Applying ultrasonic fields to separate water contained in medium-gravity crude oil emulsions and determining crude oil adhesion coefficients

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ABSTRACT

Separating produced water is a key part of production processing for most crude oils. It is required for quality reasons, and to avoid unnecessary transportation costs and prevent pipework corrosion rates caused by soluble salts present in the water. A complicating factor is that water is often present in crude oil in the form of emulsions. Experiments were performed to evaluate the performance of ultrasonic fields in demulsifying crude oil emulsions using novel pipe-form equipment. A horn-type piezoelectric ultrasonic transducer with a frequency of 20 kHz and power ranging from 80 W to 1000 W was used for experimental purposes. The influences of the intensity of ultrasonic fields, ultrasonic irradiation time, and the initial water content of crude oils were evaluated to establish the rate of water segregation from oil. The experiments applied ultrasonic-field intensities of 0.25 W/cm³, 0.5 W/cm³, 0.75 W/cm³ and 1 W/cm³ to synthetic emulsions with 10%, 15%, 20%, and 25% of the water in crude oil. Crude oil demulsification occurred for each ultrasonic field intensity tested for all the samples tested. Function β involving adhesion coefficients was expressed in terms of wave-field intensity and initial concentration of water in each of the three crude oil samples tested. The experiments demonstrated that despite the absence of any chemical demulsifier involved, water separation caused by applying ultrasonic fields was effective and occurred rapidly. As the intensity of the ultrasonic field applied increased, the amount of water segregated from crude oil also increased. Subjected to constant field intensity, higher initial water cuts (up to 15% or so) in the crude oil samples and higher ultrasonic irradiation times, resulted in greater segregation of water from crude oil in percentage terms. However, in samples with initial water cuts of 20+% long irradiation times (~5 min), resulted in a decline in water separation compared to 2-min tests. Ultrasonic field treatments offer commercially-viable and environmentally-friendly alternatives to treatments using chemical demulsifiers as they reduce desalination requirements of wastewater.

1. Introduction

Crude oils, as they are produced, are complex mixed fluids, associated with salt-water often in the form of persistent emulsions that are difficult to breakdown. Some of these water-in-oil emulsions are actually developed during crude oil handling at the surface as it passes through pipework and valves and its pressure and temperature changes. In addition to becoming bound up in emulsions, some of the produced water becomes fixed in other components that natively exist in crude oil, such as waxes, resins, asphaltenes, and solids [1–4].

During crude-oil production, its association with water is both undesired and costly, particularly during midstream transportation and storage in pipelines, terminal tanks, and ships [1,5]. Water-free crude oil is a requirement for efficient pipeline flow and refining processes.
The water and salt contents of crude oil need to be minimized before it can be efficiently refined. Breaking water-in-oil emulsions are often required to achieve sufficiently low water contents in crude oil sold to refineries; that means reducing the water cuts of crude oils to less than 0.5% weight percent [3,5–7]. Breaking down water-in-oil emulsions in crude oil can be difficult, requiring complex treatments for large scale processing plants. A broad range of novel technologies and their applications, introduced in recent years, plays a substantial role in improving standards of living and access to energy and resources, such as CO2 emission [8], energy resources [9], enhanced oil recovery [10], iron corrosion [11], bioreresources [12–14].

Demulsifying crude oils can be achieved in laboratory and field scales [15,16] by various processes, which in addition to dosing them with chemical demulsifiers, include treatments involving: centrifuges [17], filtration through membranes [18], and microwave irradiation [19,20]. These demulsification procedures demonstrate high efficiency when applied to light crude oils. However, as crude oils become denser with longer-chain hydrocarbon molecules making them more viscous, these demulsification processes become less efficient and often require the addition of chemical demulsifiers to be effective.

Various methods have been proposed and tested in the past two decades to exploit ultrasound to enhance water-in-oil demulsification [21–27], and oil in produced water emulsification [17], some of which have been patented [28]. More recently, some ultrasound methods have been scaled up for field-scale processing plant applications [22,29–34]. Specifically, for demulsification operations, two ultrasound effects have been identified as influencing the separation of water from oil. These are the production of standing waves and cavitation [21,28,30]. Cavitation depends on how ultrasound treatments are applied and can either enhance droplet formation in fluids involving liquid phases that are immiscible or indeed stimulate emulsification [21]. Cavitation in powerful ultrasound treatments can cause larger drops to disperse into smaller droplets by micro-jet activity fracturing the drops leading to fragmentation [36]. On the other hand, in certain conditions and fluid types, cavitation can stimulate the coagulation of drops instead of their fragmentation [36], thereby enhancing water and crude oil separation [21]. The bulk viscosity of the emulsified oil-in-water fluids and the emulsion droplet size play a role in the efficiency of ultrasonic wave fields in separating oil and water [37].

Ultrasonics has been successfully used in demulsification mixtures of water with natural plant oils and hydrocarbon-based solvents as well as crude oil [21,25,37]. Several advantages of ultrasonic methods have been reported by researchers. One of the advantages of ultrasonic methods is the reduction of the emulsification time and the formation of smaller droplets compared with that generated by the conventional method [15,38]. The beneficial effects of ultrasonic waves on crude oil viscosity were previously recognized by Abramov et al. [39]. They showed that a viscosity reduction of 16% could be obtained by the application of the ultrasonic waves implemented by a downhole tool in the wellbore [39]. An ultrasonic method for enhanced oil recovery has also been proposed [40]. An average 3-fold productivity increase was accomplished for a production well employing a chemical treatment assisted technique for industrial applications. The objective of this work is to evaluate the potential for the ultrasound method to replace conventional desalting and dehydration equipment.

In this experimental work, the ultrasonic field was tested to evaluate its demulsification performance when applied to three types of medium-gravity (25° to 30°API). These crude oils were used as the basis for forming synthetic emulsions (water-in-oil) with various water concentrations: The tests conducted on these fluid mixtures with synthetic emulsions included 1) varying the ultrasonic irradiation time (between 1 and 5 min), 2) varying the initial water concentrations in the crude oil samples (10%, 15%, 20%, and 25%), and 3) varying the ultrasonic field intensity (0.25 w/cm², 0.5 w/cm², 0.75 w/cm², and 1 w/cm²).

Three distinct crude oil (codes 010, 020 and 030) qualities are used in this study: crude oil 010 (25°API), crude oil 020 (28°API), and crude oil 030 (30°API). The crude oil samples were injected with fresh water and chemical emulsifiers: 75 ppm for 010 oil and 50 ppm for the 020 and 030 samples. These combinations were subjected to five minutes of high-speed electric mixing to produce homogeneous emulsified fluid samples. The samples were each then irradiated for 1, 2, and 5 min. The effectiveness in separating the water from the crude oil in each sample was evaluated by establishing the differences between the initial and final contents of the water. Additionally, the adhesion coefficient function (β) was measured, for each of the three samples of crude oil tested in terms of ultrasonic field intensity and the initial water concentration of crude oil.
2. Emulsification and demulsification theory

2.1. Desalting methods

2.1.1. Settling coalescing

In this procedure, for each desalting unit, usually, three equal-sized vessels or fluid reservoirs are employed in a cyclic process. While one vessel is being filled, another is undergoing settlement and the third is being discharged. This operation is repeated continuously for each of the reservoirs. Ceasing the flow in the settlement reservoir enhances the settlement by the gravity of brine molecules. These appear in the form of large water droplets in the stationary crude oil [50,51].

2.1.2. Chemical coalescing

In this method, chemical injection speeds up the settlement of brine droplets. The outer wall of the brine droplet in crude oil is surrounded by a layer of oil acting as a protective outer wall to the brine droplet. The injection of demulsifier chemical(s) causes this protective wall around the oil droplets to break down. This action enables the small water droplets to be absorbed into each other (coalesce) to form larger droplets and settle by gravitational forces. The quantity of chemicals injected depends on the physical and chemical characteristics of the crude oil and, in particular, the brine concentration in the crude oil and the temperature of the mixed fluid. Typically, dosages vary between about 5 and 100 parts per million (ppm). If too much demulsifier chemical is injected there is a risk that the water-in-oil emulsification may actually be enhanced. Excess demulsifiers may fragment the water droplets too much leading to smaller droplets that form emulsions more readily. Consequently, the dosage of injected chemicals must be tested in the laboratory and calculated before deployment [50,51].

2.1.3. Thermal coalescing

One of the effective factors in separating brine from oil is the use of heat. Consequently, many oil processing plants utilize heater-treaters to break water-in-oil emulsions. The heat reduces the viscosity of the crude oil and weakens the strength of the protective outer wall surrounding the brine droplets. As a result, water droplets are absorbed into one another and, once they reach a certain size, tend to settle by gravitational forces. The heat energy introduced has to be carefully controlled should not be exceeded by a certain amount. The ideal temperature for a heater-treater tends to be about 55 °C (130°F). At higher temperatures the lighter hydrocarbons are vaporized, leaving the liquid crude oil fraction with higher specific gravity, which is counterproductive for water oil separation. At locations where ambient temperatures are high, this typically means that little or no heating of the crude oil is required, as oil arriving from the subsurface is typically at temperatures of 50 °C or higher [50,51].

2.1.4. Mechanical coalescing

Mechanical coalescence is typically achieved using two distinct techniques:

A) Filtration, with the crude oil, passed through special filters. A downside to using a filter is that they require regular cleaning which adds to field operating costs.

B) Centrifuging, applying centrifugal force to knock out the heavier water from the crude oil. The crude oil is placed in special containers that rotate around their central axes. As a result, brine droplets coalesce towards the rotating vessel’s walls and settle by gravitational force.

Since the production rate of oilfield fluid (crude oil plus water) tends to be high in a large commercial oil field, the use of mechanical methods has often not been practical, due to vessel and energy costs. It is through a technique that is usefully employed in laboratory testing [50,51].

2.1.5. Electrical coalescing

In cases where brine is difficult to separate from the oil, typically in cases where tight emulsions are hard to break, electricity can be employed. Many oil processing plants use electrostatic-treater separators to break these tight water-in-oil emulsions. These electrical devices use positive and negative electricity poles with high-voltage for separating brine from oil. The voltages typically range from about 17,000 to 21,000 V in these vessels. The small spherical water droplets in the emulsions are drawn together under the influence of the electrostatic force imposed upon them and tend to become elliptical in shape. As a result, adjacent droplets connect more easily with one another, and they begin to settle out due to gravitational forces. If the droplets of water become too large in these electrostatic treaters, the electrical current then tends to fragment them inhibiting water settlement. Consequently, the voltage employed in these treaters has to be carefully tuned to optimize water separation. The distance between the water droplets should be kept low and their diameters should not be prevented from growing too large. For this reason, in desalting units, water is injected before the crude oil enters the electrostatic treater so that in addition to dissolving the salt particles, the distance between the droplets of brine decreases and the diameter of the water droplets is prevented from becoming too large [50,52].

2.2. Methods for generating ultrasonic waves

2.2.1. Piezoelectric vibrators

The interaction of mechanical pressure and electrical force in an environment is called the piezoelectric effect. For example, some crystals produce electrical force due to mechanical pressure, and, conversely, the created potential difference between opposite sides of such crystal causes compression and expansion. Sustaining such compression and expansion over time causes oscillation and the creation of ultrasonic waves. These materials are referred to as piezoelectric and typically exist in macrocrystalline forms. The piezoelectric effect is present only in crystals that do not have symmetrical structures. Quartz crystals often exist in piezoelectric forms and were used to create ultrasonic waves for the first time. Although many natural crystalline materials display piezoelectric properties, in the industry the crystals that are commonly used to generate sustained ultrasonic waves are made of artificially produced ceramics. Some of the materials used industrially are crystalline mixtures of lead zirconate and lead titanate which are highly piezoelectric. These materials act as a useful means for converting electrical energy into mechanical energy (as ultrasonic wave-forms), and vice versa, are referred to as transducers [53,54].

2.2.2. Magnetostriiction

Ferromagnetic materials, under the influence of the magnetic field involving small magnetic dipoles, become spontaneously aligned with their influencing dipoles. The shape and size of these materials change in these fields and can be made to oscillate according to the frequency of imposed alternating currents, leading to the production of ultrasonic waves. The intensity of the ultrasonic waves generated via the magnetostriiction method is low. This method is more practical for generating ultrasonic waves in the laboratory [55].

2.3. Ultrasonic waves in emulsion systems

As shown in Fig. 1, oil droplets tend to be dispersed in water-in-oil emulsions. The effectiveness of the breakdown of such emulsions and ultimate separation of water and oil components depends on several factors. The ultrasonic force on the emulsion system causes the separation of droplets from the fluid due to the difference in density and the velocity of sound passing through the two fluid components.

The ultrasonic force acting on the suspension consists of two forces, primary and secondary. The primary ultrasonic force causes the droplets to be moved towards the nodes or anti-nodes of the ultrasonic waves, where they accumulate due to the constraints imposed by the secondary ultrasonic force [56,57]. Low-frequency ultrasonic-wave
energy plays a key role in the separation of water components from oil in fluid systems dominated by water-in-oil emulsions. The excessive energy of the ultrasonic wave can either induce dispersion of water droplets or, depending on conditions and fluid compositions, prompt water droplets sticking to coalesce leading eventually to demulsification [22]. However, in low energy density conditions, a small increase in the intensity of the ultrasonic wave can reduce the time it takes for two droplets to adhere or coalesce.

In Fig. 2, the movement of water droplets under the influence of an ultrasonic field is illustrated diagrammatically.

The Fig. 3 illustrates an example of the effects an ultrasonic wave can cause in a water-in-oil emulsion. Fig. 3A shows water droplets dispersed without the application of ultrasonic waves. Fig. 3B shows that droplets of water are induced to coalesce in oil when ultrasonic waves are applied [50].

3. Materials and methods

3.1. Ultrasonic desalting equipment suitable for water-bearing crude oils

Many ultrasonic containers used in historically published experiments consist of a metal vessel with an ultrasonic wave transmitter located at the top end, which worked in a discontinuous cycle. However, a container design suitable for the continuous desalting of crude oil has not previously been designed. A chamber similar in characteristics to a crude oil transportation pipeline possesses the necessary features for a continuous desalting process. By placing ultrasonic transducers at the two ends of the pipe, a static ultrasonic wave can be generated. A static ultrasonic wave is desirable because it contains nodal points. Droplets of water in the crude oil can accumulate at these nodal points and gradually expand, thereby accelerating their separation and settling [58–60]. Therefore, a pipe-form desalter was designed and constructed for laboratory use (Fig. 4).

The steel pipe-form desalting pilot equipment with an ultrasonic-wave generator and glass pipe chamber are both employed as crude oil containers (Fig. 4). The steel chamber has a higher ultrasonic separation efficiency than the glass chamber due to the greater reflection of the ultrasonic waves within the steel pipe. However, filming the desalting of crude oil process is only possible in the glass chamber. The setup of the glass device is supported by two polyethylene piece holders fixed to a wooden plate. The polyethylene holders restrict vibrations of the crude oil chamber during the while the ultrasonic waves are generated and impacting the glass tube. Both crude oil containers, steel, and glass are fitted with two transducers that are located at both ends of the chambers.

In most published experiments of this type, static ultrasonic waves are produced by a single transducer and a reflector of the ultrasonic waves is also positioned in the device. The incident and reflected ultrasonic waves, then produce static waves and nodes within the device. In the experimental equipment used for this study, instead of using wave reflectors, two transducers are employed, one at either end of the pipe-form device. Both transducers produce ultrasonic waves at the same time. The interactions between these waves coming from opposite directions in the pipe produce static waves and nodes in the crude oil containers. The ultrasonic pilot equipment was constructed to make the connection of the transducers to the oil container as simple as possible. This enables the transducers to be redeployed easily between the steel and glass devices. Two Teflon gaskets seal the spaces between the pipe container and the transducer without any welding or screws being required. This prevents leakage of the oil out of the container and also simplifies the insertion and removal of the transducers from the pipe-form device. The main components of the ultrasonic transducer are illustrated in Fig. 5.

Testing of the devices revealed that ultrasonic transducers employed in the process of water separation and demulsification need to be of low...
frequency. The transducers need to encompass frequencies around 20 kHz and power ranging from 80 W to 1000 W. Transducers with the power of 100 W are suitable for separation while transducers with the power of 1000 W are better suited to emulsification. There was no pre-constructed electrical circuit suitable for the desalting pipe-form process equipment as designed employing a transducer with 100 W power and 20 kHz frequency. Two custom-designed circuits were installed, one for each transducer with software placed on circuit controllers to integrate the control function of the two transducers.

By involving two electrical circuits the transducers were able to operate at powers of 25 W, 50 W, and 75 W in addition to 100 W. This made it possible to test and record the effects of the intensity of the produced ultrasonic fields on the desalting process applied to the crude-oil samples tested. Fig. 6 shows one of the electrical circuits built for the production of ultrasonic waves in the pipe-form pilot device.

3.2. Crude oil characterization

Three different crude oil samples (codes: 010, 020 and 030) were tested in the pipe-form ultrasonic desalting pilot device. The 010 crude

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Fig. 3. Physical characteristics of droplets of water in oil. A) Without ultrasonic waves applied B) with ultrasonic waves applied [53].

Fig. 4. Pipe-form ultrasonic desalination device for desalting crude oil.

Fig. 5. The main components of the ultrasonic transducer inserted at either end of the desalting device.
oil was the heaviest (~25°API) tested and contained the most asphaltene. As the quantity asphaltene in crude oil increases, the separation of water drops from the crude oil typically becomes more difficult. This is because asphaltenes typically act as an emulsifier for water drops and generally inhibit water droplet formation and segregation. The crude oil samples 020 and 030 tested display gravities between 28 and 30°API. The key difference between them is that the crude oil sample 030 has a substantially higher salt content compared to the crude oil sample 020 sample. Table 1 shows the crude oils characterizations. The salt was added during the preparation of the emulsions to examine the ability of the treatments to desalt the samples.

3.3. Experimental procedures

Freshwater and chemical emulsifier were injected into each sample of crude oil. The combination was then subjected to 5 min of mixing in a high-speed electric mixer resulting in a homogeneous fluid containing water-in-oil emulsions. The quantity of freshwater injected ranged from 5% to 25% of the total fluid. 75 ppm of chemical emulsifier injected into the crude oil sample 010 (~25°API) and 50 ppm was injected into the crude oil samples 020 and 030 (28°API). These concentrations corresponded to the quantities used in the desalting units in the processing units of these fields. All emulsified samples were then placed in a 100 cm³ volume ultrasonic chamber for the ultrasonic field testing. Two tests were performed for each condition.

The input power to the pipe-form ultrasonic testing equipment was varied between 25 W and 100 W to test the prepared samples over a range of ultrasonic field intensities. Additional tests were conducted applying a range of ultrasonic irradiation times. During these tests, the temperature of the samples in the pipe-form device increased up to about 50 °C. The heat generated by this temperature increase acts to enhance water and oil separation during the tests, as was observed in published experiments conducted on heavy oil in an ultrasonic bath [17].

Following the irradiation periods, the fluid samples were removed from the ultrasonic chamber and poured into a graduated cylinder. This enabled the ultrasonic effects on the segregation of fluids to be

![Image](image_url)

**Fig. 6.** The electrical circuitry configured to produce ultrasonic waves in the pipe-form device shown in Fig. 4.

**Table 1**

Crude oils characterizations.

| Crude Oil Code | Unit | Standard |
|---------------|------|----------|
| Gravity °API  | ASTM D4052 | 25 28 30 |
| Conductivity %w/w | ASTM D2624 | 21 18 20 |
| Viscosity mm²/s | ASTM D445 | 19 20 21 |
| Asphaltenes %w/w | Sara | 2.4 1.1 0.5 |
| Salinity concentration ppm | IP 77-72 | 28 21 19 |

![Image](image_url)

**Fig. 7.** The effects of the quantity of initial water in the crude oil sample 010 on the percentage of water separation achieved at different intensities of the ultrasonic field applied. a) Field intensity of 0.25 w/cm²; b) Field intensity of 0.5 w/cm²; c) Field intensity of 0.75 w/cm²; d) Field intensity of 1w/cm².
quantified applying the industry standard bottle test. This test is traditionally used to quantify the effects (quantity and time impacts) of emulsifiers and demulsifiers on crude oils containing water-in-oil emulsions. This test requires the ultrasonic-treated samples to be placed in an oven for 12 h at a temperature of 80 °C. The quantity of water separated from the oil samples is then measured.

4. Results and discussion

4.1. The Impact of irradiation time and the amount of initial water on the quantity of separated water

4.1.1. Crude oil sample 010

The impacts of the duration of ultrasonic irradiation applied and the quantity of primary water in the crude oil samples is illustrated for the crude oil sample 010 in Fig. 7a–d. The ultrasonic irradiation time equals zero refers to the test without ultrasound. Two distinct trends are observed depending on the initial water concentrations in the samples. The water separation efficiency enhances with an increase in water content. At higher water contents, the volume percentage of water separated is decreased, but the amount of water separated is increased. For the samples with 10% and 15% water concentrations three straightforward trends are apparent (Fig. 7). The following results could be obtained from Fig. 7: 1) The higher the initial water content (up to 15%), the lower the water segregation percentage achieved; 2) the longer the ultrasonic irradiation time (up to 5 min), the greater the water separation percentage achieved; and, 3) The greater the irradiation intensity (up to 1 W/cm³), the greater the water segregation percentage achieved. These trends are consistent with the expectations of previous research [17,49,61].

The trends observed in Fig. 8 for the experiments performed with 20% and 25% of initial water in the crude oil 010 samples are more complex. Up to two minutes of irradiation time, the trends are similar to samples with 10% and 15% initial water. However, at most irradiation intensities the sample with 20% initial water actually achieved slightly higher water separation and 1- and 2-minutes irradiation time than the sample with 15% initial water. In all cases tested the sample with 20% initial water achieved higher ultimate water separation than the sample with 25% initial water. It is for the longer ultrasonic irradiation time of 5 min that the samples with 20% and 25% initial water contents show distinct behavior. At all irradiation intensities, those samples achieved less ultimate water separation than they did at 2 min irradiation time. Moreover, the water separation achieved for the longer ultrasonic irradiation duration (5 min), water separation was substantially less than that achieved by the samples with 10% and 15% initial water contents. These differences become more extreme as ultrasonic irradiation intensity increases (i.e., Fig. 7d). These more complex trends are consistent with the dispersion of larger droplets and their reduction to smaller droplets during longer exposures to high-intensity ultrasonic fields. This phenomenon confirms that the time crude oil is exposed to ultrasonic waves has a significant influence on water separation performance. An optimum irradiation duration time should be determined for each crude oil type processed [27,62–64].

The impacts of the irradiation time and the intensity of the ultrasonic field on the crude oil sample 010 are more distinctly illustrated for samples with specific initial water concentrations in Fig. 8. As the intensity of the ultrasonic field increases, the percent of water segregated from crude oil also clearly increases.

4.1.2. Crude oil sample 020 (low salt)

Fig. 9 shows the water separation results from the crude oil sample 020 tested. The results show similar overall trends for the crude oil sample 010 but differ in some important details. Firstly, the degree of water separation continues to increase for irradiation times up to 5 min for the crude oil sample 020 with 20% initial water content. This was not the case for the crude oil sample 010 with 20% initial water content.

Fig. 8. Impact of ultrasonic field intensity on the amount of water separated from crude oil sample 010 with different initial water concentrations. a) 10% initial water b) 15% initial water c) 20% initial water d) 25% initial water.
suggesting that longer ultrasonic irradiation times are generally more effective in separating water from this lighter crude oil at the upper limit of the medium-gravity range. Secondly, the ultimate water separated from the crude oil sample 020 is consistently higher than from the crude oil sample 010 irradiated for similar durations and at similar intensities. For instance, at 10% initial water concentrations and 5-minute ultrasonic irradiation times the water separation percentages for the crude oil sample 020 is 98% (@1 W/cm$^3$) and 85% (@0.25 W/cm$^3$) compared to 92% (@1 W/cm$^3$) and 80% (@0.25 W/cm$^3$) for the crude oil sample 010. Furthermore, at 25% initial water concentrations and 5-minute ultrasonic irradiation times the water separation percentages for the crude oil sample 020 is 86% (@1 W/cm$^3$) and 74% (@0.25 W/cm$^3$) compared to 80% (@1 W/cm$^3$) and 66% (@0.25 W/cm$^3$) for the crude oil sample 010.

**4.1.3. Crude oil 030 (high salt)**

Fig. 10 shows the water separation results from the crude oil sample 030 tested. The results show similar overall trends for the crude oil sample 020 but differ slightly in some important details. The ultimate water separated from the crude oil sample 030 (high salt) is consistently higher than from the crude oil sample 020 irradiated for similar durations and at similar intensities. For instance, at 10% initial water concentrations and 5 min ultrasonic irradiation times the water separation percentages for the crude oil sample 030 is 100% (@1 W/cm$^3$) and 87% (@0.25 W/cm$^3$) compared to 98% (@1 W/cm$^3$) and 85% (@0.25 W/cm$^3$) for the crude oil sample 020. Furthermore, at 25% initial water concentrations and 5 min ultrasonic irradiation times the water separation percentages for the crude oil sample 030 is 86% (@1 W/cm$^3$) and 72% (@0.25 W/cm$^3$) compared to 86% (@1 W/cm$^3$) and 74% (@0.25 W/cm$^3$) for the crude oil sample 020. For the samples with 25% initial water concentrations irradiated at 0.25 W/cm$^3$, water separation from the crude oil sample 020 is slightly higher than for the crude oil sample 030, which is contrary to the results for other conditions applied to these two samples.

The detailed comparison of Figs. 9 and 10 reveals that the percentage of water separated from the crude oil sample 030 (high salt), is slightly higher than for the crude oil sample 020. This trend is similar to that observed in microwave experiments that have also shown that the segregation of emulsion water droplets from crude oil occurs more readily if the crude oil’s salt content is higher [65].

For each crude oil tested there is an optimum irradiation duration that is most effective at separating water for the higher water cut samples and this optimum is less than 5 min. Water separation rates increase quite significantly for irradiation durations less than the peak duration (Figs. 8–10). The cause of this is consistent with more droplets approaching the pressure antinodes as irradiation duration increases. As this occurs the secondary acoustic forces increase and enhance the coalescence of the droplets [25,63].

Mechanistically the changes of water content achieved after the optimum ultrasonic irradiation duration value are probably due to emulsion droplets becoming dispersed into smaller droplets in response to the cavitation phenomenon. Some of the dissolved salt in the water phase tends to be transferred to the oil phase in this process and as a result, gets trapped in some of the crude oil components such as asphaltene and resin molecules [17]. Hence, irradiating crude oil for durations beyond the optimum are likely to result in less effective de-salting of crude oil.

**4.2. Separated water percentages for adhesions coefficient calculations**

To derive the adhesion coefficient functions, the separated percentages of water resulting from the ultrasonic irradiation experiments were computed for all the sample tests conducted. Selected values of the volumetric percentage and mole density quantities of the water segregated from the crude oil samples caused by the influence of ultrasonic irradiation % (@0.5 W/cm$^3$) are listed in the Tables 2–4 for initial water concentrations of 10%, 15%, and 20%.

It is the mole density values (right-hand columns in Tables 1–3) that
are specifically used in the calculation of the adhesion coefficients.

### 4.3. Adhesion coefficients derived from experimental data

The adhesion coefficient functions were derived by applying the population-balance model. This model assumes that the water droplets in crude oil are dependent upon and related to an empirically-derived function $\beta$ related to adhesion coefficients. Previously published studies on crude oil samples have applied a drop-pair model to assess the displacement of water droplets in crude oil samples tested with microwave and ultrasonic irradiation [65]. In those experiments, the irradiation time in the microwave chamber was 15 s [66], while in the experiments conducted for this study the ultrasonic irradiation time was between 1 and 5 min. A comparison can be made of the adhesion coefficients calculated from the results of the experiments conducted in microwave systems [65] and the results of the ultrasonic experiments presented here for the same crude oil samples. The adhesion coefficients

### Table 2

Quantities of water segregated from the crude oil sample 010 under the influence of selected ultrasonic wave conditions tested.

| Ultrasonic irradiation duration (Min) | Ultrasonic field intensity (W/cm$^2$) | Percentage of initial water concentration (%) | Volumetric percentage of separated water (%) | Mole density of separated water (Mol/cm$^3$) |
|--------------------------------------|---------------------------------------|----------------------------------------------|---------------------------------------------|-------------------------------------------|
| 1                                    | 0.5                                   | 10                                           | 6.8                                         | 0.000503                                  |
| 2                                    | 0.5                                   | 10                                           | 10.0                                        | 0.00798                                   |
| 5                                    | 0.5                                   | 10                                           | 10.0                                        | 0.01297                                   |
| 1                                    | 0.5                                   | 15                                           | 3.9                                         | 0.00291                                   |
| 2                                    | 0.5                                   | 15                                           | 10.2                                        | 0.00757                                   |
| 5                                    | 0.5                                   | 15                                           | 15.0                                        | 0.00151                                   |
| 1                                    | 0.5                                   | 20                                           | 9.4                                         | 0.009695                                  |
| 2                                    | 0.5                                   | 20                                           | 15.6                                        | 0.01156                                   |
| 5                                    | 0.5                                   | 20                                           | 13.5                                        | 0.00998                                   |

### Table 3

Quantities of water segregated from crude oil sample 020 under the influence of selected ultrasonic wave conditions tested.

| Ultrasonic irradiation duration (Min) | Ultrasonic field intensity (W/cm$^2$) | Percentage of initial water concentration (%) | Volumetric percentage of separated water (%) | Mole density of separated water (Mol/cm$^3$) |
|--------------------------------------|---------------------------------------|----------------------------------------------|---------------------------------------------|-------------------------------------------|
| 1                                    | 0.5                                   | 10                                           | 7.9                                         | 0.00585                                   |
| 2                                    | 0.5                                   | 10                                           | 10.0                                        | 0.00974                                   |
| 5                                    | 0.5                                   | 10                                           | 10.0                                        | 0.01373                                   |
| 1                                    | 0.5                                   | 15                                           | 7.1                                         | 0.00528                                   |
| 2                                    | 0.5                                   | 15                                           | 13.3                                        | 0.00983                                   |
| 5                                    | 0.5                                   | 15                                           | 15.0                                        | 0.01418                                   |
| 1                                    | 0.5                                   | 20                                           | 6.4                                         | 0.00476                                   |
| 2                                    | 0.5                                   | 20                                           | 11.8                                        | 0.00873                                   |
| 5                                    | 0.5                                   | 20                                           | 17.2                                        | 0.01271                                   |

### Table 4

Quantities of water segregated from crude oil sample 030 under the influence of selected ultrasonic wave conditions tested.

| Ultrasonic irradiation duration (Min) | Ultrasonic field intensity (W/cm$^2$) | Percentage of initial water concentration (%) | Volumetric percentage of separated water (%) | Mole density of separated water (Mol/cm$^3$) |
|--------------------------------------|---------------------------------------|----------------------------------------------|---------------------------------------------|-------------------------------------------|
| 1                                    | 0.5                                   | 10                                           | 7.3                                         | 0.00542                                   |
| 2                                    | 0.5                                   | 10                                           | 10.0                                        | 0.00934                                   |
| 5                                    | 0.5                                   | 10                                           | 10.0                                        | 0.01389                                   |
| 1                                    | 0.5                                   | 15                                           | 6.0                                         | 0.00447                                   |
| 2                                    | 0.5                                   | 15                                           | 11.6                                        | 0.00859                                   |
| 5                                    | 0.5                                   | 15                                           | 15.0                                        | 0.01429                                   |
| 1                                    | 0.5                                   | 20                                           | 7.3                                         | 0.0054                                   |
| 2                                    | 0.5                                   | 20                                           | 12.1                                        | 0.00899                                   |
| 5                                    | 0.5                                   | 20                                           | 17.9                                        | 0.01325                                   |
calculated from the population-balance model for water droplets in crude oil were determined separately in this study for tests conducted in both microwave and ultrasonic fields.

The mole densities of the separated water droplets under the influence of ultrasonic wave (e.g. Tables 1–3) represent the inputs to the population-balance model. The numerical value of the value of \( \beta \) in the adhesion coefficient function is then obtained by minimizing the error between the amount of water separated in terms of mole density and the experimental values obtained by applying a differential evolution (DE) optimization model \([65,67]\). By applying the MatLab curve fitting algorithm, formulaic relationships were obtained expressing adhesion coefficient function \( \beta \) for each crude oil sample tested in terms of coefficients A and B according to Eq. (1).

\[
\beta = A \times \text{(initial water content)}^B
\] (1)

The relationships used to derive the numerical values of coefficients A and B for the ultrasound experiments conducted in this study are related to four adhesion coefficients (i.e., \( A_1; A_2; B_1; B_2 \)) are expressed as Eqs. (2) and (3).

\[
A = A_1 \times \left( \text{field intensity} \left( \frac{W}{cm^2} \right) \right) + A_2
\] (2)

\[
B = B_1 \times \left( \text{field intensity} \left( \frac{W}{cm^2} \right) \right) + B_2
\] (3)

Table 5 lists the adhesion coefficients calculated using Eqs. (2) and (3) for crude oil samples tested with an ultrasonic field intensity of 0.5 W/cm\(^2\).

The values in Table 5 can be compared with those presented in Table 6 that list the A and B factors calculated using Eqs. (2) and (3) for crude oil samples tested with microwave irradiation.

The values listed in Tables 5 and 6 are expressed graphically in Fig. 11. Apart from factor \( B_2 \), the values of the adhesion coefficients for the microwaved samples are orders of magnitude lower than for those treated with ultrasonic irradiation. However, the relative magnitudes between these adhesion coefficients (i.e., \( A_1 \) to \( A_2 \) to \( B_1 \) to \( B_2 \)) are similar to the samples with ultrasonic and microwave radiation. The numerical values of ultrasonic wave adhesion coefficients are higher than microwave coefficients owing to the longer ultrasonic irradiation times involved in the tests. If temperature could be controlled in the microwave chamber, the water separation achieved with microwave treatments over longer durations could be much more significant. The adhesion coefficient shows the relative change in magnitude trends in function \( \beta \) for the three oil samples that are the same for samples treated with ultrasonic and microwave radiation.

| Crude Oil Type | \( A_1 \) | \( A_2 \) | \( B_1 \) | \( B_2 \) |
|----------------|---------|---------|---------|---------|
| crude oil 010  | 1.01E−09| −2.85E−10| 0.6674 | −0.7979 |
| crude oil 020  | 1.80E−09| −6.79E−10| −0.232 | −0.04143 |
| crude oil 030  | 2.33E−09| −8.52E−10| −0.429 | −0.02919 |

| Crude Oil Type | \( A_1 \) | \( A_2 \) | \( B_1 \) | \( B_2 \) |
|----------------|---------|---------|---------|---------|
| crude oil 010  | −2.88E−15| 2.35E−12| 0.004606| −1.148 |
| crude oil 020  | −7.95E−15| 1.50E−12| 0.002308| −0.5782 |
| crude oil 030  | −1.19E−13| 8.75E−12| 0.0219| −1.788 |

Fig. 11. Comparison of ultrasonic (Table 5) and microwave (Table 6) derived adhesion coefficients for the crude oil samples 010, 020 and 030 a) factor \( A_1 \) b) factor \( B_1 \) c) factor \( A_2 \) d) factor \( B_2 \).
The separation by ultrasound is less effective. Water separation efficiency for experimental purposes.

Three crude oils with codes 010, 020, and 030 were assessed. An ultrasonic horn with a frequency of 20 kHz and power ranges of 80 W to 1000 W was applied in the experiments. Higher water cuts and emulsions. An ultrasonic field with a frequency of 20 kHz and power ranges of 80 W to 1000 W was applied in the experiments. Three crude oils with codes 010, 020, and 030 were assessed.

Conduct ultrasonic-field experiments on medium-gravity crude oils with microwave treatments.

Fig. 13. Water contents of processed crude oil sample 030. The “witness” sample is treated with the conventional electrostatic treatment process. The sample treated with ultrasonic irradiation removes the most salt.

Figs. 12 and 13 reveals that samples treated with both microwave and ultrasonic irradiation are both more effective in removing salt and water from the crude oil sample 030 than the conventional desalting and electrostatic treatment processes that are applied to the oil types evaluated in existing oil processing plants.

The longer ultrasonic irradiation times possible compared with the relatively short microwave treatment times, constrained by temperature considerations, is the reason that treatments applied to the studied oil samples are more effective in terms of salt and water removal than microwave treatments.

5. Conclusions

An ultrasonic tubular shape device was designed specifically to conduct ultrasonic-field experiments on medium-gravity crude oils with high water cuts and emulsions. An ultrasonic horn with a frequency of 20 kHz and power ranges of 80 W to 1000 W was applied in the experiments. Three crude oils with codes 010, 020, and 030 were assessed for experimental purposes.

The experiments conducted provide useful insight into the performance of ultrasonic fields applied to remove water and mineral salts from crude oils. The increased amount of initial water and ultrasonic irradiation time will consequently enhance the quantity of water separated under a specific field intensity. However, at 20% and 25% of initial water concentrations, the separation by ultrasound is less effective. Water separation efficiency decreased for longer irradiation times (5 min or more) relative to low irradiation times (2 min). Therefore, the amount of time that the crude oils with water emulsions are exposed to ultrasonic waves has an optimal value in terms of the separation percentage achieved. Consequently, the optimal irradiation time has to be determined specifically for each type of crude oil processed. Increasing the intensity of the ultrasonic field tends to elevate the percent of water segregated from crude oil.

The percentage of water separated was higher in the mineral-salt-rich sample (crude oil 030). This suggests that the higher the salt content of crude oil, the more effective the ultrasound fields are likely to be in achieving separation of emulsion droplets of water from crude oil and in desalting the crude oil samples. Furthermore, the comparison of ultrasonic and microwave adhesion coefficient values shows that trends followed by these coefficients in terms of field intensity and initial water percentage in the studied crude oils are the same. The numerical values of ultrasonic wave coefficients tend to be orders of magnitude higher than corresponding microwave adhesion coefficients. This is due to the longer ultrasonic irradiation times compared to the short irradiation times possible with microwaves due to temperature constraints. Overall, the key benefits of applying ultrasonic techniques have been demonstrated by this study.

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