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Separation of crude oil from wastewater using high-frequency ultrasound

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Abstract: Crude-oil and gas production always generates a large amount of stable oily wastewater. An alternative treatment involves the use of high-frequency ultrasound to induce oil separation. A controlled 1 MHz ultrasonic chamber was used to separate a 2500ppm (g_{oil}/g_{water}) synthetic Brazilian crude-oil-in-water emulsions without the use of chemical additives. An oil separation efficiency of about 70% was achieved with a power input of around 40 W applied for 2 minutes. In contrast, the non-acoustic reference resulted in an oil separation of only around 3.6%.

Keywords: High-frequency ultrasound, acoustic radiation force, resonance control, oil-in-water emulsion, emulsion separation.

Introduction

A large amount of oily wastewater is constantly generated during the crude-oil and gas production [1]. The wastewater can be considered a crude-oil-in-water microemulsion, with oil concentrations varying up to 5000 ppm, which must be treated before being discarded or reused following strict environmental regulations and oil industry specifications [2]. Such microemulsions remain a challenge for the oil industry due to their high stability, which makes them difficult to separate and recover [3]. Their treatment is commonly performed using the following processes: gravity separators and bed coalescers [4] that are essentially large settling tanks for water and settleable solids, hydrocyclones [5] in which the oily water is forced through a conical spiral that accelerates and displaces it along the internal walls, and the oil droplets tend to be displaced towards the central part of the spiral, facilitating their extraction, gas flotation [6] injection of dissolved or induced gas is a technique used to increase the difference in density between water and oil droplets to promote separation, and chemical additives [7] that usually are water-soluble and aid the flocculation process in which particles are aggregated together.

An alternative treatment involves the use of the acoustic radiation force, generated by high-frequency ultrasound, to induce oil separation. Although this technique is being applied in industries such as chemical, food processing, and medicine, its application for separating crude-oil from wastewater is still under development [8]. In the ultrasonic technique for treating oily wastewater, an acoustic chamber is used to produce high-frequency ultrasound that promotes oil separation by inducing the agglomeration and/or coalescence of oil droplets [9]. This process increases the size of the droplets and their buoyant capability, consequently enhancing the separation process.

A dual closed-loop control system was implemented in the ultrasonic chamber to maintain consistent resonance and stable delivery of acoustic energy. This setup actively monitors the resonance frequency and modulates input power through proportional-integral and proportional controllers. Similar feedback mechanisms have been successfully employed to enhance acoustic control and precision in treatments such as selective cell trapping and resonator-based bulk mode acoustophoresis [10, 11]. These methodologies offer reliability despite varying loads and temperatures, making them ideal for intricate emulsified systems.

Acoustic separation

An acoustic chamber vibrates at its resonance frequency driven by a piezoelectric transducer, which generates the so-called acoustic radiation force within a sample cavity of the chamber. In oily wastewater, the collision of emulsified oil droplets can be improved by this force, which can induce coalescence between droplets and aid in their separation [12].

The acoustic contrast factor is a characteristic value that defines the direction and magnitude of the acoustic force. This parameter contains the relative physical

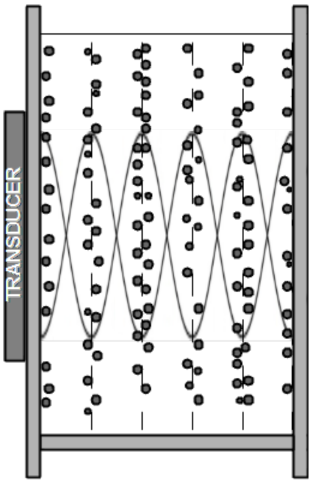

Fig. 1: Representation of High-frequency ultrasound agglomerating oil droplets in water at the antinodes.

properties of the fluid, or continuous phase, and the emulsified droplets or diluted particles [13]. For an oil-in-water emulsion, the factor indicates the oil droplets motion due to the acoustic standing wave field generated inside a chamber. The differences in density and compressibility between the continuous phase and the droplet, or particles, define the acoustic contrast factor ϕ_{ac} as follows:

$$\phi_{ac} = (5\rho_{dr} - 2\rho_F)/(\rho_{dr} + \rho_F) - \beta_{dr}/\beta_F, \quad (1)$$

in Eq. (1), ρ_F and ρ_{dr} are the densities of the fluid medium and emulsified droplet, respectively, β_F and β_{dr} are the compressibilities of the fluid and the droplet, respectively. The acoustic contrast factor is negative for oil-in-water emulsions, indicating that the oil droplets will be pushed toward the antinodes of the acoustic standing wave field by the acoustic radiation force.

Fig. 1 presents a schematic representation of the action of high-frequency ultrasound on crude-oil droplets emulsified in water, in which gray circles represent oil droplets emulsified in water, two sinusoidal lines represent the acoustic standing wave field created by a piezoelectric transducer, and the vertical dashed lines indicate the positions of the antinodes. In the figure, the oil droplets are driven toward the pressure antinodes of the standing acoustic wave, which promotes droplet agglomeration and increases the separation potential.

Materials and Methods

A high-frequency acoustic chamber is a resonant device with a piezoelectric transducer that is electrically excited by a sinusoidal voltage signal at a frequency of around $1\,\mathrm{MHz}$. The transducer causes the entire chamber to resonate, and the vibration produced is transmitted to any sample inside the device. Two parallel aluminum plates, spaced 20mm apart by an acrylic "U" shaped component, create cavity in the chamber. The first plate acts as a coupling layer, and the second serves as a reflecting element. The acoustic standing wave field was generated between the aluminum plates by four $1\,\mathrm{MHz}$ square PZT-4 piezoceramic plates. Their positive electrodes were bonded to the $6.4\,\mathrm{mm}$ thick coupling layer. The resonance frequency of the chamber depends primarily on the dimensions of the chamber cavity, the properties of the emulsion, and the process temperature.

For the optimal operation of the acoustic chamber, a dual closed-loop control system is implemented. This system ensures that the driving frequency remains aligned with the resonance of the chamber and accurately regulates the acoustic power delivered to the ultrasonic device. The electrical power is continuously monitored and modulated, taking into account the effects of electrical noise, fluctuations in amplifier gain, and the acoustic characteristics of the sample, as detailed in [14]

The control system utilizes a proportional-integral (PI) controller to minimize tracking deviations in the resonance frequency caused by temperature variations. Another simpler proportional controller is used to maintain the acoustic power at a predetermined level, as determined by previous empirical findings, by modulating the input voltage. This robust system achieves resonance, thereby maintaining both resonance stability and power delivery throughout the treatment period. These measures ensure the consistency of experimental conditions and enable the comparison of outcomes with those from non-acoustic treatments.

A synthetic wastewater containing approximately 2,500 ppm of oil (g_{oil}/g_{water}) was used to simulate microemulsions typical in the oil industry. The preparation of the samples involved the dissolution of the surfactant, the surfactant was dissolved in deionized water at $80\,^{\circ}\mathrm{C}$, the Brazilian petroleum (approximately 24 °API) was added at room temperature (including a 15% increase due to preparation losses). The quantities used are presented in Tab. 1.

Tab. 1: Table synthetic wastewater components.

Component	Quantity
Deionized water	200 g
Surfactant	300 mg
Crude-oil	500 mg $(+15\%)$

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Tab. 2: Liquids properties.

Component	Property	Quantity
Water	Dens. ρ_F $[kg/m^3]$	998
	Comp. β_F $[m^2/s]$	4.51×10^{-10}
Crude-oil	Dens. ρ_{dr} $[kg/m^3]$	909.2
	Comp. β_{dr} $[m^2/s]$	5.53×10^{-10}

An UltraTurrax ICA T25 homogenizer was used to initially mix the components at 10,000 rpm for 3 minutes to ensure full incorporation, followed by emulsification at 15,000 rpm for 30 minutes. Synthetic wastewater presented an oil content decrease of about $4\,\%$ in one hour.

Results

A controlled 1 MHz acoustic chamber was used to separate synthetic Brazilian crude-oil-in-water microemulsions. No chemical additives were used to help the separation process. The ultrasonic test at high frequency followed this procedure: the oil concentration in the synthetic wastewater was initially measured for a period of 10 minutes. Subsequently, a power of around 40 W ($\approx$ 0.42W/cm^2) was applied to the chamber, with a sonication duration of 2 minutes. Lastly, the oil measurement was conducted for approximately 35 minutes. Both continuous lines in Fig. 2, referred to as *emulsion treatment*, represent the oil content variation during the ultrasonic test. In this figure, a reference test is also presented, indicated by a dotted line. The oil separation efficiency was determined using measurements from an oil-in-water content analyzer (HD-100, Advanced Sensors).

Fig. 2 shows that the acoustic field was switched on after approximately 10 min, as indicated by the label in the plot. Shortly after the onset of sonication (2 min), the oil content of the emulsion (solid line) drops rapidly, whereas the reference test (dotted line) exhibits only minor fluctuations around its initial value.

Before ultrasound ($t < 10$ min), both curves overlapped at $\approx\ 2,590$ ppm (g_{oil}/g_{water}), confirming negligible gravity-driven separation on the time scale investigated. Upon sonication, the treated synthetic wastewater sample dropped by $\approx 27\,\%$ within 5 min (time required to stop the oil content analyzer, perform the ultrasonic treatment, and restart the analyzer), and stabilized near 660 ppm (g_{oil}/g_{water}) after 35 min. It can also be observed in Fig. 2 that once the chamber reached its resonant condition and the ultrasound was applied, the oil concentration in the treated sample decreased significantly. These results

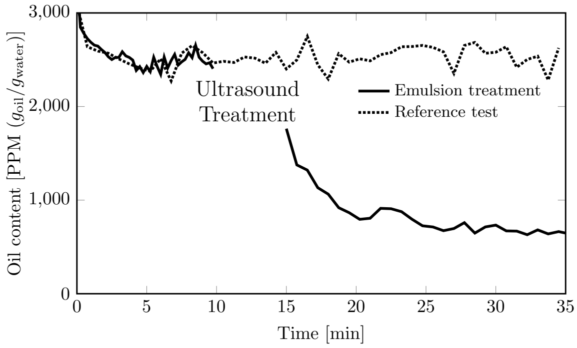

Fig. 2: Oil content variation.

are summarized in Tab. 3.

The separation process, therefore, removed almost 70 % of the initial oil content. In contrast, the reference remained between 2,400 and 2,600 ppm (g_{oil}/g_{water}). The findings confirm that radiation forces agglomerate droplets and accelerate buoyancy, delivering enhanced separation without the need for chemical additives or mechanical filters. As previously mentioned, the acoustic radiation force pushes the oil droplets emulsified in water toward the antinodes of the standing wave. This behavior is consistent with expectations, as the acoustic contrast factor ϕ_{ac} of the synthetic wastewater is negative, $\approx$ -0.32, this value can be calculated with the parameters listed in Tab. 2 and using Eq. (1).

The controlled application of high-frequency ultrasound successfully separated oil from a Brazilian crude-oil-in-water microemulsion with an initial concentration of about 2,500 ppm (g_{oil}/g_{water}) without using chemical additives. The results demonstrated oil separation efficiency slightly above 70%, achieved with a power input of around 40 W ($\approx$ 0.42W/cm^2) applied for 2 minutes. In contrast, the reference test (non-acoustic) yielded an oil separation of only approximately 3.6%.

Highlights

The controlled application of high-frequency ultrasound significantly improved the oil separation compared to the non-acoustic separation. These results suggest that ultrasound constitutes a promising tech-

Tab. 3: Oil separation test

Test	Inicial PPM $[g_{oil}/g_{water}]$	Final PPM $[g_{oil}/g_{water}]$
Ultrasound	$2595.2(\pm 77.4)$	$665.5(\pm 33.6)$
Reference	$2474.6(\pm 103.2)$	$2536(\pm 124.8)$

nique for crude-oil separation. To validate the findings presented, additional tests of the unconsidered conditions will be required for further confirmation. However, high-frequency ultrasound can be regarded as an environmentally friendly alternative as it eliminates the need for the use of chemical additives to induce oil separation. As outlined in Tab. 3, the application of ultrasound reduced the oil content from approximately 2,595 to 665 ppm (g_{oil}/g_{water}), while the control sample remained almost the same.

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