# **The influence of low frequency electric field on the coalescence of water drops in emulsion shear flow**

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#### **ABSTRACT**

Crude oil often mixes with water to form water-in-oil emulsions as a result of factors such as high shear at the production wellhead and surfactants that are naturally present in crude oil. The present study is aimed at determining the conditions leading to electrocoalescence of water droplets in the flow of the emulsion. An important part of this preliminary work concerns the creation of an experimental cell and installation designed to study the massive coalescence of droplets in a shear flow of an emulsion under the action of an applied inhomogeneous alternating electric field. The flow of the emulsion through the cell is created by a syringe pump. We determine the effect of exposure by the volume of the separated phases of the emulsion in the drainage tank. Further research will be related to the study of various emulsions with varying concentration and flow rate and possibly the frequency of the applied field. The research results will be useful for creating devices for electrocoalescence in the flow.

#### **KEYWORDS**

electrocoalescence • nonuniform • electric field • ITO glass • photolitography

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# **Introduction**

Refining crude oil often requires the extraction of large amounts of water. Crude oil often mixes with water to form water-in-crude emulsions as a result of factors such as high shear at the production wellhead and surfactants that are naturally present in crude oil  $[1,2]$ . These emulsions are undesirable and required emulsification to remove dispersed water and associated inorganic salts in accordance with manufacturing and shipping specifications. In addition, demulsifying these crude oil emulsions reduces corrosion and catalyst poisoning and invariably maximizes the overall profitability of crude oil recovery. Recently, there has been a growing research interest in developing effective solutions to the problems associated with the transportation and processing of crude oil emulsions, as well as restrictions on the discharge of produced water  $[3,4]$ . There are various methods for the separation of emulsions  $[5-7]$ . Moreover, a more efficient demulsification process can be achieved through the use of synergistic effects by combining one or more of these methods. The undisputed favorite among them is the electric method. This method includes both exposure to direct current, alternating (LF, RF, MW) field  $[8-10]$ . Electrical methods can be used at any stage of emulsion separation. However, it is advisable to use these methods immediately before or during centrifugation because it obtains the greatest efficiency. Most of the work is aimed at studying electrocoalescence at low frequencies. In electrocoalescence, an electric field is

applied to a dispersion of conductive water droplets in a poorly conductive oil to force the droplets to coalesce in the direction of the field [\[11\].](#page-6-5) As a rule, the behavior of coalescence can be described in three stages: the droplets approach each other, the film thinning / drying process, and film rupture, leading to the coalescence of droplets-droplets. However, there are other mechanisms, such as the formation of droplet chains, dipole – dipole coalescence, electrophoresis, dielectrophoresis and random collisions. The type of electric field, such as alternating, direct and pulsed direct current, plays a significant role depending on the design and setup of the system. Other factors, such as the average droplet size and the residence time of the liquid mixture under the influence of the electric field, stand out due to the efficiency of coalescence [\[12\].](#page-6-6) One of these is the conductivity of the water phase. Dielectric parameters are important characteristics, which depend both on the content of the dispersed phase and on the parameters of the electromagnetic field  $[2,13]$ . Experiments have been reported using silicone oils, vegetable oils and hydrocarbons in a droplet fixer that allows the use of low viscosity oils  $[14,15]$ . Another factor is the nature and concentration of surfactants at the drop-oil interfac[e \[16,17\].](#page-6-9) It was found that the time of electrocoalescence decreases with increasing interfacial tension. This study shows that increasing the interfacial tension gradient will shorten the coalescence time, which is important for improving the mixing efficiency of the droplets  $[18,19]$ . The influence of oil viscosity and energy consumption on the rate of coalescence was also investigated [\[20,21\].](#page-7-1) In this case, the viscosity of the dispersed system depends on both temperature and the content of the dispersed phase [\[22,23\].](#page-7-2) Much attention is paid to the numerical modeling of electrocoalescence using molecular dynamics (MD) modeling [\[24\].](#page-7-3) The results showed that the merging of droplets depended on the strength of the electric field [\[25\].](#page-7-4) The deformation of a water drop in a dielectric oil phase in the presence of external pulsating electric fields is numerically analyzed by the finite element method [\[26\].](#page-7-5) Proprietary software Comsol Multiphysics is used for modeling.

# **Materials and Methods**

## **Emulsion preparation**

An emulsion consisting of water microdroplets suspended in tetradecane was chosen as the object of the study (Fig. [1\)](#page-1-0). Deionized water, purified by the Milli-Q Advantage A10 system (EMD Millipore), was emulsified in tetradecane C (Reachim) using a stirrer.

<span id="page-1-0"></span>

**Fig. 1.** Emulsion microstructure

To stabilize the emulsion, a nonionic surfactant, sorbitanmonooleate (SPAN 80, Sigma-Aldrich), at a weight concentration of 0.5 % was previously dissolved in tetradecane.

Using digital image processing in the ImageJ program, the diameters and the number of droplets were obtained and a graph of the probability density of the droplet size distribution was plotted. The data were fitted with a log normal distribution, from which an average droplet size of 9  $\mu$ m was determined.

#### **Experimental setup**

For the research, a laboratory setup (Fig. [2\)](#page-2-0) was assembled based on an Olympus IX71 optical microscope, with an integrated high-speed camera Photron FASTCAM SA5. The experimental cell is installed on the table of an optical microscope. An Agilent 33522A signal generator is used to generate an electric field in the cell, followed by amplification through a Tabor Electronics 9100 high-voltage low-frequency amplifier.

<span id="page-2-0"></span>

**Fig. 2.** Laboratory setup diagram: 1 - microscope; 2 - camera; 3 - signal generator; 4 - amplifier; 5 - experimental cell

The cell for electrocoalescence was placed on the stage of an optical microscope. The cell is a sandwich of two glasses between which a Teflon gasket 100 µm thick was installed in which a channel was cut. Glass with an electrically conductive layer of indium tin oxide ITO (Indium TinOxide) was used as the lower substrate, on the surface of which sawtooth electrodes with a distance of 200 µm between the tops were etched by the photolithography method. An alternating bipolar voltage was applied to the electrode system from a 33522A arbitrary waveform generator (Agilent Technologies), amplified with a Tabor 9100 amplifier (Tabor Electronics Ltd.).

## **Experimental cell**

The main part of the laboratory setup is an experimental cell, which is a sandwich of two glasses with a Teflon gasket 100 µm thick between them. A channel 1 mm wide and 10 mm long is cut into the Teflon gasket. Holes were made on the upper substrate for the movement of the liquid under study. Electrically conductive glass with an indium-tin oxide layer (ITO glass) was used as the lower substrate, on the surface of which a sawtooth microelectrode system was etched using the photolithography technique. For this, a negative photoresist (Allresist AR-N 4400-50) was applied to the glass using a

centrifuge for smooth distribution of the substance (Spin 150) at 1000 rpm for 120 sec. After that, the photoresist layer was dried in an oven (WiseStir MSH - 20D) at 90 °C for 90 min under a laminar flow cabinet. Next, the stage of exposure under ultraviolet light was performed through a film mask, which was made by the method of photo output. Thereafter, the glass substrate was heated in an oven at 100 °C for 10 min to re-cure. After the steps described above, the non-exposed areas of the photoresist were washed off. The next stage of manufacturing involves etching an electrically conductive layer not protected by a photoresist; for this, a 10 % aqueous solution of hydrochloric acid was prepared, where a glass substrate with a previously applied thin layer of zinc was placed. Then, after etching the electrically conductive layer, the areas of the exposed photoresist were removed using methylpyrrolidone. After that, the required configuration of microelectrodes was obtained on the glass surface. To protect the electrodes, a protective layer of electrically insulating acrylic varnish (PLASTIK-71) was applied to the glass surface. To connect the microelectrodes to the amplifier output, a place was left, and then elastic conductive tape was glued. After that, the cell was assembled by gluing the glasses together using epoxy glue.

The measuring cell is a flat channel of rectangular cross-section without geometric features (Fig. [3\)](#page-3-0). Electrodynamically, the complexity of the design is due to the presence of specially shaped electrodes on the inner wall of the channel. The electrodes are located in a local area at the entrance to the channel to monitor electrocoalescence. When the emulsion flows through the channel, firstly it enters the area of action of the electric field, then continues to flow through the channel without features.

<span id="page-3-0"></span>

**Fig. 3.** Schematic of manufacturing the lower substrate of the experimental cell (left) and a system of electrodes with an interelectrode distance of 200 um (right)

## **Results**

The emulsion (1 ml) was pumped through the cell, where it was exposed to an inhomogeneous alternating electric field with a frequency of 500 kHz and a voltage of 300 V for all tests. At the exit, as mentioned earlier, the emulsion was collected in a tared drainage container. After passing the emulsion through the cell, we obtained separate phases of water and oil. Photographs of this process at various volumetric flow rates  $(1, 8 \text{ and } 16 \text{ ml } / \text{ h})$ , which correspond to speeds 10, 80, 160  $\mu$ m/s obtained with a highspeed camera are shown in Figs.  $4-6$ . The survey was carried out in the area where the electrodes were applied. The vertical line shown in Figs. 4[–](#page-4-0)6 indicates the boundary of areas with electrodes (left part) and without electrodes (right part).

<span id="page-4-0"></span>

**Fig. 4.** The effect of an electric field on a water-in-oil emulsion at a volumetric flow rate of 1 ml/h (speed 10 µm/s). There are no electrodes to the left of the green dividing line, and an area with electrodes to the right

<span id="page-4-1"></span>

**Fig. 5.** The effect of an electric field on a water-in-oil emulsion at a volumetric flow rate of 8 ml/h (speed 80 µm/s). There are no electrodes to the left of the green dividing line, and an area with electrodes to the right

<span id="page-4-2"></span>

**Fig. 6.** The effect of an electric field on a water-in-oil emulsion at a volumetric flow rate of 16 ml/h (speed 160 µm/s). There are no electrodes to the left of the green dividing line, and an area with electrodes to the right

As you can see in Fig. [4.](#page-4-0) the emulsion moves from left to right, and upon reaching the electrode zone, water droplets begin to coalesce, falling out into the free phase. In the case of a flow rate of 2 ml/h, small droplets are combined into large droplets of the order of 200–300 µm.

Increasing the flow rate up to 8 and 16 ml/h (Figs. [5](#page-4-1) and [6\)](#page-4-2), the process is identical, but the coalescence of drops is worse due to the high speed of the drops, and in the electrode region, large drops are strongly deformed, and these drops are pulled out and broken into secondary drops. This is the reason for the low rate of separated water at a flow of 16 ml/h.

Based on the test results, the dependence of the separated water on the volumetric flow rate of the emulsion liquid through the cell was built. After passing through the cell, the liquid was drained into the drainage chamber and settled for 24 hours. The dependence is built on the basis of assessing the levels of liquids in the drainage chambers after settling during the day.

Figure [7](#page-5-0) shows the dependence of the water separation on the liquid flow rate through the cell. At a volume flow of 1 ml/h, 30 % of water was separated, at 2 ml/h  $-$ 25 %, at 4 ml/h – 18 %, at 8 ml/h – 8 % and at 16 ml/h - less than 3 %. With an increase in the volumetric flow rate of the liquid, the drops do not have time to fully combine with each other, as in the case of 1 ml/h.

<span id="page-5-0"></span>

**Fig. 7.** Water separation from the emulsion vs volumetric flow rate

## **Conclusions**

Based on the experiments results, we can say that this method of emulsion separation is applicable. The electric field acts on the water droplets, which leads to their coalescence in the interelectrode space. Water droplets, passing through the cell, combine and thereby increase in size, which leads to the separation of the emulsion into its constituent phases. The difference in the amount of water remaining is that as the volumetric flow rate increases, the flow rate increases, which leads to high shear flow. In this case, the drops are strongly deformed, and the connection may not occur. The formation of secondary droplets was also observed due to the strong deformation of large droplets, which consisted in stretching along the flow and disintegrating into several small droplets. We assume that this method is appropriate because it does not require a lot of time, the use of special chemical demulsifiers, and at the exit from the cell, you can immediately get separate phases from each other. The use of this method in combination with centrifugation and the use of chemicals will increase the efficiency of emulsion separation by more than 90 %. However, the value of the indicator will greatly depend on the composition of the original emulsion and requires research in each specific case. Unfortunately, in the experiment, the concentration of emulsion droplets was not controlled, this imposes restrictions on the study of a couple of droplets or single droplets, even though we present photographs of the exposure process obtained using a microscope.

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