 / [R \text{Ln}^2(D_j / D_p)] \quad (1)$$

Where,  $r$  is the particle radius,  $\varepsilon_c$  is the relative permittivity of the continuous phase,  $\nabla$  is the del vector operator,  $D_j$  is the copper jacket's diameter,  $D_p$  is the Perspex cylinder's diameter,  $E$  is the local Root Mean Square (RMS) of electric field, and  $\text{Re}[K]$  is the real part of the Clausius–Mossotti factor, given by

$$K = \frac{\varepsilon_d^* - \varepsilon_c^*}{\varepsilon_d^* + 2\varepsilon_c^*} \quad (2)$$

Where,  $\varepsilon_d^*$  and  $\varepsilon_c^*$  are the complex permittivity of the continuous

and dispersed phase (Table1), respectively, and  $\varepsilon^* = \varepsilon - j\delta/\omega$  with  $\delta$  represents the conductivity,  $\varepsilon$  is the relative permittivity, and  $\omega = 2\pi f$  represents the angular frequency. In all the equations,  $c$  and  $d$  refer to the continuous and dispersed phases, respectively. When  $K > 0$ , the particle experiences positive electrophoresis, where the particle moves towards high electric-field gradient regions. Likewise, when  $K < 0$ , the particle experiences negative dielectrophoresis and moves away from high electric-field gradient regions [12].

## 3. Materials and Methods

### 3.1. Simulation of Drop-Drop Coalescence Using NUE Method

#### 3.1.1. Schematic Diagram of the Separator

Separator apparatus consisted of a Perspex cylinder with a copper jacket. Diameter and height of the Perspex cylinder were equal to 2 and 34 cm, respectively. Diameter and height of the copper jacket were equal to 2.2 and 33 cm, respectively considering two holes in the two sides of the jacket to facilitate visualization of coalescence phenomenon. Meanwhile, high-voltage supplier was connected to middle electrode, placed in the Perspex cylinder (brass electrode). In contrast, lower one (cooper jacket) was grounded. A schematic presentation of the system for the related experiments is shown in (Figure 1).

#### 3.1.2. Production of High Voltage and Characterization by Camera

A positive polarity high-voltage direct-current power amplifier (TREK20/20C) was used to generate high voltages. A high-speed digital video camera (Photon FAST CAMS A5), equipped with a micro-lens (NAVITAR12\_Zoom Lens) was used to observe phenomena within the batch cylinder with a framing rate of 20,000 fps (frame per second). It was focused on the area where drop-drop coalescence occurs.

#### 3.1.3. Apparatus Used for Coalescence and Separation Process

In this work, the cylinder was filled with sunflower oil. Top of this cylinder had a tiny hole allowing a needle to be inserted for introducing water droplets by a syringe falling in oil to create drop–drop coalescence. In the experiments, sunflower oil was used and a cold halogen lamp (veritas, constellation 60, 6650K) was applied for lighting; lighting intensity could be accurately adjusted to facilitate focusing. For the dispersed phase, deionized water not containing any surfactant was utilized. Properties of the used liquids are given in (Table 1). The Image-Pro software was used along with the images obtained by the camera to measure the water droplets' diameter. Three waveform types were used to study their effects on coalescence behavior. The experiments were done at 23  $^{\circ}\text{C}$ , and diameter of the water droplets was in approximate range of 650 - 1200  $\mu\text{m}$ .

**Table 1:** Properties of the liquids used in the experiments

Liquids	Conductivity ( $\mu\text{S/m}$ )	Viscosity (mpas)	Density(kg/m <sup>3</sup> )	Dielectric constant
De-ionized water	5.49	1	1000	80
Sunflower oil	$7.62 \times 10^{-5}$	46.5	922	4.9

**Table 2:** Range of the electric field, in which a complete coalescence is predictable for all the three different waveform types.

Waveform type	min. Amplitude (V/mm)	max. Amplitude (V/mm)
Ramp	34	150
Sinus	28	110
Pulse	22	100

### 3.2. Preparation of "Oil –in- Water Emulsion" and Its Demulsification Process

In this work, the emulsion was prepared by shaking the mixture of 1 v/v % sunflower oil and deionized water in a separating funnel for more than 40 times. Volume of the solution was equal to 60 ml for different percentages of oil. For example, for calculating separation efficiency, at first, water was poured into a pipe separator (glass container) with 40 cm of diameter. Then, it was added to 1 v/v % of oil. A ruler was used to measure height of the oil before being emulsified. After emulsification process, the emulsion was placed under the electric field. Separation lasted about 10 min and then, height of the oil was measured separately, which could ultimately be calculated by having these amounts [13].

**% Efficiency = [(Initial height of oil - Final height of oil) / (Initial height of oil)]  $\times$  100**

## 4. Results and Discussion

### 4.1. Basic Panel of the Electrocoalescence Process

Coalescence process of water droplets in sunflower oil was recorded at 20,000 fps and was shown in the sequences of images and movies. When water droplet was allowed to fall in oil, then it was influenced by a high strength electric field under NUE. Formation of a secondary droplet depends on dominating process. In this regard, the following significant results were found:

**Complete Coalescence:** The droplet was pumped into its bulk phase, and no necking occurred, leading to a complete coalescence because pumping process was faster than necking process. The droplet experiences a repulsive Coulomb force by the adjacent electrode. This force is not sufficiently strong (in the case of complete coalescence) to contribute in occurrence of necking process.

**Incomplete Coalescence:** Necking process was faster than pumping process, and in this kind of coalescence, some tiny secondary droplets were put into the continuous phase. Necking and pumping processes are always in competition with each other.

**No Coalescence:** This phenomenon was caused by extreme elongation of two droplets after their collision, and extensive pushing of the produced longer droplet into the continuous phase. Predominance of each of these processes depends on some parameters like

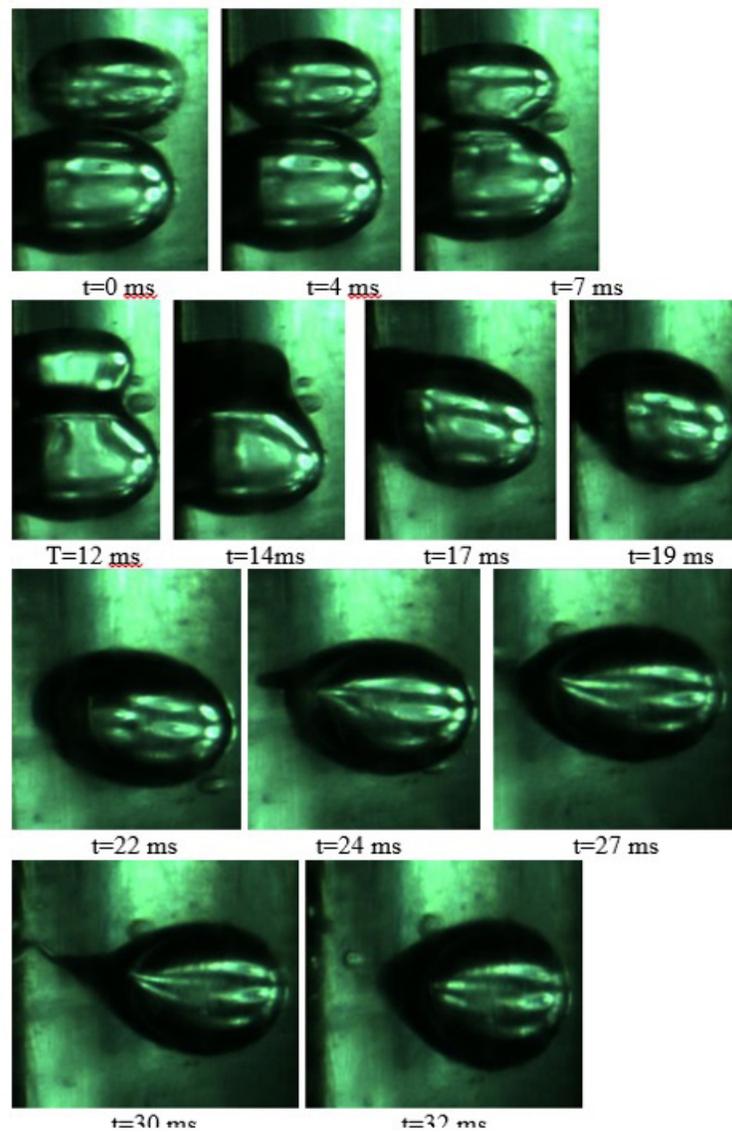
waveform, voltage, and frequency.

### 4.2. Determination of Optimum Amplitude, Frequency, and Waveform Types for Producing Complete Coalescence

In the current work, the effect of three different waveform types on coalescence behavior was investigated, applying various voltages and frequencies. The following results were obtained during this research: No secondary droplets were observed utilizing amplitude less than 100 V/mm while generating all the three mentioned waveform types. The results showed that the effect of frequency on coalescence behavior was negligible using electric field strength lower than 100 V/mm (for all the three waveform types produced complete coalescence). Application of an electric field over 100 V/mm caused development of secondary droplets in all the previously mentioned waveform types except ramp waveform. No secondary droplets were observed using ramp waveform and generating maximum amplitude of 150 V/mm and frequency of 60 - 70 Hz (Figures 2 & 3), which could be a remarkable advantage of ramp waveform in comparison with the other mentioned waveform types. The effect of frequency on coalescence behavior was negligible using electric field strength less than 100 V/mm. Still, its effect was significant in higher amplitudes more than 100 V/mm. Optimum and or maximum amplitude and optimum frequencies were equal to 150 V/mm and 60 Hz, respectively (to produce complete coalescence). Using amplitude of up to 150 V/mm and frequency of 60 Hz, the camera showed that coalescence could be made without creating any secondary droplets only by ramp waveform type (Figures 2 & 3).

### 4.3. The Effect of Frequency on Coalescence Behavior

The frequency played an essential role by applying amplitude higher than 100 V/mm (Figures 2 & 3 compared to Fig. 4). Despite using the same waveform type (ramp) and optimum amplitude of 150 V/mm, different results would be obtained using different frequencies. Low frequency (20Hz) led to incomplete coalescence compared to Figs. 2 & 3. Necking process would be faster than pumping process by applying low frequency (20 Hz). In this kind of coalescence, some tiny secondary droplets will be put into the continuous phase due to necking process (Figure 4).



**Figure 4:** Incomplete coalescence process of two droplets using the ramp-ac waveform type and generating amplitude of 150 V/mm and 20 Hz of frequency. Diameter of the water droplets was in approximate range of 1000 -1200  $\mu\text{m}$ .

#### 4.4. The Effect of Higher Amplitude on Coalescence Behavior

An extreme elongation of droplet was observed after a high-speed and energetic collision by applying a high amplitude up to 200 V/mm for ramp-ac waveform and using an optimum frequency of 60 Hz, because of extensive pushing of the droplet into the continuous phase, which finally resulted in division of the two attached droplets in two parts again (Figure 5).

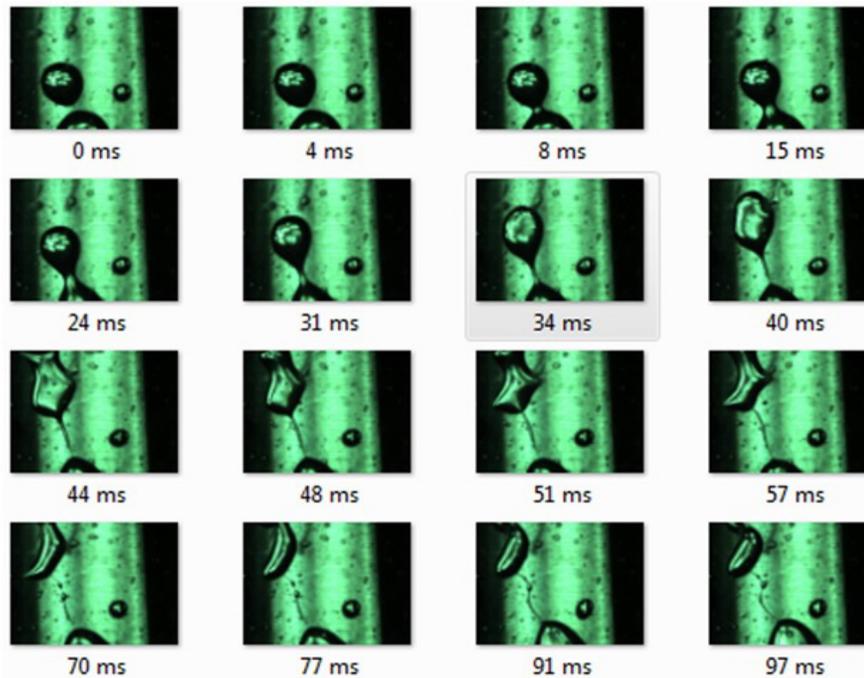
In this work, pulse waveform was found to work by lower amplitude than the other waveform types, but there was no complete coalescence in amplitude range of 20 - 100 V/mm. The electric field ranges, in which a complete coalescence is predictable for sinusoidal and pulse-ac waveform types are as follows: between 28 - 110 V/mm for sinusoidal and between 34 - 150 V/mm for ramp-ac waveform types, respectively (Table 2).

#### 4.5. Investigation of the Other Two Waveform Types' Effect Compared to Ramp Waveform by Applying Optimum Amplitude and Frequency

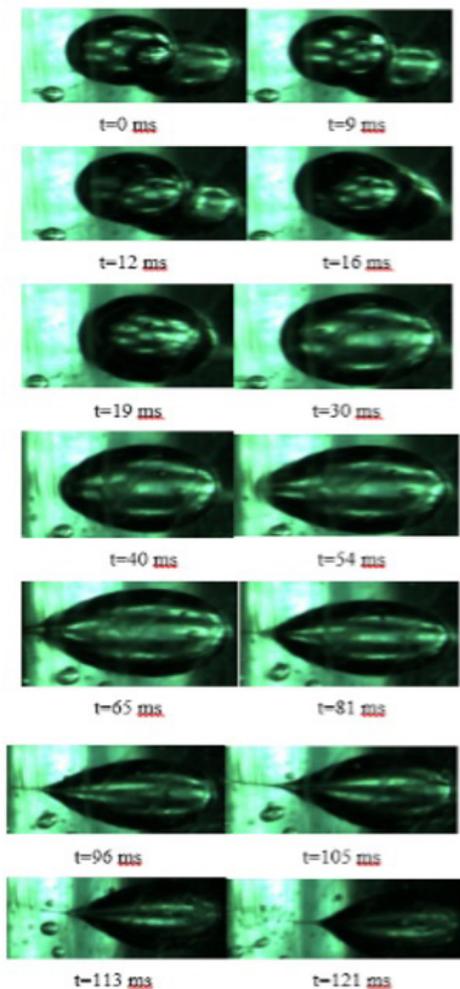
##### 4.5.1. Sinusoidal Waveform

(Figure 6) demonstrates an incomplete coalescence using a sinusoidal waveform by applying optimum amplitude and frequency of 150 V/mm and 60 Hz. Significantly smaller secondary droplets were developed by spreading the sinusoidal waveform as shown in Fig. 6 after two droplets' coalescence and making bigger droplets. Necking led to this phenomenon in this case; necking process was faster than pumping process.

(Figure 6) shows a pattern for an incomplete coalescence because of producing some secondary droplets, where necking process was dominated compared to pumping process.



**Figure 5:** A pattern of no-coalescence using ramp-ac waveform type and applying amplitude and frequency of 200 V/mm and 60 Hz, respectively. Diameter of the water droplets was in approximate range of 650 - 800  $\mu\text{m}$ .



**Figure 6:** Deformation of two water droplets as a pattern of incomplete coalescence under NUE using the sinusoidal waveform type and applying amplitude and frequency of 150V/mm and 60Hz, respectively. Diameter of the water droplets was in approximate range of 900 - 1000  $\mu\text{m}$ .

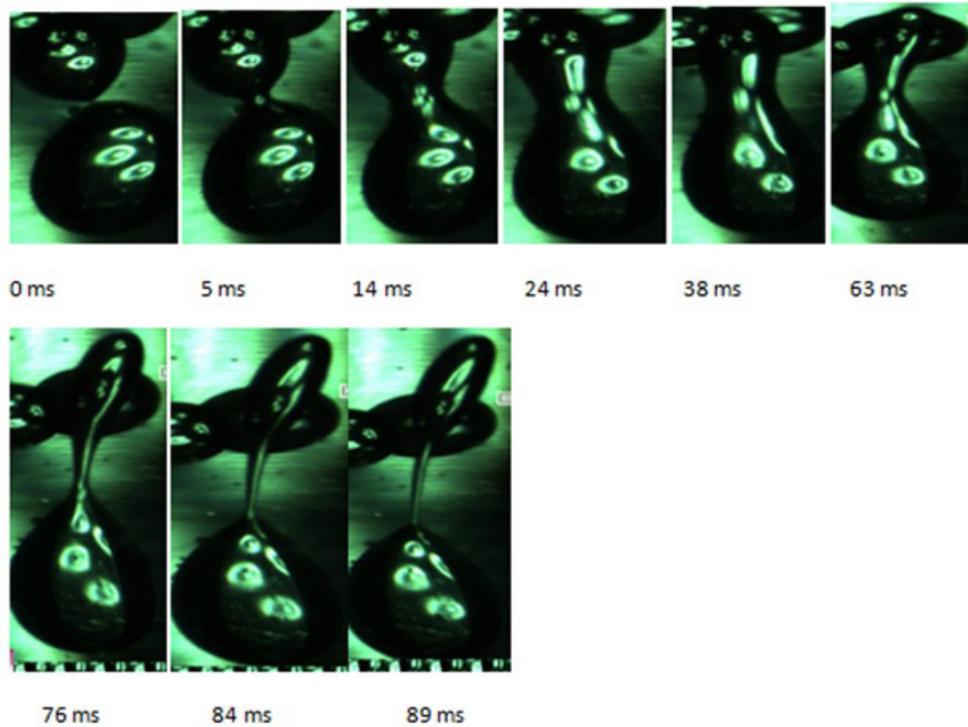
#### 4.5.2. Pulse Waveform

(Figure 7) shows a drop-drop coalescence by generating an optimum amplitude of 150 V/mm and an optimum frequency of 60 Hz using pulse waveform. Generating pulse waveform led to no-coalescence because it caused separation of the two coalesced droplets again due to extreme elongation of two droplets after collision (Figure 7). In contrast, it led to a complete coalescence using a ramp waveform with the same circumstances (Figs. 2 & 3).

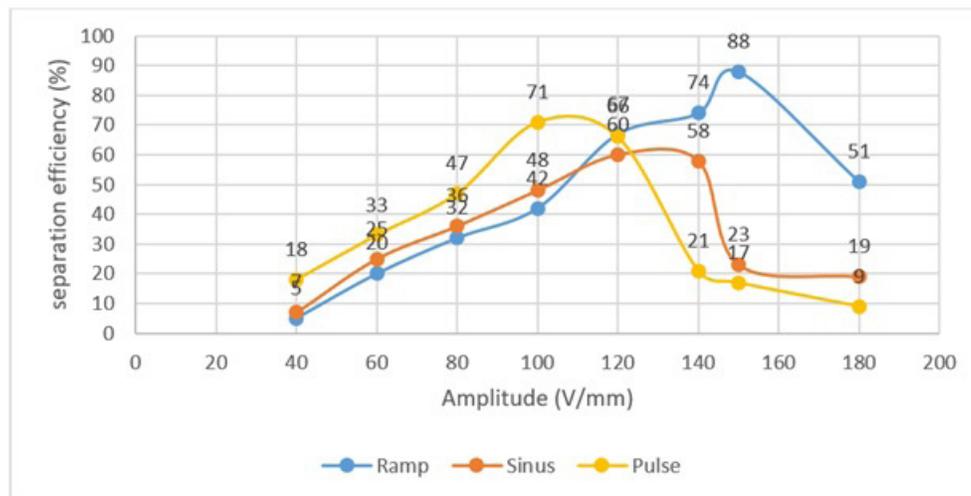
The waveform's effect is significant so that, ramp waveform's advantage was remarkable compared to the pulse-ac waveform (Figure 7) and sinusoidal waveform (Figure 6). As can be seen in Figure 7, the droplets have got an extreme elongation (due to extensive pushing of the droplet into the continuous phase) and finally, two droplets come back to their initial form after they would be separated again.

### 5. Demulsification of the Synthesized Oil -in -Water Emulsion

Finally, for understanding whether this type of electrical field influences similarly to demulsification process of the synthesized emulsion, an oil -in -water emulsion was prepared and finally, was demulsified under the mentioned electric field. (Figure 8) illustrates separation efficiency of oil -in -water emulsion using different waveform types and applying different amplitudes with a constant frequency of 60 Hz (1 v/v. % emulsion of sunflower oil in de-ionized water). As shown in Figure 8, all the three waveform types have higher demulsification rate by generating amplitude between 100 - 150 V/mm and the ramp waveform also was the best kind of waveform among all three types of waveform for demulsification process and 60 Hz and 150 V/mm were optimum frequency and amplitude, correspondingly.



**Figure 7:** Deformation of water droplets- as a pattern of no complete coalescence under NUE using the pulse-ac waveform type and applying amplitude and frequency of 150 V/mm and 60 Hz, respectively. Diameter of the water droplets was in approximate range of 700 - 1000  $\mu\text{m}$ .



**Figure 8:** Separation efficiency of oil-in-water emulsion using different waveform types and applying different amplitudes with a constant frequency of 60 Hz

## 6. Conclusion

When the droplet was allowed to fall in oil, it was influenced by high strength electric field under NUE. Formation of a secondary droplet depends on dominating process. In this regard, the following significant results were achieved:

**Complete Coalescence:** The droplet was pumped into its bulk phase, and no necking occurred, leading to a complete coalescence because pumping process was faster than necking process (ramp waveform Figures. 2 & 3). The droplet experiences a repulsive Coulomb force by the adjacent electrode. This force is not suffi-

ciently strong (in the case of complete coalescence) to contribute in occurrence of necking process.

**Incomplete Coalescence:** As shown in (Figure 6), necking process was faster than pumping process by generating sinusoidal waveform and finally, led to an incomplete coalescence. Necking and pumping processes are always in competition with each other. In this work, incomplete coalescence occurred using a low frequency of 20 Hz (Figure 6).

**No Coalescence:** No coalescence was caused by very fast and energetic coalescence and extensive pushing of the droplet into

the continuous phase. Predominance of each of these processes depends on some parameters like waveform type (pulse-ac, Figure 7) and a voltage higher than 200 V/mm (Figure 5).

The water droplets' diameter falling in oil was in approximate range of 650 - 1200  $\mu\text{m}$ .

After the experiment, it was found that the ramp waveform was the best type of waveform among all the three types of waveform to avoid producing secondary droplets and make a complete coalescence. Generating 60 Hz and 150 V/mm was also considered as optimum frequency and amplitude, correspondingly. For demulsification of the oil -in -water emulsion, the ramp waveform was the best kind of waveform among all the three types of waveform (88% of separation efficiency) similar to a complete coalescence occurred in the first simulation process.

### Nomenclature

E	field intensity (V/m)
r	droplet radius (mm)
R	the distance between the droplet and the central electrode (mm)
$D_p$	Perspex cylinder diameter (mm)
$D_j$	copper jacket diameter (mm)
$\epsilon^*$	complex permittivity
$\epsilon_c$	relative conductivity of the suspending medium
$\delta$	conductivity (S/m)
$\omega$	angular frequency of the electric field
f	frequency(Hz)
$\nabla$	del vector
$\pi$	integer =3.14
c, d	continuous, disperse

### 7. Acknowledgments

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