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1 The impact of electric field on the demulsification efficiency in an  
2 electro-coalescence process

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20 Abstract

21 In this study, the influence of the electric field and its determining parameters such as voltage,  
22 inter-electrode distance and frequency on the demulsification efficiency of a water-in-oil emulsion  
23 was studied. The numerical analysis showed that the water droplets trapped onto the hydrophobic  
24 titanium dioxide (TiO<sub>2</sub>) coated electrodes and at 2 mm inter-electrode distance can increase the  
25 electric field intensity above 300 kV/m, where droplet rupture takes place. Experimental study  
26 showed highest demulsification efficiency of 76% while applying AC voltage of 250 V, frequency  
27 of 200 kHz and inter-electrode distance of 4 mm.

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32 Keywords: Water-in-oil emulsion; Dielectrophoresis; Demulsification; High frequency alternating  
33 current.

## 34 1. Introduction

35 Naturally, crude oil contains large amount of dispersed water droplets with an average radius of  
36 50  $\mu\text{m}$ . Turbulent mixing, agitation through downhole wellbore, surface chokes and other drilling  
37 activities disperse these water droplets and emulsifies the droplets in the oil system [1]. Crude oil  
38 also contains other substances such as asphaltenes, resins, naphthenic acids and other solid  
39 particles [2, 3]. These natural surfactants prevent the rupture of the water droplets hence  
40 stabilizes the emulsion by preventing coalesce of the water droplets in crude oil [4]. The existence  
41 of these water droplets in oil results in corrosion of the pipeline and reduces the efficiency of the  
42 pumps and heat exchangers [1, 5]. In the industry, demulsification of water-in-oil emulsion is  
43 carried out by adding chemical demulsifiers such as poly alkylene oxides, poly alkylene glycol,  
44 ethylene oxide, polyamines, fatty alcohols, alkyl phenols, fatty acid ester and mercaptans [6].  
45 These chemicals are expensive and can create toxic byproducts, thus, better demulsification  
46 processes are still being investigated. The primary aim of the demulsification process is to  
47 coalesce the water drops which then can be easily separated by gravity. To promote droplet  
48 coalescence, Romanova et al. (2022) injected aluminum nitride (AlN) suspended in acetone as  
49 an additive in the emulsion and ultrasonicated the emulsion [7]. In the process, by adding 8 v/v%  
50 additive, 99% demulsification was achieved within 3 min of ultrasonication. Cao et al. (2022)  
51 modified a polyacrylonitrile (PAN) microfiltration membrane with polytetrafluoroethylene (PTFE)  
52 nanofiber to demulsify a water-in-oil emulsion [8]. In this study, the membrane casted with 9%  
53 PTFE achieved 99.99% demulsification efficiency along with a permeate flux of 4,909 LMH. Zhu  
54 et al. (2022) coated a recycled wet wipe with hydrogel and carried out demulsification of an  
55 oil/water emulsion stabilized by cationic surfactants [9]. During demulsification, the negatively  
56 charged hydrogel surface attracted the positively charged surfactants which promoted  
57 demulsification. The process achieved <99% demulsification efficiency with a flux of 3,057 LMH.  
58 Apart from chemical demulsifiers, the coalescence of water droplets can also be promoted by

59 adding biological demulsifiers, thermal treatment, membrane demulsification, electrical  
60 demulsification and microwave irradiation [10]. Among the different technologies used for the  
61 demulsification of water-in-oil emulsions, the electrical demulsification technology is considered  
62 as one of the simplest, the most environmentally friendly and the most efficient technology with  
63 lowest energy demand [11, 12]. In electrical demulsification, a uniform or non-uniform electric field  
64 is applied to polarize the water droplets. The net charge on water droplets induces electrophoretic  
65 (EP) force in a uniform electric field. The polarization of water droplets generates dielectrophoretic  
66 force (DEP) in a non-uniform electric field. The presence of EP or DEP in the system promotes  
67 coagulation of the water droplets. The main challenges for an electrical demulsification process  
68 include formation of pearl chain, electrochemical reactions on the electrode surface and formation  
69 of secondary droplets. To avoid formation of pearl chains, high frequency non-uniform AC electric  
70 field has been suggested by Zhou et al. (2022) [13]. Mousavi et al. (2014) carried out  
71 demulsification of a water-in-oil emulsion using poly(methyl methacrylate) coated polished brass  
72 electrodes and applied a pulsatile electric field (PEF) [14]. In the study, by applying PEF at 100  
73 Hz to a sunflower oil-based emulsion, coalescence was achieved without the formation of  
74 secondary droplets. The study also found that by applying direct current (DC) in sawtooth  
75 waveform is better for prevention of secondary droplet formation than applying DC with square or  
76 half-sinusoidal waveform. Less et al. (2008) studied demulsification of water-in-oil emulsion using  
77 the Aibel Vessel Internal Electrostatic Coalescer (VIEC) where the electrodes were insulated with  
78 an epoxy coating [15]. In the study, applying an alternating current (AC) at high voltage with the  
79 addition of 20 ppm chemical demulsifier resulted in the removal of 95% water from the emulsion  
80 within 5 mins, whereas, without applying an alternating current only 80% water was removed from  
81 the emulsion. Li et al. (2022) studied the electrocoalescence process of water droplets in  
82 sunflower oil in a non-uniform electric field and in laminar flow conditions using different electrode  
83 arrays [16]. The numerical study showed that higher electric field intensity can be achieved in the  
84 electrocoalescence process by using the mesh electrode array compared to the grid electrode

85 array. Moreover, the study also found that, at high Reynold's number, the demulsification  
86 efficiency increases. Table 1 summarizes the mechanisms, advantages and disadvantages of the  
87 different demulsification techniques.

88 In this work, for a better and deeper understanding of the electrocoalescence process, we  
89 numerically simulated and experimentally demonstrated the influence of electric field on the  
90 demulsification efficiency by varying the electric field determining parameters (including voltage,  
91 frequency and distance between electrodes). In addition, the impact of the entrapment of water  
92 droplets on the surface of the electrodes was investigated. An optimal design of an  
93 electrocoalescence process to achieve high demulsification efficiency with low energy demand  
94 was suggested.

95 **Table 1.** Summary of different demulsification technologies with their advantages and disadvantages.

Demulsification technique	Mechanism	Advantages	Disadvantages	References
Chemical demulsification	Chemical demulsifiers are amphiphilic in nature and reduces the surface tension of the emulsion by adsorbing oil and water on its surface. The reduction of surface tension promotes coalescence of the water droplets.	-Simple operation -Low residence time -Cost effective	-High toxicity -Non-renewable	[17]
Biological demulsification	In biological demulsification, bacteria is used to reduce the interfacial tension which in turn promote coalescence of the droplets.	-Low toxicity -Biodegradable -High efficiency at extreme conditions	-Vulnerable to sudden operation changes	[18]
Thermal demulsification	In thermal demulsification, application of heat reduces the viscosity of oil and water. This reduces the mechanism strength of the interfacial films and increases mobility of the particles. These promotes coalescence of the water droplets.	-Dissolves paraffin crystals -No harmful byproducts	-Loss of light oil fraction -Lower separation efficiency -Increased corrosion risk	[19]
Membrane demulsification	In this process, a membrane is used to separate the oil/water droplets from the bulk solution.	-Free of chemical contaminants -Low energy requirement	-Concentration polarization	[8]

Electrical demulsification	In this process, electric field is used to cause collision among the water droplets which in turn will promote droplet coalescence.	-Environment friendly -Lower energy consumption	-Difficult to regenerate membranes -Formation of secondary droplet	[20]
Microwave irradiation	In this process, by using microwaves, the molecules of the components in the dispersed phase are rotated which increases ionic conduction of the particles. The increased ionic conduction promotes coalescence of the particles.	-Low treatment time -Selective heating of the dispersed phase -No chemical contaminants	-Low separation efficiency	[21]

97 2. Methodology

98 2.1 Preparation of the water-in-oil emulsion

99 QALCO performance oil #68 was utilized to prepare the water-in-oil emulsion. Table 2  
100 summarizes the properties of the used oil. The total volume of the water-in-oil emulsion used  
101 during all the studies was 100 ml. The water content in the emulsion was varied between 2.51-  
102 11.27 wt%. The oil and water were dispersed using an Ultra Turrax T25 (IKA) disperser in a 500  
103 mL beaker. The disperser can operate at a dispersion speed between 3,000 and 25,000 rpm. The  
104 oil and water mix were dispersed for 5 to 20 minutes. The concentration of water in the water-oil  
105 emulsion was measured using a moisture analyzer (OHAUS, Germany). The moisture analyzer  
106 works by heating the sample to 120 °C to evaporate all the available water in the emulsion. The  
107 difference in weight before and after evaporation indicates the moisture content of the emulsion.

108 **Table 2.** Properties of the oil used for preparing the water-in-oil emulsion.

Flash Point	220°C
Appearance	Clear Amber Yellow
Physical State	Liquid
Vapor Density (air=1)	>1
Boiling Point	>260 °C
Solubility in Water	Negligible
Specific Gravity	0.875
Viscosity	42-50 cSt
Conductivity	<0.001 μS/cm

109

110 To find the lowest water content, samples were taken from different locations of the sample holder  
111 and it showed a gradual decrease in water content along the depth of the sample holder where  
112 the lowest water content was found at the top and the highest water content was found at the  
113 bottom. The stability of the water-in-oil emulsion was evaluated by examining the water  
114 concentration on the surface of the water-in-oil emulsions prepared under different conditions,  
115 such as mixing speed, dispersion time, and water content in the emulsion. Under these conditions,  
116 water content was plotted as a function of time. To identify emulsion stability, the slope of the  
117 points was determined. Higher stability was indicated by lower slope.

118 The effect of water content on the stability of the water-in-oil emulsion was studied by varying the  
119 water content between 2.51 and 11.27 wt% at a constant dispersion speed of 10,000 RPM and 5  
120 minutes of dispersion time. As shown in **Figure S1** of the Supplementary Information, higher  
121 water content in the emulsion reduced the stability of the prepared emulsion. This is because, at  
122 higher water concentrations, the number of water droplets in the emulsion is higher. This presents  
123 more opportunity for the water droplets to collide, coalesce and form larger droplets which then  
124 settle easily by gravity and reduce the stability of the emulsion. Since the demulsification  
125 experiments will be carried out for 60 mins, and the stability of the emulsion for both 2.51 wt%  
126 and 4.71 wt% water-in-oil emulsion is almost similar with slopes of -0.0004 and -0.0005,  
127 respectively, a water content of 4.71 wt% was used to prepare the emulsions. Using 4.71 wt%  
128 water and 5 minutes of dispersion time, the effect of dispersion speed on the stability of the water-  
129 in-oil emulsion was studied. As presented in **Figure S2** of the Supplementary Information, a  
130 dispersion speed of 5,000 RPM gave the lowest stable emulsion presented by the highest slope  
131 of its tendency line. Whereas dispersion speed of 10,000 RPM and higher gave almost the same  
132 stability of the emulsion. Therefore, in this study, dispersion speed of 10,000 RPM was used to  
133 prepare the water-in-oil emulsion. At 10,000 RPM dispersion speed and 4.71 wt% water content,  
134 the effect of dispersion time on the stability of the water-in-oil emulsion was studied. As shown in

135 **Figure S3** of the Supplementary Information, a dispersion time of 5 minutes presented the highest  
136 stable water-in-oil emulsion where the difference of water content of the emulsion after 180  
137 minutes was only 0.24 wt%. The very unstable emulsion after longer dispersion times is mainly  
138 due to the heat generated by the high-speed mixer. It was noticed that at 10,000 RPM the  
139 temperature of the emulsion increased from 23°C at 5 minutes dispersion time to 42°C after 20  
140 minutes of dispersion time (**Figure S4** of Supplementary Information). The increased temperature  
141 promotes coalescence of the water droplets which reduces stability of the emulsion. It can be also  
142 noticed from **Figure S2** and **Figure S3** that, although the initial water concentration for the  
143 dispersion speed and dispersion time study was fixed at 4.71 wt%, after dispersion, the initial  
144 water content was not identical. This is because, while using the homogenizer, mechanical  
145 heating was induced in the system as indicated by **Figure S4**. Thus, at higher dispersion time  
146 and dispersion speed water was lost from the system as indicated by lower initial water content  
147 at higher dispersion time and speed. This was also a factor for deciding the optimum dispersion  
148 time and speed. The selected dispersion time of 5 mins and dispersion speed of 10,000 RPM  
149 showed lowest loss of water due to mechanical heating. Hence, we considered the emulsion to  
150 be more stable while using these operating conditions.

151 The water droplet size analysis was carried out using a MALVERN Zetasizer nano (UK) and can  
152 be seen in **Figure S5** of the Supplementary Information. As seen in **Figure S5**, increasing the  
153 water content in the emulsion also resulted in increased water droplet size in the emulsion. The  
154 largest and smallest average water droplet size in the prepared water-in-oil (W/O) emulsion was  
155 found to be 797 nm and 291 nm for W/O emulsion with 11.27 wt% and 2.51 wt% water,  
156 respectively. Generally, the presence of bigger water droplets makes the emulsion less stable,  
157 whereas smaller water droplet size would result in higher emulsion stability.

## 158 2.2 Experimental setup

159 The experimental setup for the demulsification process comprised of a high-frequency AC power  
160 supply (G 2000, Redline Technologies), an oscilloscope (MDO3024, Tektronix) utilized to  
161 measure the current (A) and the voltage (U), two titanium electrodes, and a glass vial (length: 10  
162 cm, diameter: 15 mm). The titanium electrodes were 2 mm in diameter. The electrodes surface  
163 was coated with a 200 nm thin film insulating layer of hydrophobic titanium oxide (TiO<sub>2</sub>, rutile).  
164 The electrodes were coated with TiO<sub>2</sub> hydrophobic layer in order to reduce the electrowetting  
165 effect and the high-pass filter effect. In addition, the TiO<sub>2</sub> coating will eliminate electrochemical  
166 reactions on the surface of the electrodes, prevent the formation of gas bubbles, and corrosion of  
167 the electrodes.

168 Figure 1 (a) shows a schematic diagram of the experimental set up. During the demulsification  
169 process, 10 mL of the stable water-in-oil emulsion was placed in the glass vial and a high  
170 frequency AC voltage was supplied by the AC power supply where the waveform of the current  
171 and voltage measured by the oscilloscope was a Sine wave. The water content at the top of the  
172 emulsion was analyzed after 15, 30, 45 and 60 minutes of demulsification. The demulsification  
173 performance using high frequency AC was compared with low frequency AC and direct current  
174 (DC). All experiments were repeated three times and the average of the results is reported.  
175 Moreover, the standard deviation of the results is expressed in the form of error bars. The  
176 demulsification efficiency ( $\eta_w$ ), represented by the efficiency of water removal, was calculated as:

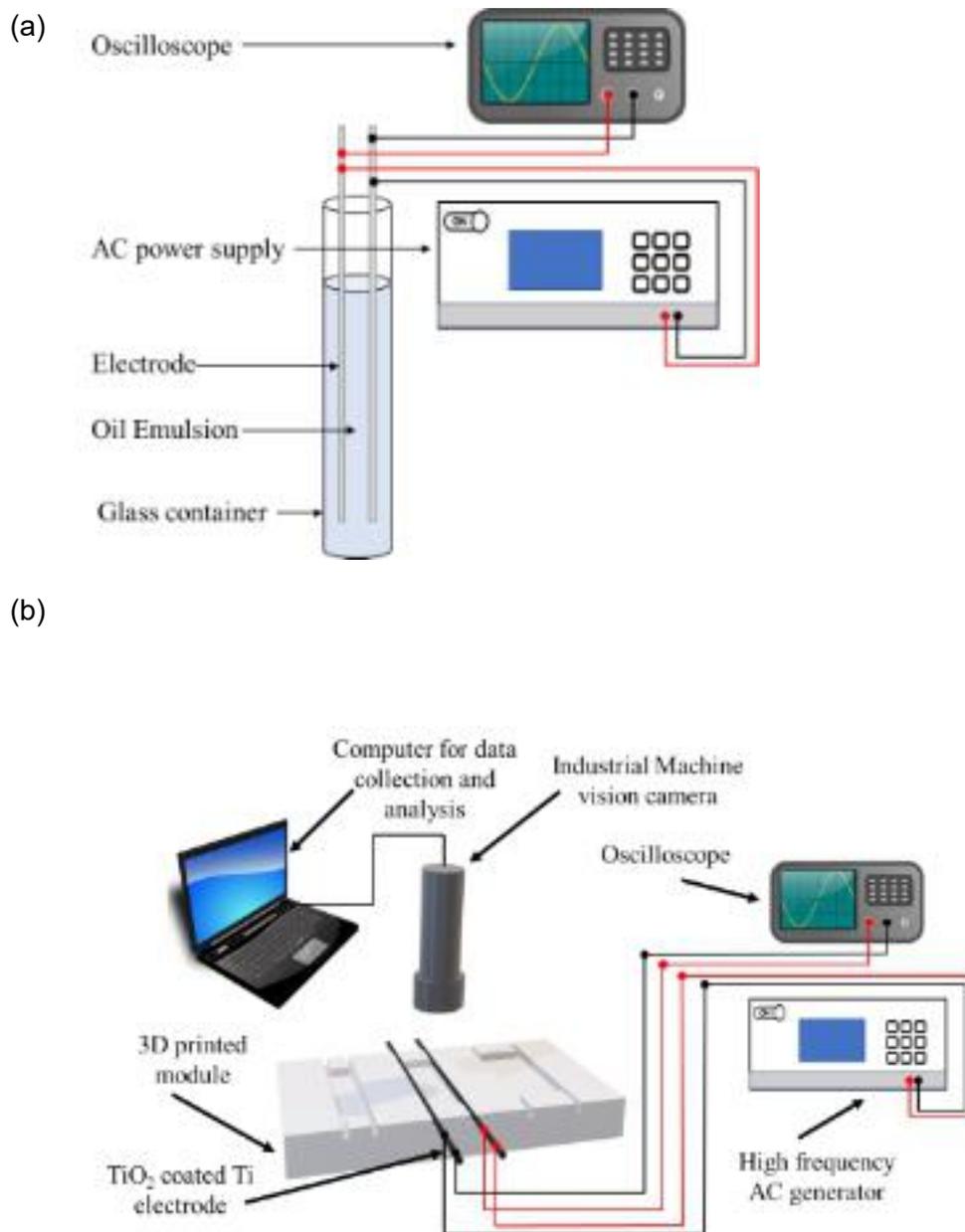
$$\eta_w = \frac{C_{wt} - C_{w0}}{C_{w0}} \quad (1)$$

177

178 where,  $C_{w0}$  is water content at the beginning of the experiment (wt%),  $C_{wt}$  is the water content at  
179 the end of demulsification process (wt%). To observe the formation of the water droplets, a 3D  
180 printed module with channels for electrodes and water-in-oil emulsion was designed (

181 Figure 1 (b)). The 3D printed module was equipped with an industrial machine vision camera  
182 (MER-112-32U3C, Daheng Imaging) connected to a computer for data collection and analysis.

183



184

185 **Figure 1.** (a) schematics of the experimental setup, (b) droplet coalescence observation setup.

### 186 3. Numerical Analysis

187 Dielectrophoresis (DEP) is an electrokinetic effect of translational motion of neutral or charged  
188 particles instigated by dielectric polarization in an inhomogeneous electric field [22]. The  
189 dielectrophoretic force ( $F_{DEP}$ ) due to the non-uniform electric field on the produced dipole moment  
190 of the particle is dependent on the dielectric properties and size of the particle, the frequency of  
191 the applied field, and the electrical properties of the medium [23]. The DEP force acting on a  
192 spherical particle can be calculated as [24]:

$$F_{DEP} = 4\pi a^3 \epsilon_0 \epsilon_M \text{re}[\tilde{K}](E \cdot \nabla)E \quad (2)$$

193 Here,  $a$  is the radius of the particle (m),  $\epsilon_0$  is the permittivity of free space ( $8.854 \times 10^{-12} \frac{F}{m}$ ),  $\epsilon_M$  is  
194 the relative permittivity of the medium (F/m),  $E$  is the field intensity ( $\frac{V}{m}$ ) and  $\text{re}[\tilde{K}]$  is the real part  
195 of Clausius-Mossotti (CM) factor, which is calculated as [23]:

$$\tilde{K} = \frac{\tilde{\epsilon}_p - \tilde{\epsilon}_M}{\tilde{\epsilon}_p + 2\tilde{\epsilon}_M} \quad (3)$$

$$\tilde{\epsilon} = \epsilon - \frac{j\sigma}{\omega} \quad (4)$$

196 Here,  $\tilde{\epsilon}_p$  is the complex permittivity of the particle (F/m),  $\tilde{\epsilon}_M$  is the complex permittivity of the  
197 medium (F/m),  $\tilde{\epsilon}$  is the complex permittivity (F/m),  $\sigma$  is the conductivity ( $\frac{S}{m}$ ),  $\omega = 2\pi f$  is the angular  
198 frequency ( $\frac{rad}{s}$ ),  $f$  is the frequency (Hz),  $j = \sqrt{-1}$ , the geometric gradient of the square of the  
199 electric field ( $E$ , V/m) is calculated by [23]:

$$(E \cdot \nabla)E = \frac{1}{2} \nabla |E|^2 \quad (5)$$

200 As presented in Equation 2, the gradient of the square of the electric field can be used as an  
201 indicator of the DEP force in a DEP system with given properties of particle and medium.  
202 Therefore, the gradient of the square of the electric field was simulated to present the impact of

203 the different parameters on the demulsification efficiency. Numerical analysis using COMSOL  
204 Multiphysics 5.5 was performed to predict the electric field strength generated at different inputs  
205 of voltage ( $U$ , volt) and different inter-electrode distances ( $d$ , meter).

206 The sketch for the simulated geometry is shown in Figure 2 (a). The simulation was conducted in  
207 two dimensions, assuming that the height of the electrode is infinite. The electric potential was  
208 solved at a set of boundary conditions to calculate the gradient of the square of the electric field.  
209 The medium between the electrodes was assumed to be oil with a known specific conductivity.  
210 This assumption was made because the medium is a continuous phase of oil with minute water  
211 droplets behaving as particles. To solve this problem for the applied currents and frequencies,  
212 the quasi-electrostatic form was used. The root mean square (rms) of the electric field is given  
213 by:

$$E = -\nabla\varphi \quad (6)$$

214 Here,  $\varphi$  is the rms of the electrostatic potential which can be given by Laplace's equation  
215 (assuming that the medium is liquid only with the absence of any particles and it is homogeneous):

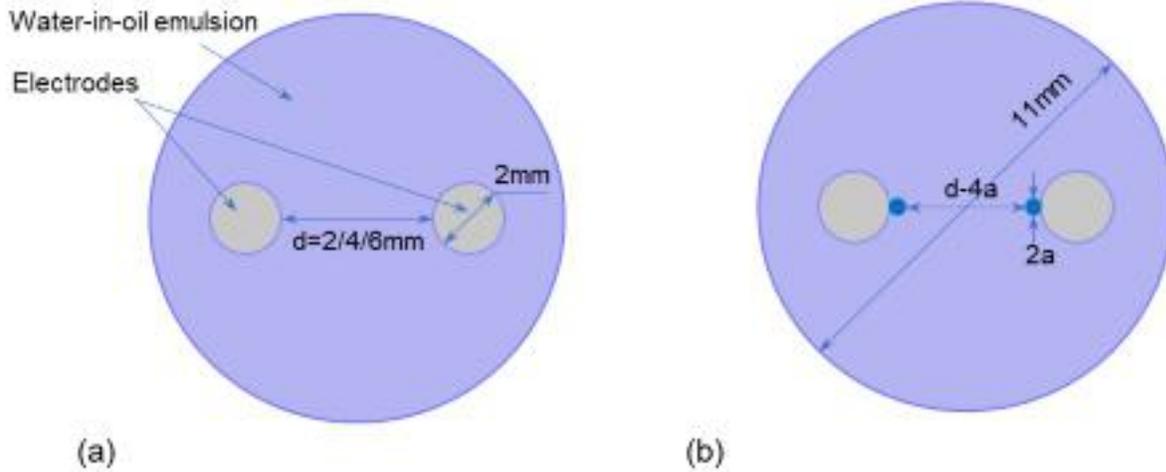
$$\nabla^2\varphi = 0 \quad (7)$$

216 Fixed boundary conditions were applied on the surface of the charge carrying electrodes:

$$\varphi_1 = U_0 \quad (8)$$

$$\varphi_2 = 0 \quad (9)$$

217 Here,  $U_0$  is the rms of the oscillating potential drop (volt). An 'extremely fine' mesh was applied to  
218 carry out the simulations. To further improve the accuracy of the results, the mesh size can be  
219 reduced.



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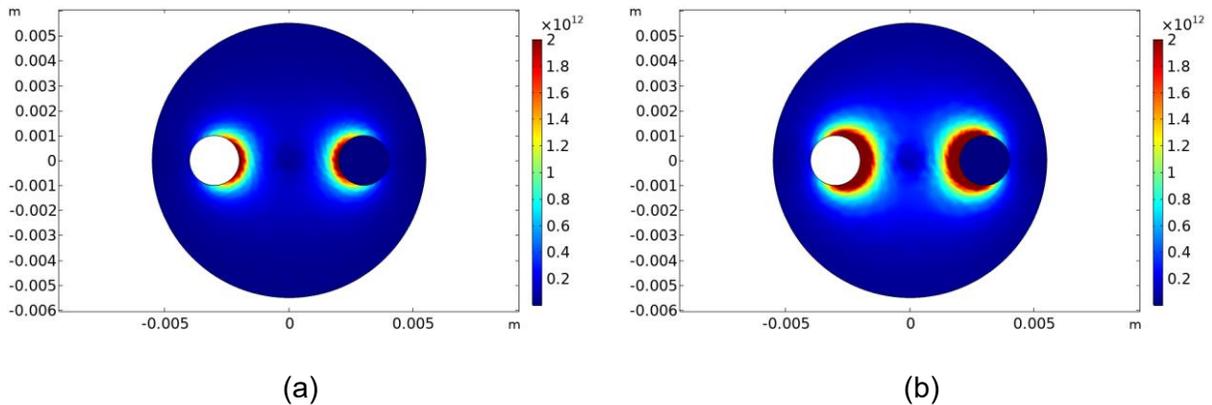
222 **Figure 2.** (a) Sketches of the geometry considered in the simulation of the two electrodes (b) the  
 223 considered geometry for the simulation of the entrapment of water droplets (blue circles) on the  
 224 surface of the electrodes (gray circles).

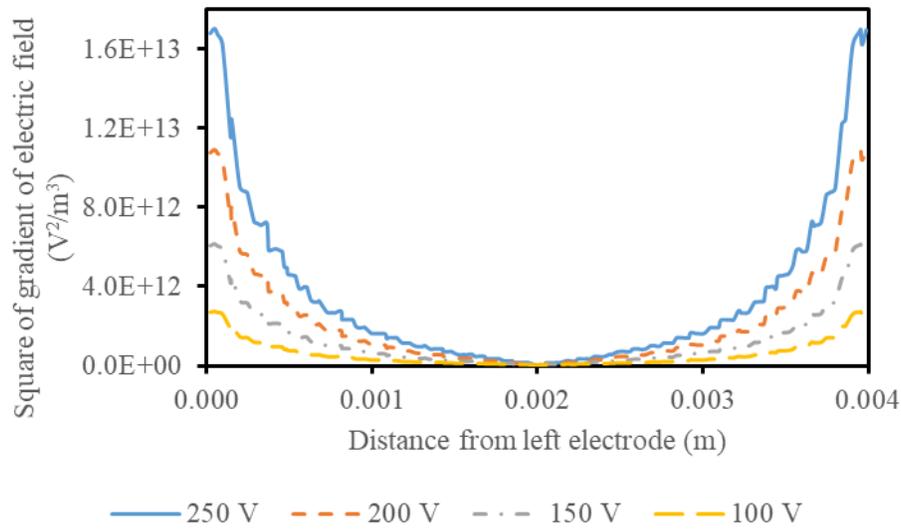
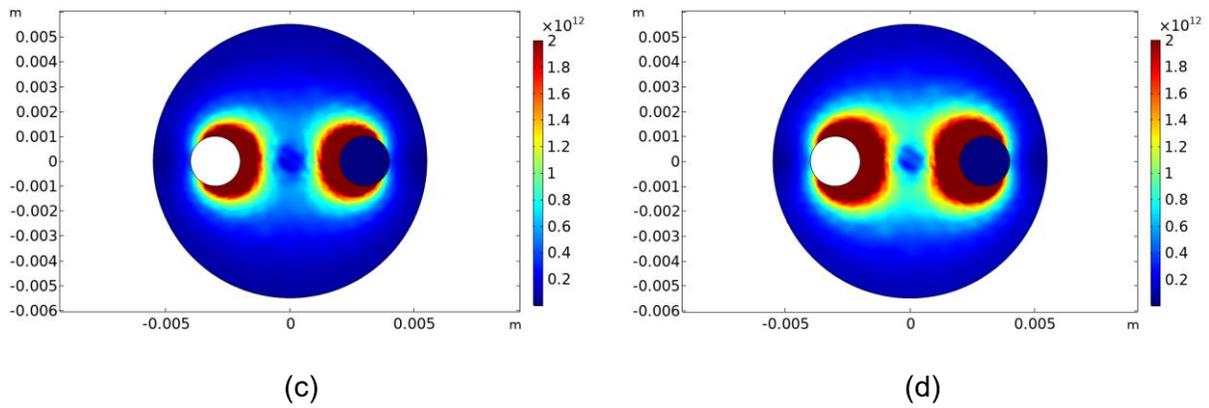
225 To study the influence of trapped water droplets on the surface of the electrodes on the electric  
 226 field strength, two water droplets of radius 'a' were placed on the surface of each electrode,  
 227 between the two electrodes (Figure 2 (b)). Although the  $\text{TiO}_2$  film is resistive with a conductivity  
 228 of  $10^{-13}\Omega^{-1}\text{cm}^{-1}$ , high frequency signal can pass through the film. During this study, the range of  
 229 frequency was between 175 and 250 kHz. At this high frequency, the electric field in the system  
 230 is not affected by the isolation film. This can result in formation of charges on the surface of the  
 231 trapped droplets. The numerical analysis on the geometric model was performed using the similar  
 232 assumptions (i.e. continuous oil medium and infinite electrode height) and similar boundary  
 233 conditions given in Eqs. (8) and (9). Due to the significantly higher conductivity of water droplets  
 234 compared to that of oil (continuous phase), electrodes and water droplets together are considered  
 235 to be one conducting body and forms a new electrodes pair as soon as the water droplets are  
 236 trapped on the surface of electrodes. The trapped water droplets are therefore assumed to be a  
 237 protruding point out of electrode surface. The gravity force will move the formed water droplets

238 downward when the radius of the water droplets exceeds 0.1 mm [25]. Hence, this is the largest  
239 radius of water droplets that would be trapped on the electrodes and is considered in our  
240 simulation.

### 241 3.2 Results of numerical analysis

242 The numerical analysis to study the effect of voltage on the DEP force was carried out at an inter-  
243 electrode distance of 4 mm with four different voltages of 100, 150, 200 and 250 V. The DEP  
244 force represented as the gradient of the square of the electric field is shown in Figure 3. As shown  
245 in Figure 3, the area under the influence of DEP force increases with increasing voltage. The  
246 maximum gradients of the squared electric field were  $2 \times 10^{12}$ ,  $4.5 \times 10^{12}$ ,  $8 \times 10^{12}$  and  $1.3 \times$   
247  $10^{13} \text{ V}^2/\text{m}^3$ , for the applied voltages of 100, 150, 200 and 250 V, respectively. Moreover, for all  
248 the applied voltages, the gradient of the squared electric field intensity decays very rapidly away  
249 from the electrode surface, reaching approximately  $0 \text{ V}^2/\text{m}^3$  at a distance of 1.4 mm away from  
250 the electrode surface. This implies that the maximum DEP force is located in the vicinity of the  
251 surface of the electrodes, while the minimum and even null DEP force is located at the midpoint  
252 between the two electrodes.





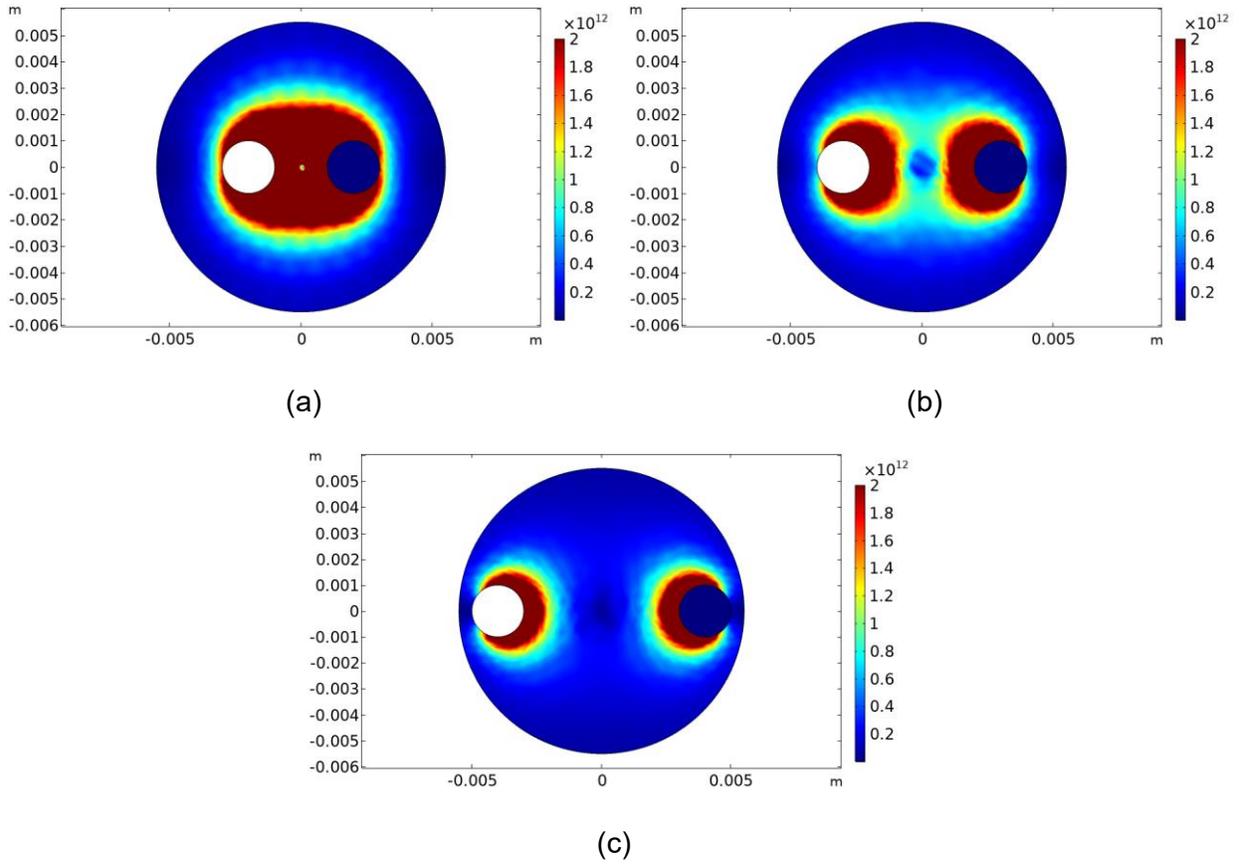
(e)

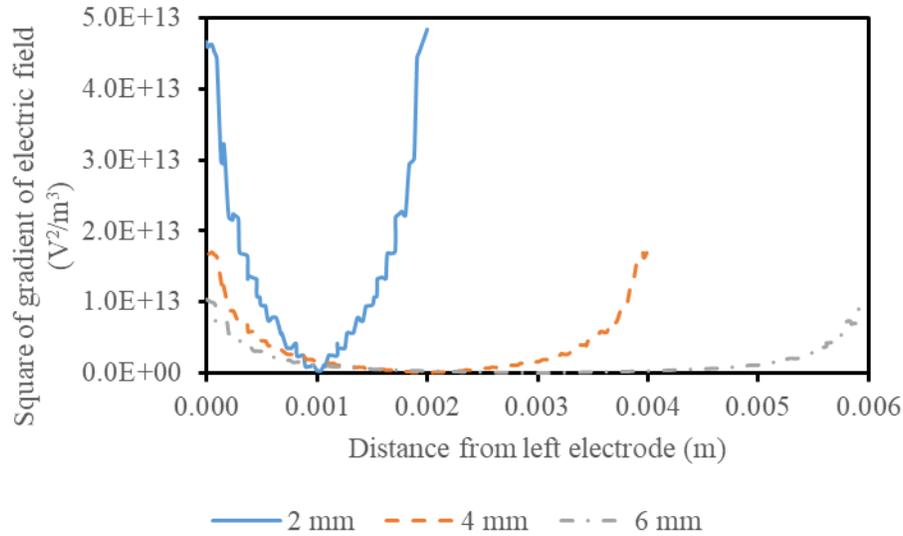
253 **Figure 3.** The DEP force field distribution defined as  $(\nabla|E|^2)$  for four applied voltages (a) 100 V (b)  
 254 150 V (c) 200 V (d) 250 V (e) Squared of the gradient of electric field intensity between the  
 255 electrodes at different applied voltages (inter-electrode distance 4 mm, frequency 200 kHz).

256 The numerical analysis to study the effect of the inter-electrode distance on the DEP force was  
 257 carried out at 250 V with three different inter-electrode distances of 2, 4 and 6 mm. The DEP force  
 258 represented as the gradient of the square of the electric field is shown in Figure 4. As shown in  
 259 Figure 4, the area under the influence of DEP force increases with decreasing the distance. The  
 260 maximum gradients of the squared electric field were  $0.9 \times 10^{12}$ ,  $1.2 \times 10^{13}$ , and  $3.75 \times 10^{13}$   
 261  $V^2/m^3$ , for inter-electrode distances of 6 mm, 4 mm, and 2 mm, respectively. Moreover, for all the

262 inter-electrode distances, the gradient of the squared electric field decays very rapidly away from  
263 the electrode surface, reaching approximately  $0 \text{ V}^2/\text{m}^3$  at a distance of 0.8 mm away from the  
264 electrode surface for an inter-electrode distance of 2 mm and at a distance of 1.3 mm away from  
265 the electrode surface for inter-electrode distances of 4 mm and 6 mm (Figure 4 (d)).

266





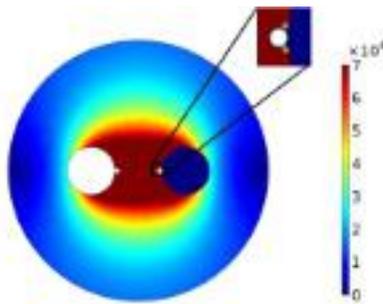
(d)

267 **Figure 4.** The DEP force field distribution defined as  $(\nabla|E|^2)$  for inter-electrode distances of (a) 2  
 268 mm (b) 4 mm (c) 6 mm (d) Squared of the gradient of electric field intensity between the electrodes  
 269 at different inter-electrode distance (applied voltage 250 V, frequency 200 kHz).

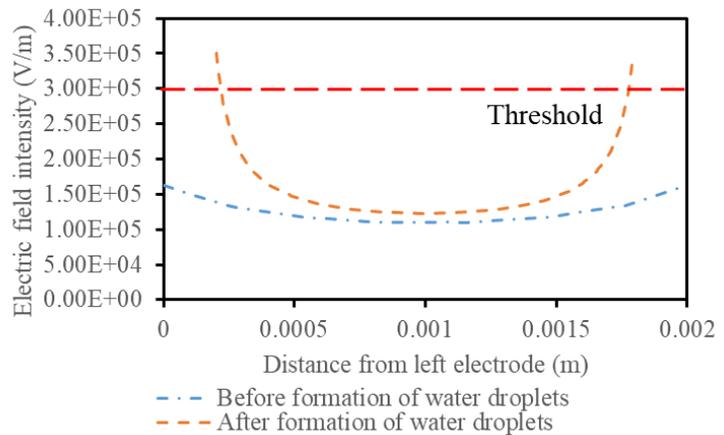
270 The influence of the entrapment of water droplets on the surface and between the electrodes on  
 271 the electric field strength was simulated and compared with the electric field strength generated  
 272 without water droplets trapped on the electrodes surface (Figure 5). The simulation was  
 273 performed at two different inter-electrode distances of 2 mm and 4 mm. It can be seen from Figure  
 274 5 that for the two inter-electrode distances, 2 mm and 4 mm, the electric field strengths with no  
 275 water droplets attached to the surface of the electrodes are less than those with water droplets  
 276 attached to the surface of the electrodes. The impact of the attached water droplets to the surface  
 277 of the electrodes on enhancing the electric field can be obviously seen as the protruded electric  
 278 field distribution shown in Figure 5 (c), and quantitatively demonstrated in Figure 5 (b) and (d).  
 279 The electric field strengths in both electrode arrays with water droplets trapped on the surface of  
 280 the electrodes are largely enhanced. The maximum electric field strengths were increased about  
 281 110% and 106% for inter-electrode distances of 2 mm and 4 mm, respectively. Compared to the

282 homogenous distribution of charges on the surface of the electrodes with no water droplets  
 283 trapped, more charges are aggregated at the tips of the water droplets, and hence inducing a  
 284 very inhomogeneously distributed electric field with the maximum electric field strength located at  
 285 the tips of the water droplets. Eow et al. suggested that an electric field intensity of 300 kV/m and  
 286 more would cause water droplets to break up and re-emulsify the system and therefore the reduce  
 287 the demulsification efficiency, the voltage that results in an electric field intensity of 300 kV/m is  
 288 therefore defined as threshold voltage [26]. It can be seen from Figure 5 (d) that even with the  
 289 trapped water droplets, the maximum electric field strength generated by the electrode array with  
 290 distance  $d$  of 4 mm is still far lower than the threshold electric field intensity. Therefore, the water  
 291 droplets although trapped on the electrodes surface are expected not to be broken up before they  
 292 settle down by gravitational force. However, the electric field strength in the vicinity of the water  
 293 droplets attached to the electrodes is higher than the threshold in the 2 mm distanced electrode  
 294 array (Figure 5 (b)). Therefore, we could expect a break-up phenomenon occurring and hence a  
 295 lower demulsification efficiency at an applied voltage of 250 V.

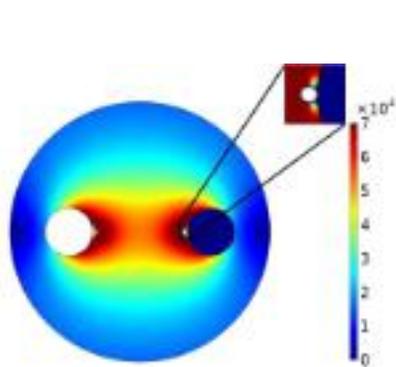
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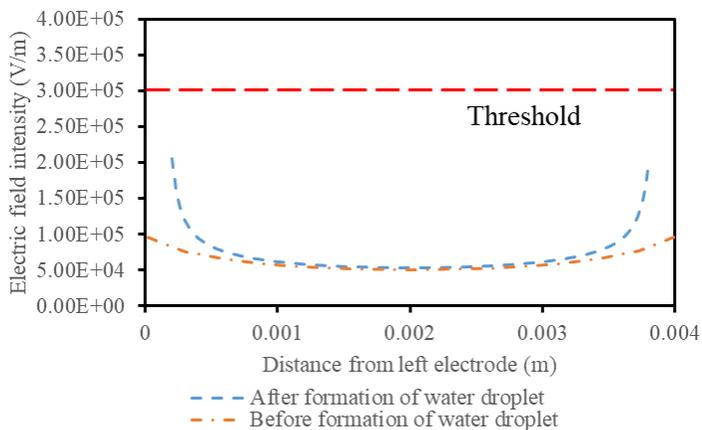
(a)



(b)



(c)



(d)

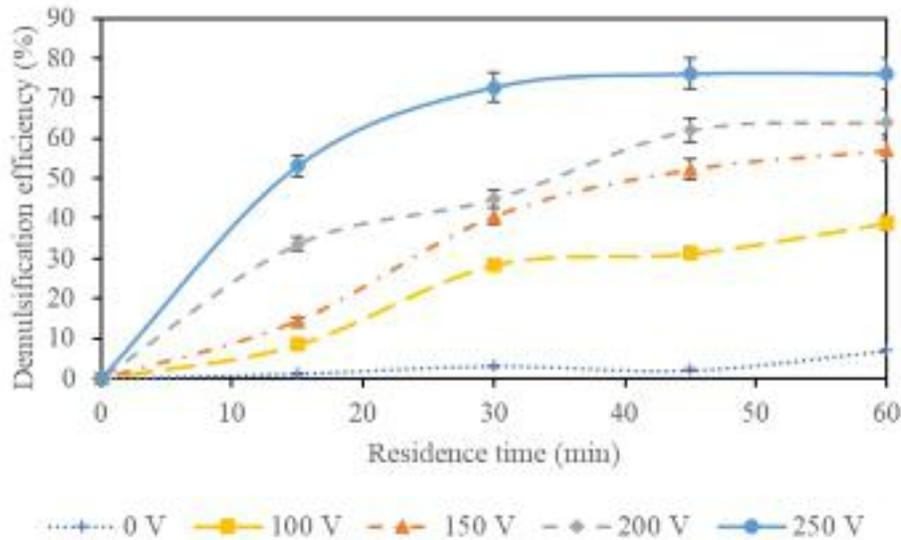
297  
 298 **Figure 5.** Comparison of electric field between two electrodes (white and blue large circles) with  
 299 and without water droplet (white small circles) trapped on electrodes surface with an inter-  
 300 electrode distance of 2 mm (a, b), 4 mm (c, d) (applied voltage 250 V, frequency 200 kHz).

## 301 4. Experimental results and discussion

### 302 4.1 Effect of electric field strength

303 The efficiency of electro coalescence is mainly dependent on the electric field strength, which  
 304 determines the velocity of water droplets and the intensity of their collision. Applied voltage and  
 305 inter-electrodes distance are two key parameters influencing the electric field strength. The  
 306 influence of voltage on the demulsification efficiency was experimentally examined with varied  
 307 voltages of 100, 150, 200 and 250 V, at a constant frequency of 200 kHz and an inter-electrode  
 308 distance of 4 mm. As shown in Figure 6, higher voltage presents better demulsification efficiency.  
 309 An applied voltage of 250 V presented the best demulsification efficiency, where after 60 minutes  
 310 of demulsification time the demulsification efficiency reached 76.1%, while the lowest  
 311 demulsification process occurred with the lowest applied voltage of 100 V. The improvement of  
 312 demulsification with increased voltage was also observed by Kim et al. (2002) and Ramadhan et

313 al. (2023) [27, 28]. Kim et al. (2002) increased the applied voltage from 1 kV/m to 5 kV/m and  
314 improved the demulsification efficiency by 28%, whereas Ramadhan et al. (2023) increased the  
315 voltage from 1000 V to 4000 V and improved the demulsification efficiency by 18%. The strength  
316 of the DEP force field is directly proportional to the gradient of squared electric field and hence  
317 the square of the applied voltage. At higher voltages it is expected that the water droplets in the  
318 emulsion will be more affected by the DEP force and thereby experience more coalesce [29]. That  
319 also explains why the demulsification efficiency at 250 V increases rapidly compared to the other  
320 applied voltages and achieves a relatively stable demulsification efficiency after 45 minutes. The  
321 DEP force is expected to promote frequent collision of the water droplets, due to these collisions,  
322 the water droplets coalesce forming bigger water droplets, when the radius of the droplets  
323 increases to 0.1 mm or more, the droplets will separate from oil under the influence of the  
324 gravitational force [25]. Moreover, it was noticed that the application of voltage was accompanied  
325 with an increase in temperature of the emulsion where the emulsion temperature increased from  
326 22°C to 34°C after 60 min when applying 250 V. This increase of temperature could facilitate the  
327 collision of water droplets and increase the efficiency of the demulsification process [30]. In  
328 addition, the fact that the maximum demulsification efficiency occurred while applying the highest  
329 voltage, implies that no water droplets break-up occurred. The simulation results showed that  
330 water droplets will not break-up at an applied voltage of 250 V, a frequency of 200 kHz, and an  
331 inter-electrode distance of 4 mm (Figure 5 (d)).



332

333 **Figure 6.** Effect of applied voltage on water removal efficiency (inter-electrode distance 4 mm,  
 334 frequency 200 kHz).

335 4.2 Effect of inter-electrode distance

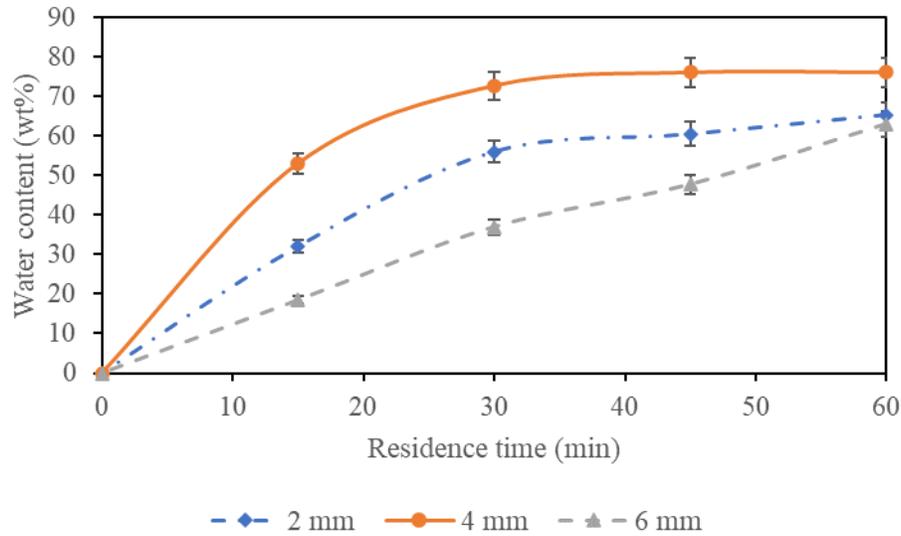
336 The effect of the inter-electrode distance on the demulsification efficiency was studied by  
 337 analyzing the demulsification efficiency at three different inter-electrode distances of 2 mm, 4 mm  
 338 and 6 mm. The applied voltage and frequency were fixed at 250 V and 200 kHz, respectively. As  
 339 shown in Figure 7, the demulsification efficiency at an inter-electrode distance of 4 mm was the  
 340 highest. Comparing the 4 mm inter-electrode distance to the 6 mm inter-electrode distance, the  
 341 induced electric field is stronger for the case of the shorter distance, which could enhance the  
 342 DEP force and hence the number of collisions of water droplets, thereby presenting higher  
 343 demulsification efficiency. According to the simulation results shown in Figure 4, the highest  
 344 electric field strength and hence the highest DEP force are generated at an inter-electrode  
 345 distance of 2 mm. However, contrary to the simulation results using an inter-electrode distance of  
 346 2 mm resulted in a lower demulsification efficiency compared to an inter-electrode distance of 4  
 347 mm. It can be seen from Figure 11 that at the 60<sup>th</sup> minute the demulsification efficiency for the 2  
 348 mm inter-electrode distance was 14% lower than the 4 mm inter-electrode distance. The very

349 strong DEP force when using the 2 mm inter-electrode distance allows water droplets around the  
350 electrodes to be rapidly moved to and trapped on the surface of the electrodes. Trapped water  
351 droplets, although still small, could enhance the electric field strength and the DEP force for  
352 attracting more water droplets on these already trapped water droplets and thereby growing very  
353 fast. As presented in the numerical analysis (Figure 5 (a) and (b)), as soon as the radius of the  
354 water droplets grows up to 100  $\mu\text{m}$ , the electric field intensity at the surface of the electrode  
355 increases by 109.58% and reaches 350 kV/m, which is much higher than the threshold of 300  
356 kV/m [26]. Due to this high electric field intensity, these 100  $\mu\text{m}$  water droplets will be ruptured  
357 immediately to release secondary much smaller water droplets [26]. Such re-generated small  
358 water droplets will re-emulsify the system and therefore the demulsification efficiency is largely  
359 reduced. In the case of the 4 mm inter-electrode distance the induced electric field is far lower  
360 than the threshold of 300 kV/m as shown in the simulation results in Figure 5 (c) and (d). The  
361 demulsification process and the accumulation of the water droplets on the surface of the electrode  
362 can be clearly seen in Figure 8. Figure 8 (a) shows how the water droplets are dispersed in oil  
363 before the application of the electric field while Figure 8 (b) shows how the water droplets formed  
364 are accumulated on the surface of the electrode after the application of the electric field.

365

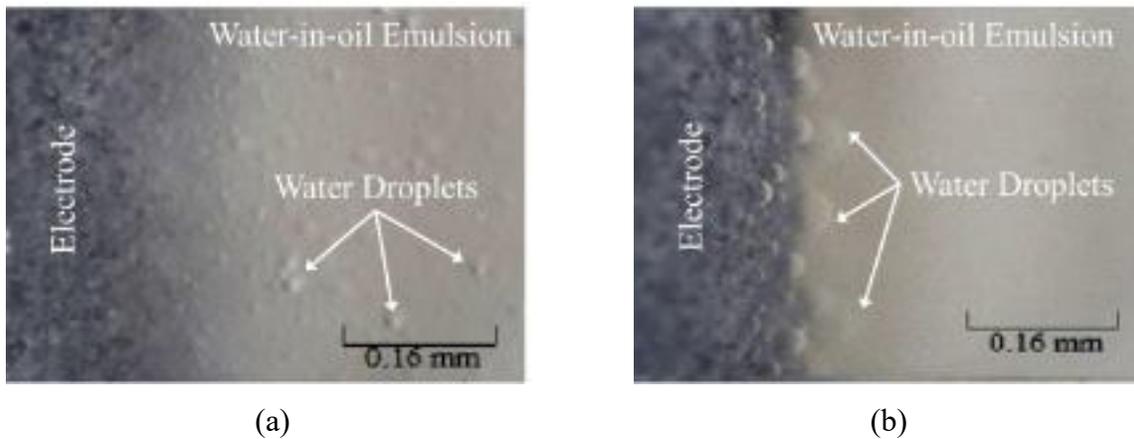
366

367



368

369 **Figure 7.** Effect of electrode distance on demulsification efficiency (applied voltage 250 V,  
 370 frequency 200 kHz).

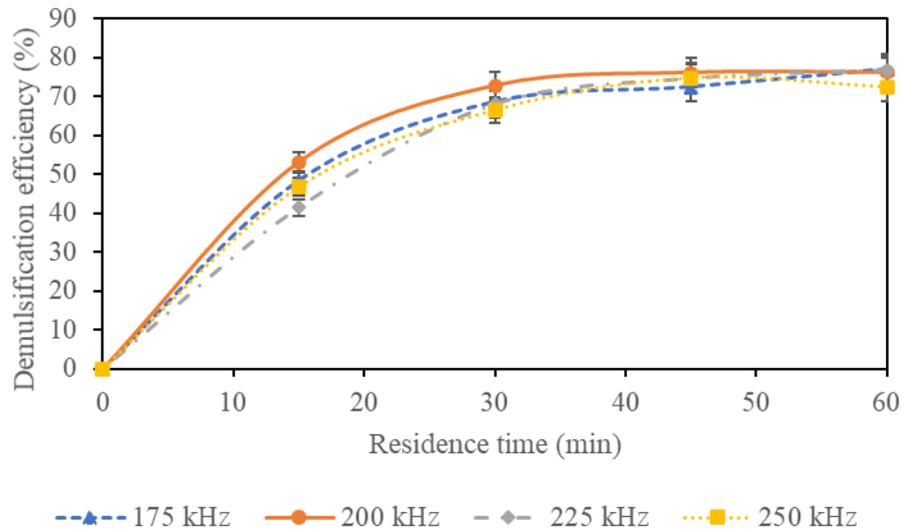


371 **Figure 8.** The water-in-oil emulsion (a) no applied electric field (b) with applied electric field with  
 372 an applied voltage of 250 V, frequency of 200kHz, and an inter-electrode distance of 4mm after  
 373 45 minutes demulsification time. (The brightness and contrast of the image has been digitally  
 374 improved)

#### 375 4.3 Effect of frequency

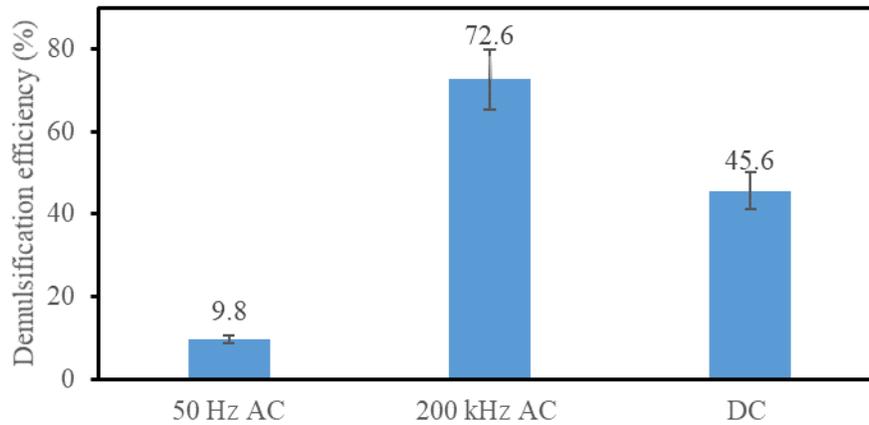
376 The effect of frequency on the demulsification efficiency was studied by analyzing the  
 377 demulsification efficiency at four different high frequencies of 175, 200, 225 and 250 kHz and

378 one low frequency of 50 Hz. The applied voltage and inter-electrode distance were fixed at 250 V  
379 and 4 mm, respectively. The application of high frequency is on one hand to avoid the  
380 electrochemical reaction on the surface of the metal electrodes and on the other hand to prevent  
381 the formation of the pearl-chain, which might induce a short circuit of the system and hence largely  
382 reduce the demulsification efficiency and even damage the demulsification system. It can be seen  
383 from Figure 9 that all examined high frequencies presented very similar tendency where the  
384 difference on the demulsification efficiencies is very insignificant. However, at a very low  
385 frequency of 50 Hz, the demulsification efficiency was poor at only 9.8% compared to a maximum  
386 demulsification efficiency of 72.6% at a frequency of 200 kHz (Figure 10). In the literature, Kim et  
387 al. (2002) and Ramadhan et al. (2023) also observed improved demulsification efficiency at  
388 elevated frequencies of 1000 Hz and 1500 Hz, respectively [27, 28]. Ramadhan et al (2023)  
389 observed a 55% improvement in demulsification efficiency while increasing the frequency from  
390 50 Hz to 2000 Hz, whereas Kim et al. (2002) obtained 13% improvement in demulsification  
391 efficiency by increasing frequency from 50 Hz to 2000 Hz. Frequency is believed to be one of the  
392 most important parameters that would affect the chain formation of water droplets during  
393 demulsification [31]. With very high frequency, the charges on the water droplets will not have  
394 sufficient time to respond to the electric field and hence the water droplets will migrate for  
395 coalescence. In addition, when compared to DC voltage, the demulsification efficiency obtained  
396 by applying high frequency AC voltage was 37% higher. This indicates that in DC the  
397 demulsification process is dominated by electrophoresis and the dielectrophoretic force is only  
398 dominant when higher frequency AC voltage is applied. Moreover, the better demulsification  
399 performance of AC voltage at 200 kHz compared to DC is attributed to the induction of more  
400 droplet-droplet collision at high frequency AC voltage and prevention of pearl chain formation [26,  
401 32].



402

403 **Figure 9.** Effect of AC frequency on water removal efficiency (inter-electrode distance 4 mm,  
 404 voltage 250 V).



405

406 **Figure 10.** Effect of frequency and different types of power supply on demulsification efficiency.

407 4.4 Energy analysis

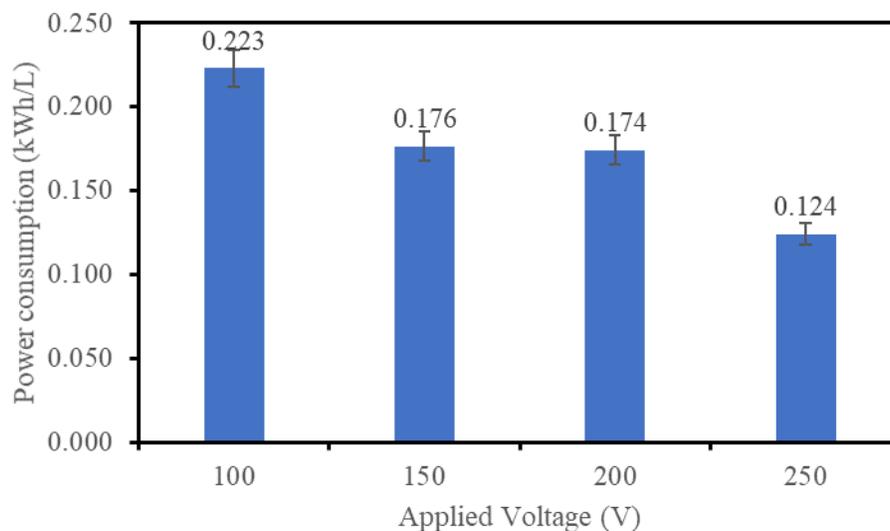
408 The specific energy consumption of the demulsification process was calculated as:

$$E = \frac{U \times I \times t}{1000 \times V} \quad (15)$$

409 Here, E is the specific energy requirement of the process (kWh/L), U is voltage (V), I is current  
410 (A), t is time (h) and V is the volume (L) of the water-in-oil emulsion. 1000 is the conversion factor  
411 used to convert watt-hr to kilowatt-hr. The voltage (U) and current (I) were continuously measured  
412 using an oscilloscope. The specific energy consumption was calculated for an inter-electrode  
413 distance of 4 mm and a frequency of 200 kHz. In order to compare the energy consumption at  
414 different applied voltages, the maximum demulsification efficiency of the lowest applied voltage  
415 was used as the base line. As shown in Figure 6 a maximum demulsification efficiency of 39%  
416 was achieved after 60 minutes with an applied voltage of 100 V. The same demulsification  
417 efficiency was achieved after 30, 15 and 8 minutes with an applied voltage of 150, 200 and 250  
418 V, respectively. Figure 11 shows that the specific energy consumption for a demulsification  
419 efficiency of 39% was 0.223, 0.176, 0.174, and 0.124 kWh/L at an applied voltage of 100, 150,  
420 200, and 250 V, respectively. The lowest specific energy consumption was achieved with the  
421 highest applied voltage due to the much lower time required to reach the targeted demulsification  
422 efficiency. Table 3 shows the specific energy consumption of different demulsification strategies  
423 that are being developed. As seen in Table 3, the specific energy consumption obtained from this  
424 study is 4.0, 54.8 and 16.9 times lower than the specific energy consumption of centrifugation,  
425 microwave irradiation and electrocoagulation based demulsification processes, respectively [33-  
426 35].

427

428



429

430 **Figure 11.** Specific power consumption at different applied voltages (39% demulsification  
 431 efficiency, inter-electrode distance 4 mm, frequency 200 kHz).

432 **Table 3.** Comparison of energy consumption of different technologies.

Demulsification Technology	Specific energy consumption (kWh/L)	Reference
Miniaturized centrifugal demulsification device	0.49	[35]
Microwave irradiation assisted demulsification	6.80	[34]
Parallel-plate electrocoagulation	2.1	[33]
Electro-coalescence using TiO <sub>2</sub> coated electrodes	0.124	This study

433

#### 434 4.5 Comparison with previous studies

435 Table 4 shows comparison of electro-coalescence performance between this study and other  
436 studies. As seen from Table 4, the 76% demulsification efficiency obtained from this study is one  
437 of the highest. Higher demulsification efficiency of 82.5% was obtained by Hu et al. (2021) for an  
438 emulsion containing 90 v% water [36]. The higher water content means higher number of water  
439 droplets to collide and coalesce which resulted in higher demulsification efficiency in the study.  
440 For a water content of 20%, Kim et al. (2002) designed and used a hydrostatic demulsifier to  
441 obtain 90% demulsification efficiency while using 200 ppm Betzdearborn as chemical demulsifier  
442 [27]. The improved demulsification efficiency in the study is attributed to the chemical demulsifier  
443 since the same study reported 0% demulsification without addition of demulsifier. Compared to  
444 other studies that has achieved greater than 70% demulsification efficiency, the current study  
445 used lower applied voltage and demulsification time. Ramadhan et al. (2023) obtained 75%  
446 demulsification efficiency within 60 mins of demulsification time, however, the reported applied  
447 voltage was 4000 V [28]. On the other hand, Taleghani et al. (2019) needed 50 hours to achieve  
448 58% demulsification efficiency [37].

450 **Table 4.** Comparison of electrocoalescence performance with previous studies.

Emulsion	Applied voltage	Frequency	Demulsification Time	Demulsifier	Demulsification efficiency	Reference
20 v% water 80 v% crude oil	2.5 kV/cm	60 Hz	3 min	75 ppm Betzdearborn	60%	[27]
		60 Hz	3 min	N/A	0%	
32.2% water 13.9 v% light oil 30.3 v% heavy oil 23.6 v% solids	2.05 V/cm	N/A	50 hrs	N/A	44%	[37]
		N/A	50 hrs	3452 ppm FeCl <sub>3</sub>	58%	
		N/A	50 hrs	3452 ppm alum	17%	
10 v% water 90 v% castor oil	4000 V	1500 Hz	60 mins	N/A	20%	[28]
10 v% water 90 v% soyabean oil						
90 v% water 10 v% kerosene	150 V	1000 Hz	11 min	N/A	82.5%	[36]
4.71 wt% water 95.29 wt% lubricant	250 V	200 kHz	45 min	N/A	76%	This work

## 452 5. Conclusion

453 In this work we numerically simulated and experimentally demonstrated the influence of electric  
454 field on the demulsification efficiency by varying the electric field determining parameters  
455 (including voltage, frequency and distance between electrodes). It was found that trapped water  
456 droplets on the electrodes surface could largely influence the electric field strength, and hence  
457 the demulsification process. The entrapped water droplets become a part of the electrode and  
458 thereby forming a new electrodes pair. The new electrodes pair has a shorter inter-electrode  
459 distance and therefore stronger electric field strength is generated with unchanged applied  
460 voltage. With the growth of the water droplets due to the electrocoalescence, the inter-electrode  
461 distance is further reduced and the induced electric field increases. When the induced electric  
462 field strength is below the threshold of 300 kV/m, the demulsification efficiency could be enhanced  
463 by increasing the electric field either through applying higher voltage or reducing the distance  
464 between electrodes. However, if the induced electric field is above 300 kV/m, the trapped water  
465 droplets will be broken up, and hence releasing many much smaller water droplets, which re-  
466 emulsifies the system and hence largely reduces the demulsification efficiency. We numerically  
467 simulated and experimentally validated such an effect using a 2 mm distanced electrodes array  
468 with the application of voltage of 250 V and frequency of 200 kHz. Due to the trapped water  
469 droplets on electrodes, the electric field was largely increased about 109.58% to 350 kV/m far  
470 beyond the threshold and hence resulted in the lower demulsification efficiency of 65%.

471 With this new understanding, a maximum demulsification efficiency of 76% was obtained by  
472 optimally applying the parameters of 250 V for voltage, 200 kHz for frequency, 4 mm for the inter-  
473 electrode distance, in a 45 min process with the most efficient energy consumption of 0.51 kWh/L.  
474 The application of high frequency and the utilization of insulation are suggested for avoiding of

475 the electrochemical reaction on the electrodes and preventing from the generation of gas bubbles  
476 as well as the pearl-chain formation of water droplets.

477 We found that the demulsification efficiency using high frequency AC electric field was 37% higher  
478 than when using DC field with other parameters constant. This is due to the much stronger  
479 collision and hence coalescence between water droplets driven by the DEP force in AC electric  
480 field with high frequency compared to that induced by EP force in DC electric field.

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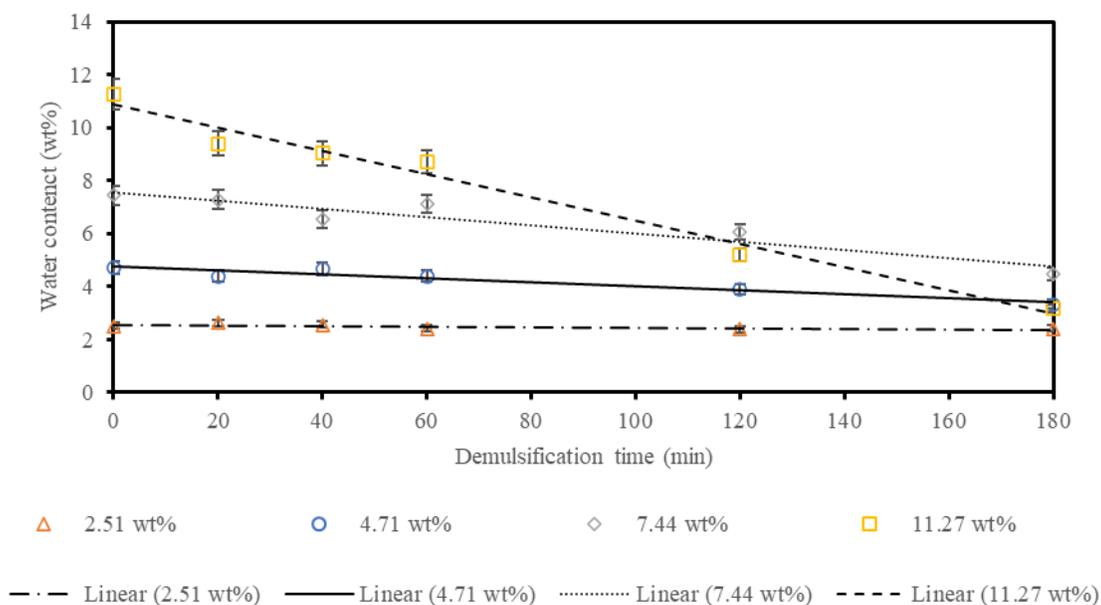
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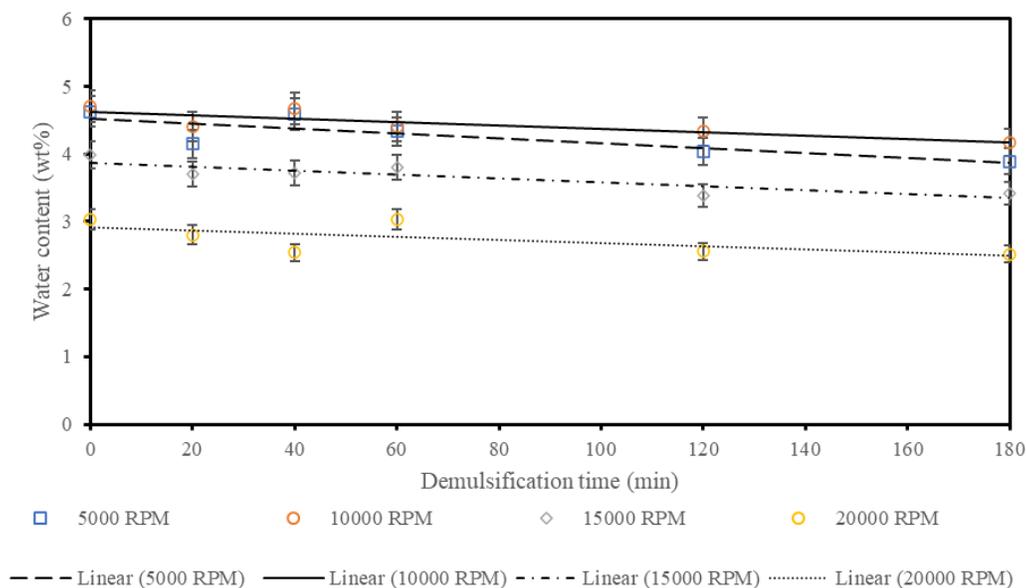
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617 Supplementary Data



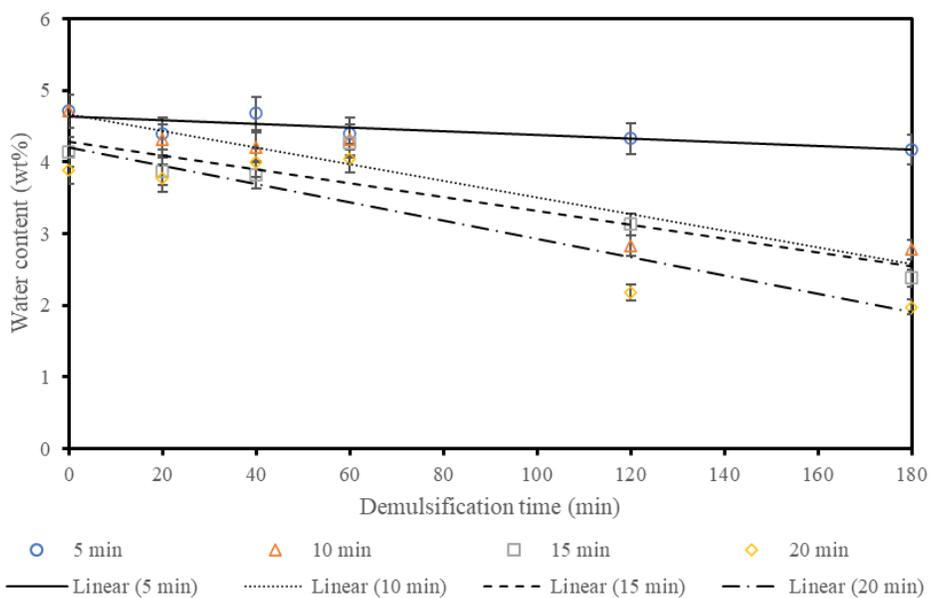
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619 **Figure S1.** Effect of water content on water-in-oil emulsion stability (10,000 RPM, 5 min).



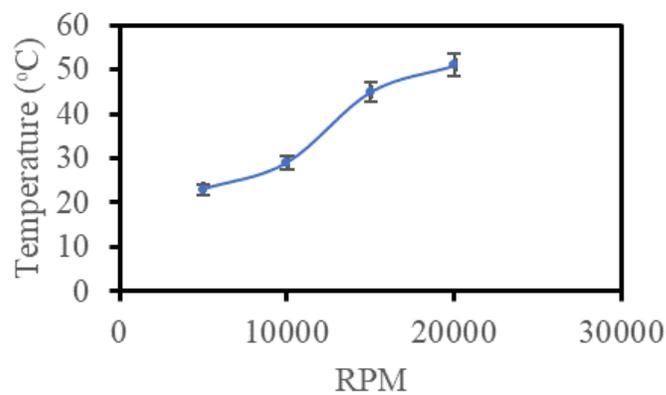
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621 **Figure S2.** Effect of dispersion speed on water-in-oil emulsion stability (5 mins, 4.71 wt%).

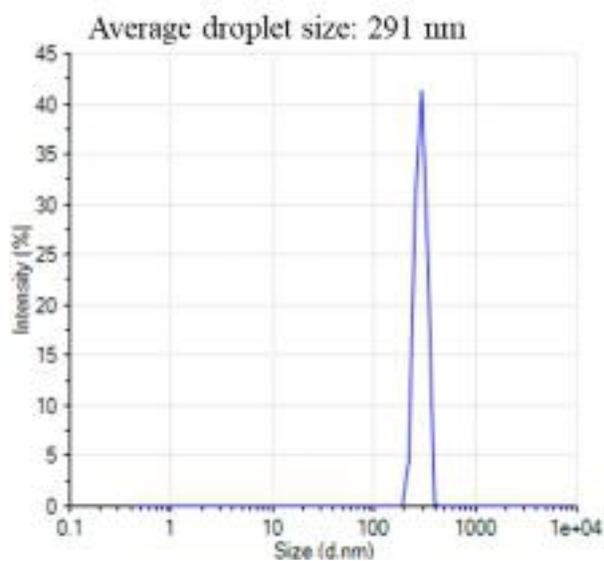


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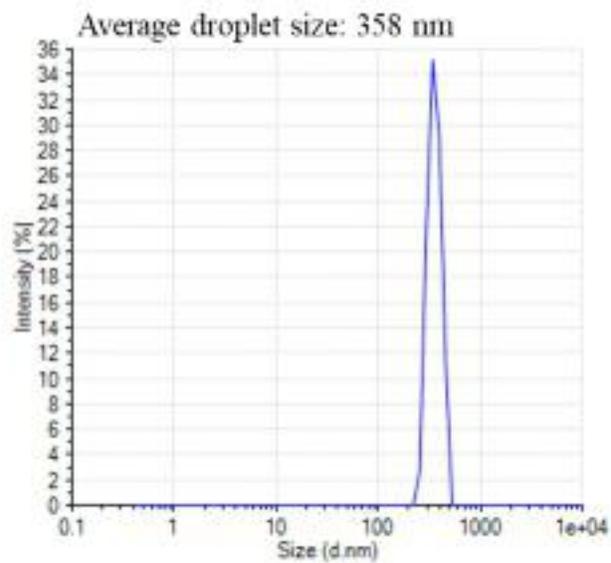
623 **Figure S3.** Effect of dispersion time on water-in-oil emulsion stability (10,000 RPM, 4.71 wt%).



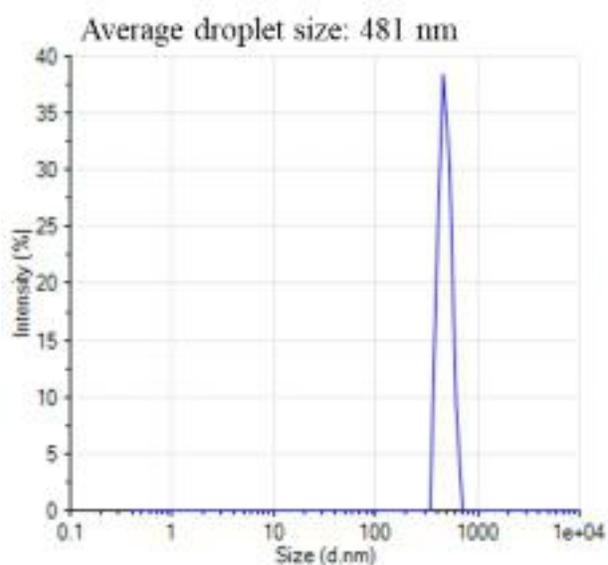
624 **Figure S4.** The temperature of the emulsion after 5 min of emulsification at different dispersion  
 625 speeds.



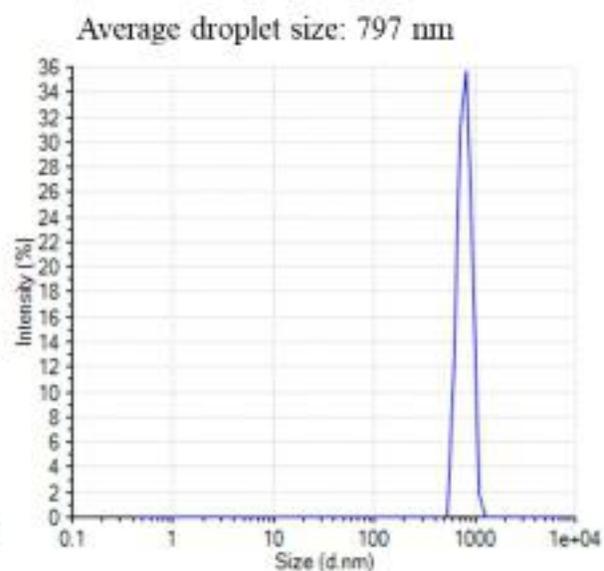
(a)



(b)

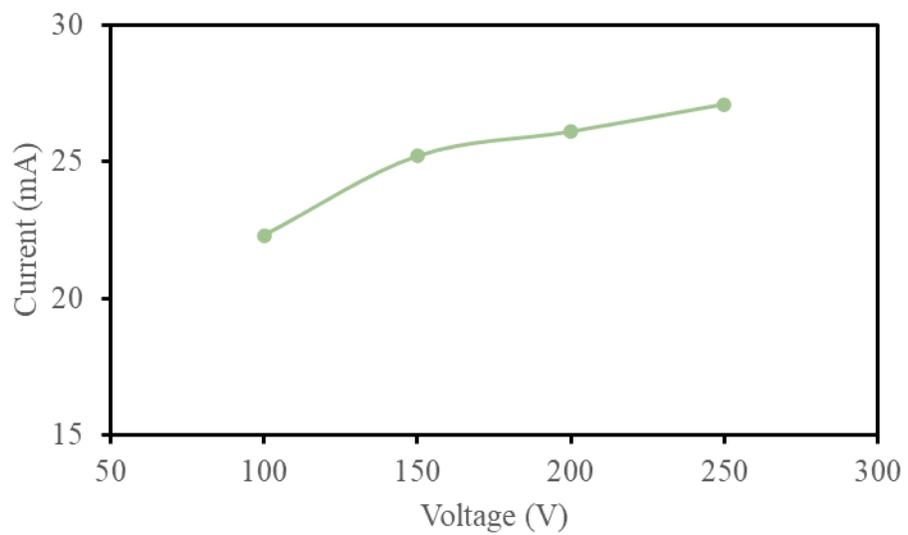


(c)



(d)

626 **Figure S5.** Water droplet size distribution for water-in-oil emulsion with (a) 2.15 wt%, (b) 4.71  
 627 wt%, (c) 7.44 wt% and (d) 11.27 wt% water.



628

629 **Figure S6.** Current voltage characteristics of the system.

630