



## Ultrasonics - Sonochemistry

journal homepage: www.elsevier.com/locate/ultson

# Water-in-oil emulsions separation using an ultrasonic standing wave coalescence chamber



Carlos Mario Giraldo Atehortúa<sup>a</sup>, Nicolás Pérez<sup>b</sup>, Marco Aurelio Brizzotti Andrade<sup>c</sup>, Luiz Octávio Vieira Pereira<sup>d</sup>, Julio Cezar Adamowski<sup>a,\*</sup>

<sup>a</sup> University of São Paulo, Department of Mechatronic and Mechanical Systems Engineering, Sao Paulo 05508-900, Brazil

Universidad de la República, Facultad de Ingeniería, Montevideo 11300, Uruguay

<sup>2</sup> University of São Paulo. Institute of Physics. Rua do Matão. 1371. São Paulo 05508-090. Brazil

<sup>d</sup> PETROBRAS/CENPES, Av. Horácio Macedo 950, Ilha do Fundão, Rio de Janeiro 21941-915, Brazil

ARTICLE INFO

Keywords: Ultrasonic coalescer Water-in-oil emulsion Frequency tracking

## ABSTRACT

The offshore extraction of crude oil produces stable water in oil emulsion. To separate this emulsion into oil and water phases, the oil/water interfacial film is commonly destroyed by the addition of chemical demulsifiers. The use of an ultrasonic standing wave force field could be an alternative to reduce the dosage of chemical demulsifiers in the coalescence process. In this work, an ultrasonic separator of water in crude oil emulsions is investigated through the use of a high frequency ultrasonic standing wave coalescence chamber. The coalescing chamber uses the acoustic radiation force to induce the coalescence of water droplets at the pressure nodes of a standing wave field. Due to temperature fluctuations, the excitation frequency is controlled to maintain the resonance in the coalescence chamber and the voltage amplitude is controlled to deliver a given acoustic power. Experimental tests using standardized emulsions of water in oil were carried out in a laboratory processing plant. The effects of ultrasound application, flow rate, initial water content, demulsifier dosage and chamber inlet temperature were analyzed. The results show that the use of the acoustic radiation force improves the emulsion separation in all the conditions analyzed, when compared with the gravitational separation technique.

### 1. Introduction

Petroleum production and extraction processes generate stable water-in-oil emulsions [1,2]. Under certain circumstances, amounts of water approaching 80%-90% may even be reached. Furthermore, during the transfer from the wellhead to the manifold, the need to release part of the gas results in substantial pressure reductions through chokes and valves. Intense mixing of oil and water occur, possibly decreasing the size of the droplets through flow-induced break-ups down to diameters averaging around ten microns [3]. Gravity separators are currently used to conduct the primary oil processing in the first stage, in which the final separation is produced by difference in the fluid densities [4]. In general, the oil to be sent to the refinery must have less than 1% of water content. To achieve this value, gravity separation is enhanced with other techniques, such as addition of a chemical demulsifier [5], electrostatic separation [6], mechanical Barrier [7], heating [8], among others. Addition of chemical demulsifiers and electrostatic separators are two of the most common techniques used. Chemical demulsifiers can be injected to neutralize the stabilizing effect of surfactants. When added, they migrate to the water-oil interface, causing rupture or weakening of the protective film around the droplets. The demulsifier effectiveness depends on the composition of the oil as well as the water pH value. In the electrostatic separator, the demulsification enhancement occurs by the application of an electric field, which promotes the contact between the droplets and enhances droplets coalescence [9]. Although electrostatic separators are widely used in the oil industry, mature oil fields may produce excessive quantities of water. Electrostatic separators containing bare metal electrodes are appropriate for water content lower than 20% [10]. The electrical conductivity between the electrodes increases with the water content, and may cause a disruptive discharge if the content of water is too high. A promising approach for water-in-oil emulsions with water content above 20% is the ultrasonic separation technique, which is not limited to emulsions having low concentrations of water, as the aforementioned electrostatic separators. The application of high frequency ultrasonic standing waves has been investigated [11,12], showing that the technique has potential to break water in oil emulsion in the oil industry.

E-mail address: jcadamow@usp.br (J.C. Adamowski).

https://doi.org/10.1016/j.ultsonch.2019.04.043

Received 15 July 2018; Received in revised form 23 April 2019; Accepted 29 April 2019 Available online 29 April 2019

1350-4177/ © 2019 Elsevier B.V. All rights reserved.

<sup>\*</sup> Corresponding author.

Typical water-in-oil emulsions have water droplets ranging from less than  $1 \mu m$  to tens of micrometers in diameter [3,13], with an average size on the order of  $10 \,\mu m$  [14]. The buoyant force and the gravity force of the water droplets in the emulsion are proportional to the droplet volume. However, the drag force is proportional to the droplet radius considering Stoke's drag for slow displacements; thus, for small droplet radius, the process is too slow and the emulsion is highly stable. Ultrasound separation technique makes use of the acoustic radiation force phenomenon to move the droplets toward pressure nodal or anti-nodal planes. In the water in oil emulsion, the water droplets immersed in oil have positive contrast factor and thus move toward the pressure nodes [15]. That effect causes droplets to clump together [16]. increasing their concentration and their probability of collision. The choice of the frequency is a compromise between the number of nodal planes and the acoustic attenuation of the emulsion for a given resonating chamber width. High frequency provides a large number of nodal planes; however, high frequency sound waves are more strongly attenuated in a fluid medium with distance. An additional reason for selecting a high frequency is to increase the cavitation threshold in the emulsion. Cavitation must be avoided to prevent emulsification.

The design of the resonating chamber is a key point in the implementation of the ultrasonic separator. It basically consists of a resonant cavity with attached piezoelectric transducers. The cavity resonant frequency depends on the speed of sound of the emulsion, which in turn depends on the temperature. If a temperature gradient occurs from the inlet to the outlet, different sections of the chamber have different resonant frequencies, limiting the system efficiency. The use of a single frequency in the entire chamber produces coalescence only in places where the resonance occurs. This paper presents the design of a coalescing chamber with multiple ultrasonic transducers operating independently. Two independent ultrasonic transducers were used along the resonating chamber in the flow direction. To maintain a standing wave in the chamber, a frequency tracking strategy is used to overcome the sound speed variation during operation, mainly due to temperature variation [17]. Dynamic testing of ultrasonic separation was carried out in a laboratory processing plant at the Petrobras research center, using synthetic water in oil emulsions.

#### 2. Ultrasonic coalescing chamber

#### 2.1. Acoustic radiation force

The acoustic separation of water-in-oil emulsion is based on the phenomenon of the acoustic radiation force [18]. When a small droplet of volume *V* is immersed in a fluid medium in the presence of a plane standing wave of pressure amplitude  $p_0$ , it experiences a primary acoustic radiation force, given by [19]

$$F = -\left(\frac{\pi V}{2\lambda\rho_0 c_0^2}\right) \phi p_0^2 \sin(2kx),\tag{1}$$

where *x* is the droplet position in relation to a pressure node,  $\lambda$  is the acoustic wavelength, *k* is the wavenumber, and  $\rho_0$  and  $c_0$  are, respectively, the density and sound velocity of the fluid in which the droplet is immersed. In Eq. (1),  $\phi$  is the acoustic contrast factor, defined as [19]

$$\phi = \frac{5\rho_d - 2\rho_0}{2\rho_d + \rho_0} - \frac{\rho_0 c_0^2}{\rho_d c_d^2},\tag{2}$$

where  $\rho_d$  and  $c_d$  are the droplet density and the sound velocity in the droplet, respectively. Droplets with positive contrast factor gather at the pressure nodes while droplets with negative contrast factor aggregate at the pressure antinodes. Water droplets in an oil medium have a positive contrast factor and, therefore, they agglomerate at the pressure nodes of the standing wave field.

In addition to the primary force, multiple droplets may also experience interparticle forces (the so-called secondary acoustic radiation



Fig. 1. (a) Coalescing chamber operation principle and (b) illustration of the ultrasonic coalescing chamber.

force [20,21]). This force originates from the wave scattering by the neighboring droplets. The interparticle force between two droplets can be either attractive or repulsive depending on their orientation in respect to the standing wave field. Yet when they are located at the pressure nodes, the interparticle force is attractive. As pointed out by Pangu and Feke [15], once the droplets are located at the pressure node due to the action of the primary force, secondary forces induce agglomeration of the droplets, thus improving the separation efficiency.

#### 2.2. Ultrasonic coalescing chamber

The separation of water-oil emulsion in a continuous flow is performed in an ultrasonic coalescing chamber formed by two ultrasonic transducers and an opposing planar reflector. The working principle of the ultrasonic coalescing chamber is illustrated in Fig. 1a. The emulsion enters the chamber at the upper inlet and then goes through an ultrasonic standing wave field. In this region, the acoustic radiation force drives the water droplets towards the pressure nodes, inducing its coalescence and thus increasing the average droplet size. The emulsion exits the chamber at the bottom outlet and the final separation of water and oil takes place in a separator vessel. The coalescing chamber operates at a frequency of approximately 1 MHz, the acoustic cavity has



Fig. 2. Electric conductance curve with the chamber filled with oil and filled with air.

an interval volume of 264 cm<sup>3</sup> and the distance between the transducers and the opposing reflector is 20 mm.

To have an ultrasonic standing wave of high pressure amplitude, the coalescing chamber must operate at resonance. However, two different types of resonance can be distinguished. The first, and the more intense, is due to the piezoelectric transducer. This resonance is essentially independent of the fluid inside de cavity. The other type of resonance is produced when the size of the cavity is a multiple of half wavelength. In this case, the resonance is produced in the fluid itself and a stationary pressure pattern is established inside the cavity. Fig. 2 shows these two different types of resonances. Here we can see the electrical conductance with and without oil inside the chamber.

Temperature fluctuations cause the sound velocity to change, shifting the resonance frequency of the acoustic cavity. In addition, the temperature at the outlet is higher than the temperature at the inlet. Consequently, the resonance frequency at the inlet is significantly different from the resonance frequency at the outlet, and the use of a single operating frequency in the cavity would produce coalescence only in places where the resonance occurs. To reduce this problem, two independent ultrasonic transducers were used. As shown in Fig. 1b, each transducer is formed by three lead zirconate titanate (PZT4) piezoelectric ceramics attached to an aluminum layer, and they are excited independently by an electronic module with a resonance frequency tracking system (see Supplementary material for details of the resonance tracking system). This ensures that regions in front of both transducers operate at the resonance, with maximum pressure amplitude.

#### 3. Experimental setup

Dynamic testing of acoustic separation with synthetic water in oil emulsions were carried out in a laboratory processing plant at the Petrobras research center. A diagram of the hydraulic circuit used in the experiments is presented in Fig. 3. It is composed of an emulsion reservoir tank, a hydraulic pump, an ultrasonic coalescencing chamber, a gravity separator vessel with a coalesced water drainage, and the oil discharge. The unit has a temperature control system, with electric heaters in the pipelines and in the separator vessel, as well as a flow control system. The ultrasonic coalescence chamber is positioned vertically in the hydraulic circuit to create a downward flow to prevent water accumulation. The static pressure in the hydraulic circuit was kept constant at 517 kPa during the tests.

To perform the tests, an oil blend with API gravity of 24.2 was



Fig. 3. Laboratory processing plant arrangement scheme.

obtained by mixing of 70% oil of 29 °API and 30% of oil of 13 °API, in volume, which is similar to a Brazilian pre-salt petroleum. The oil has viscosities of 24 and 15 cP (0.024 and 0.015 Pa·s) at 60 and 70 °C, respectively. In a first step, synthetic water in oil emulsions with water contents of 30% and 50% were obtained by mixing water and the oil blend in a pendulum mixer at 60 cycles per minute for 1 min. After this procedure, the mixture was processed in a Polytron® PT 3100D homogenizer at 10,000 rpm for 3 min to obtain a water in oil emulsion with water droplets averaging 10  $\mu$ m (D50). In all the experiment, the emulsion was prepared just before the start of each experiment. Without the addition of chemical demulsifier, the emulsion can be considered stable, since we could not observe any detectable change of the droplet size distribution.

The mean water droplet size distribution in emulsion was measured using a static laser light scattering particle size analyzer (Mastersizer S particle size stabilized by He-Ne gas lasers, Malvern Instruments Ltd). It was used a drop of emulsion in 50 mL of spindle oil in order to achieve between 10% and 30% of obscurescence. The drop size measurement was reported by D50, that is the volume median diameter.

The performance of the ultrasonic coalescence chamber to break the emulsion was evaluated with five different setups with and without the application of ultrasound. Tests were performed by varying the initial water content in the emulsion, the amount of demulsifier (Dissolvan 961), the flow rate and the temperature in the ultrasonic coalescence chamber. The electric power of the two ultrasonic transducers were kept constant at 40 W per transducer in a total power of 80 W. The parameters of each test setup are presented in Table 1.

### 4. Results and discussion

After performing the tests of Table 1, the final water content in the emulsions was obtained by Karl Fischer potentiometric titration using a Metrohm 841 Titrando equipment. For each test setup, the experiments are performed with and without ultrasound, a set of six pairs of measurements was obtained over 3 h of continuous operation. The results for the five test setups are summarized in Table 2 and Fig. 4.

## Table 1

Test setups of water in oil emulsion separation experiments.

Test setup	Initial water content [mass percent]	Amount of demulsifier [ppm]	Flow rate [cm <sup>3</sup> /min]	Temperature [°C]
1	30	25	50	70
2	30	25	100	70
3	30	50	50	70
4	30	25	50	60
5	50	25	50	70

#### Table 2

Final water content in the processed emulsion.

Test setup number	[Mass percent]		
_	Ultrasound on	Ultrasound off	
1	2.6	4.0	
2	5.0	15.1	
3	1.9	2.5	
4	2.4	4.2	
5	1.5	2.3	



Fig. 4. Results of the water separation for the five test setups of Table 1.

The results of the five tests show that the application of ultrasonic standing wave reduces the final amount of water in the emulsion in all the analyzed cases. Considering test setup 1 as a basis for comparing with the other test setups, the following considerations can be made:

- a) *Residence time* The influence of the residence time can be observed in the experiment of test setup 2, which has twice the flow rate of setup 1 (i.e. its residence time is half of test setup 1) and, consequently, the amount of water is about the double with ultrasound on and almost four times with ultrasound off. Using a 20 kHz acoustic chamber without flux, Xie and collaborators reported water content reduction in crude oil emulsions from 40% to 3.8% at 60 °C in 12 min of irradiation time [22]. Although the operation frequencies are different, both works showed the coalescing effect of ultrasonic standing waves, and our results indicate that ultrasound application can greatly reduce the residence time of the emulsion if a complementary technique is used, such as electrostatic separation.
- b) Amount of demulsifier The injection of chemical demulsifier in primary processing tanks varies from 20 to 150 ppm, depending on the composition of the oil as well as the water pH value. The final water content of test setup 1 with 25 ppm of demulsifier and ultrasound on is similar to that obtained in test setup 3 with 50 ppm of demulsifier and ultrasonic off. This comparison indicates ultrasonic standing waves can reduce the consumption of chemical demulsifier in the coalescence process, thus reducing oil production costs.
- c) *Processing temperature* The processing temperature of test setup 4 was decreased by 10 °C and kept constant at 60 °C during the test. The water content obtained with ultrasound at 60 °C was almost the same as in the test at 70 °C. This result shows that lower processing temperature can be used when ultrasound is on. Lower temperature means less power required for heating the oil.
- d) Initial water content The initial water content of test setup 5 is 66%

higher than that of test setup 1. Although the concentration of water was higher in test 5, the final water content was lower than that in test setup 1. The comparison between test 1 and test 5 shows that the increase in the water content of the emulsion interfered positively with the coalescence of the water droplets, both with ultrasound on and off. Probably, a greater number of water droplets facilitate flocculation and subsequent coalescence.

#### 5. Conclusions

The design of the ultrasonic coalescence chamber and its automatic electronic controller is a crucial step to obtain high efficiency in the application of electric power to obtain an ultrasonic standing wave. The main difficulty in the implementation of an ultrasonic demulsifier in the MHz range is the tuning of the operating frequency. This frequency must be selected in a resonance of the fluid inside the chamber cavity. In the present implementation, the use of two different transducers, with separate frequency control, makes the operation more efficient.

We demonstrated that the use of at least two separate piezoelectric transducers in the ultrasonic chamber, transversely positioned to the flow direction, allows the resonance frequency tracking to obtain a standing wave in the emulsion even when the emulsion composition and temperature vary. This behavior allows the control of the electrical power supplied to the ultrasonic transducers. It assures a constant power transmission to the emulsion in the frequency range 1.09-1.15 MHz, automatically adjusting the frequency and the magnitude of the excitation voltages of the two ultrasonic transducers to obtain a constant output power. In all the experimental tests conducted, the ultrasonic coalescence technology was observed to have potential for reducing the oil dewatering time as compared to the gravitational segregation mechanism. The results of test setup 2 indicates that the coalescing effect of ultrasonic standing waves can greatly reduce the residence time of the emulsion if a complementary technique is used. such as the electrostatic separation, which works very well for small amounts of water in the emulsion.

#### Acknowledgements

This study was partly financed by the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior/Coordination for the Improvement of Higher Education Personnel – Brasil (CAPES). We thank the support received from Petrobras/ANP (grant n. 0050.0064340.11.9), FAPESP (grant n. 2014/08185-2) and CNPq.

#### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ultsonch.2019.04.043.

#### References

- K. Kumar, A.D. Nikolov, D.T. Wasan, Mechanisms of stabilization of water-in-crude oil emulsions, Ind. Eng. Chem. Res. 40 (2001) 3009–3014.
- [2] J. Sjoblom, N. Aske, I.H. Auflem, O. Brandal, T.E. Havre, O. Sæter, A. Westvik, E.E. Johnsen, H. Kallevik, Our current understanding of water-in-crude oil emulsions. Recent characterization techniques and high pressure performance, Adv. Colloid Interface Sci. 100 (2003) 399–473.
- [3] S. Kokal, Crude-oil emulsions: a state-of-the-art review, SPE Prod. Facil. 20 (2005) 5–13.
- [4] E.W.M. Hansen, Phenomenological modelling and simulation of fluid flow and separation behaviour in offshore gravity separators, ASME Publ. PVP 431 (2001) 23–29.
- [5] C.W. Angle, Chemical demulsification of stable crude oil and bitumen emulsions in petroleum recovery – a review, in: J. Sjöblom (Ed.), Encyclopedic Handbook of Emulsion Technology, Marcel Dekker, New York, 2001, pp. 541–594.
- [6] J.S. Eow, M. Ghadiri, Electrostatic enhancement of coalescence of water droplets in oil – a review of the technology, Chem. Eng. J. 85 (2002) 357–368.
- [7] W. Meon, W. Rommel, E. Blass, Plate separators for dispersed liquid-liquid systems: hydrodynamic coalescence model, Chem. Eng. Sci. 48 (1993) 159–168.
- [8] A.B. Nordvik, J.L. Simmons, K.R. Bitting, A. Lewis, T. Strøm-Kristiansen, Oil and

water separation in marine oil spill clean-up operations, Spill. Sci. Technol. B 3 (1996) 107–122.

- [9] J.S. Eow, M. Ghadiri, A.O. Sharif, T.J. Williams, Electrostatic enhancement of coalescence of water droplets in oil: a review of the current understanding, Chem. Eng. J. 84 (2001) 173–192.
- [10] T. Frising, C. Noik, C. Dalmazzone, The liquid/liquid sedimentation process: from droplet coalescence to technologically enhanced water/oil emulsion gravity separators: a review, J. Dispers. Sci. Technol. 27 (2006) 1035–1057.
- [11] G. Ye, X. Lu, P. Han, X. Shen, Desalting and dewatering of crude oil in ultrasonic standing wave field, J. Petrol. Sci. Eng. 70 (2010) 144–144.
- [12] P. Parvasi, A. Khaje Hesamedini, A. Jahanmiri, M.R. Rahimpour, A comparative study on droplet coalescence in heavy crude oil emulsions subjected to microwave and ultrasonic fields, Sep. Sci. Technol. 48 (2013) 1591–1601.
- [13] L.L. Schramm, Emulsions: Fundamentals and Applications in the Petroleum Industry, American Chemical Society, Washington, D.C, 1992.
- [14] A.E. Bresciani, R.M.B. Alves, C.A.O. Nascimento, Coalescence of water droplets in crude oil emulsions: analytical solution, Chem. Eng. Technol. 33 (2010) 237–243.
- [15] G.D. Pangu, D.L. Feke, Acoustically aided separation of oil droplets from aqueous

emulsions, Chem. Eng. Sci. 59 (2004) 3183-3193.

- [16] T. Leong, L. Johansson, P. Juliano, S.L. McArthur, R. Manasseh, Ultrasonic separation of particulate fluids in small and large scale systems: a review, Ind. Eng. Chem. Res. 52 (2013) 16555–16576.
- [17] C.M.G. Atehortúa, N. Pérez, M.A.B. Andrade, J.C. Adamowski, L.O.V. Pereira, Design and implementation of the frequency control in an ultrasonic break waterin-oil emulsion chamber, Phys. Proc. 70 (2015) 42–45.
- [18] H. Bruus, Acoustofluidics 7: the acoustic radiation force on small particles, Lab Chip 12 (2012) 1014–1021.
- [19] T. Laurell, F. Petersson, A. Nilsson, Chip integrated strategies for acoustic separation and manipulation of cells and particles, Chem. Soc. Rev. 36 (2007) 492–506.
- [20] M.A.H. Weiser, R.E. Apfel, E.A. Neppiras, Interparticle forces on red cells in a standing wave field, Acta Acust. United Acust. 56 (1984) 114–119.
  [21] G.T. Silva, H. Bruus, Acoustic interaction forces between small particles in an indicating activity of the standard statement of the standard statement of the stat
- [21] G.T. Silva, H. Bruus, Acoustic interaction forces between small particles in an ideal fluid, Phys. Rev. E 90 (2014) 063007.
- [22] W. Xie, R. Li, X. Lu, P. Han, S. Gu, Acoustically aided coalescence of water droplets and dehydration of crude oil emulsion, Korean J. Chem. Eng. 32 (2015) 643–649.