Enhanced Acoustic Separation of Oil-Water Emulsion in Resonant Cavities

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Abstract: This paper explores the use of acoustics for the recovery of oil in oil-water emulsions. A series of experimental studies were conducted using acoustic standing waves in resonant cavities as means of trapping oil droplets and enhancing oil separation. Several cavity configurations were explored and the frequency range used was between 1 MHz-2 MHz. Oil-water emulsions studied were made using mineral oil and motor oil.

INTRODUCTION

Acoustic standing waves have been used to manipulate solid particle-fluid systems in tubes and capillaries for various applications. The exploitation of acoustic technologies has been well established in the area of particle manipulation, e.g., particle transport, separation, or removal [1]. In contrast to the extensive amount of research in particle-fluid systems, the use of acoustic technology for liquid-liquid systems, such as emulsions has been focused mainly toward fundamental studies of nucleation [2], coagulation and dispersion [3], and coalescence [4]. The recovery of oil from oil-water emulsions is an important area of technology relevant to the petroleum industry. In recent years, there has been various technology developments in this area such as the use of alternating electric fields in electrical separation of oil [5]. In the area of acoustics, Pangu and Feke have demonstrated oil recovery from an aqueous emulsion using ultrasonic standing waves in a cavity with a porous material inside the cavity [6]. However this work was limited to a single cavity geometry and a single frequency and the oil used was vegetable oil. Crum [7] used a combination of ultrasound and electrolysis techniques to flocculate micelles and then coalescing the micelles for oil recovery. In the context of oil recovery from oil-water emulsions, the application of acoustics is rather limited.

The characteristics of the oil-water emulsion vary with its rheology that is dependant on temperature, pressure, particle-particle interaction and gravitational forces [8]. Obstacles to overcome in physically separating these emulsions using ultrasound are: stability of the emulsion (surfactant), the concentration of oil, and size of oil droplets. The objective of this paper is to present the results of an experimental study of oil droplet coalescence and separation in a couple of oil-water emulsions in different types of resonant cavities and the effect of a range of different resonance frequencies, acoustic power, and electrical signal waveforms. Our goal has been to understand heavy crude oil-water emulsion separation.

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THEORY

Ultrasonic standing waves can be generated in a fluid-filled cell if the applied ultrasonic frequency on opposite ends of the cell is tuned to satisfy the condition

$$L = n\frac{\lambda}{2} \quad (n=1,2,3\ldots) \tag{1}$$

where, L is the pathlength in the fluid and λ the wavelength of the ultrasonic wave. In this case displacement nodes appear at the spacing of $\lambda/2$. At the same time, pressure nodes appear at the positions of the displacement antinodes. Fluid containing small compressible spheres suspended in such a standing acoustic wave field experience various radiation forces. This force has three separate components: (1) primary axial acoustic radiation force (F_p), (2) primary transverse acoustic radiation force, and (3) secondary acoustic radiation force (Fs). When the fluid is a two-phase system (e.g., an emulsion) the droplets of one phase in a second continuous host medium experience a time averaged primary axial acoustic force, which is generated by the interaction between droplets and the primary wave field. The primary acoustic force exerted on a droplet has been studied theoretically by many researchers [9-14]. For a compressible liquid drop this primary radiation force can be expressed as

$$F_p = -4\pi R^3 k E_{ac}.\phi(\beta,\rho).\sin(\frac{4\pi z}{\lambda}) \tag{2}$$

where E_{ac} is the energy density of the acoustic field, z is the distance from a pressure node, R is the radius of the drop, and k is the wave number of the driving frequency in the host liquid. The contrast factor ϕ is

$$\phi(\beta,\rho) = \frac{5\rho_p - 2\rho_f}{2\rho_p + \rho_f} - \frac{\beta_p}{\beta_f} \tag{3}$$

For liquids, the compressibility is expressed as $\beta = 1/\rho c^2$ where c is the sound velocity of the liquid. In case ϕ takes a negative value, F becomes the restoring force around the displacement node ($z=0$); i.e., $F>0$ for $z<0$ and $F<0$ for $z>0$. Consequently, the droplet is trapped at the displacement node. On the other hand, if ϕ takes a positive value, F becomes the restoring force around the displacement antinode

or the pressure node *(λ/4)*, and hence the droplet is trapped at the pressure node.

The acoustic contrast factor *ϕ* was calculated for mineral oil and motor oil in water using literature values of sound speed and density [15]. The literature values used for water were 0.999g/cc and the corresponding sound speed at 1480 m/s. Table **1** provides the contrast factor calculated using eq. (2) for mineral oil/water and motor oil/water.

Table 1.   Acoustic Contrast of Mineral and Motor Oil

	c (m/s)	ρ (g/cm^3)	ϕ
Mineral Oil	1430	.821	-0.53
Motor Oil	1700	.875	-0.04

When the droplets move closer to the pressure antinodes (pressure node for positive contrast factor), within a few diameters of another droplet, a secondary radiation force that is an attractive force between two spheres in an acoustic field, start to dominate. These interparticle forces drive the droplets together. Smaller droplets become larger droplets, and since the acoustic force is proportional to the particle volume , it can grow rapidly. The secondary radiation force can be expressed as [16],

$$F_s = -\frac{\kappa^2 E_{ac}}{2\pi}\left(1 - \frac{\beta_p}{\beta_f}\right)^2 \frac{V_1 V_2}{d^2} \tag{4}$$

where V_1 and V_2 are the volumes of the interacting droplets, d is the separation distance between the centers of the droplets. As the droplets move through the liquid under the influence of the various acoustic forces mentioned above they experience hydrodynamic drag which is given by the drag force F_d [17]

$$F_d = -4\pi\left(\frac{1 + 2\hat{\mu}/3}{1 + \hat{\mu}}\right)\mu R V^o \tag{5}$$

where μ is the viscosity of the fluid, $\hat{\mu}$ is the ratio of viscosity of the drop to the continuous phase and V^o is the speed of the drop. Table **2** lists viscosity values at 40°C for water, light mineral oil, and 10W-40 motor oil for comparison. Motor oil is an order of magnitude more viscous than mineral oil. Consequently, motor oil takes somewhat longer to separate for the same values of applied power.

Table 2.   Viscosity Values

	Water	Mineral Oil	Motor Oil
Viscosity (cP @40 °C)	0.65	27.5	319

In addition to the interdroplet force, the droplets also experience a radiation force that acts in the direction perpendicular to the direction of the sound beam which is horizontal in the our case. Although this force is estimated to be almost two orders of magnitude smaller than the primary radiation force, it is sufficient to induce clumping of the droplets [18-21]. This force counters the buoyancy force of the droplets.

Although there are plenty of models available to predict the acoustic behavior of oil drop movement in water; there does not exist a model to determine practical implementation of acoustic separation of oil and water for industry use. An objective of this study was to determine if acoustic separation could be performed on stable heavy crude oil-water emulsions. This is a particularly challenging problem because the density difference between water and heavy crude oil is very small. This paper explores the pragmatic performance of oil-water separation experimentally in simple resonant cavities without any artifacts in it by exploring two parameters aforementioned, transducer area by way of shape and frequency.

MATERIALS

All the experiments conducted use Stanford Research System 30 MHz *vs* 345 function generator, ENI 310L RF power amplifier, Tektronix oscilloscope, Alan Industries 50V10 attenuator, and Panametric piezo transducers of center frequencies 1 MHz, 1.5 MHz,. 2.25 MHz. Oil-water emulsions were prepared using an Omni-Ruptor 250 Homogenizer. Oils used for the emulsion were Mineral oil and Pennzoil 10W40 motor oil. The motor oil emulsions were made with the surfactant Triton-x100 (phenol) for stabilization. An HP 4194A impedance analyzer was used for the electrical characterization of the transducers used and to determine the appropriate frequency sweep range.

MINERAL OIL-WATER EMULSION EXPERIMENTS

The emulsions used in these experiments were made of 10% by volume mineral oil in water prepared using the homogenizer. Three different shaped containers were used to setup standing waves in the emulsion: a rectangular quartz Starna (Starna Cells Inc.) cuvette, a cylindrical transducer, and a curved bottom cuvette. The frequency spectra obtained from frequency sweep measurements of the containers with piezoelectric transducers attached on the outside were composite spectra of the fluid-filled cavity and the container wall. Maximum sound transmission into the cavity occurred at the thickness mode resonance frequency of the container wall that is dependent on the material and thickness of the container wall. Therefore, resonant frequency appropriate for each container was used to set up standing waves for the experiment that provided the maximum energy into the cavity. Several transducer arrangements were tested using the rectangular cuvette. The first transducer arrangement used two transducers that were placed on opposite walls of the vertical cuvette (10 mm x 10 mm x 45 mm) filled with the emulsion. Initially, one transducer was used as the source (typical excitation voltage ~ 30V pk-pk) and the other as the receiver to detect the resonance condition (standing wave) in the cavity. The cavity had several resonance frequencies between 1-2MHz and tests were performed at each of these frequencies. At each resonance frequency, mineral oil was observed to separate from the emulsion and form lines inside of the cuvette as in Fig. (**1**). A second transducer setup was

tested which involved connecting both transducers to the power amplifier and placing an adjustable attenuator on one transducer. This was done to observe the effects of applying power from opposite sides of the cavity to modify the standing wave pattern while varying the power on one transducer and keeping the other fixed. The frequency was set to 1.345MHz at 30V pk-pk and the attenuator was adjusted between 1-10dB. In each case, regardless of the signal attenuation, the mineral oil collected as lines in the cuvette by the standing waves produced. In all of the experiments previously described, once the power to the source transducer was turned off the oil could be seen rising to the top of the cuvette rapidly due to gravity. In an attempt to accelerate the rate at which the oil rose to the top, another transducer was placed at the bottom of the cuvette. The bottom transducer was excited at 49 kHz with an excitation voltage of 19.2V pk-pk using a separate function generator and amplifier. The frequency was chosen somewhat arbitrarily. Using the setup with the phase inverter all three transducers were turned on at the same time and after a few seconds only the transducers on the vertical walls were turned off. The presence of the excitation from below decreased the time of oil rising by half as compared to no additional excitation.
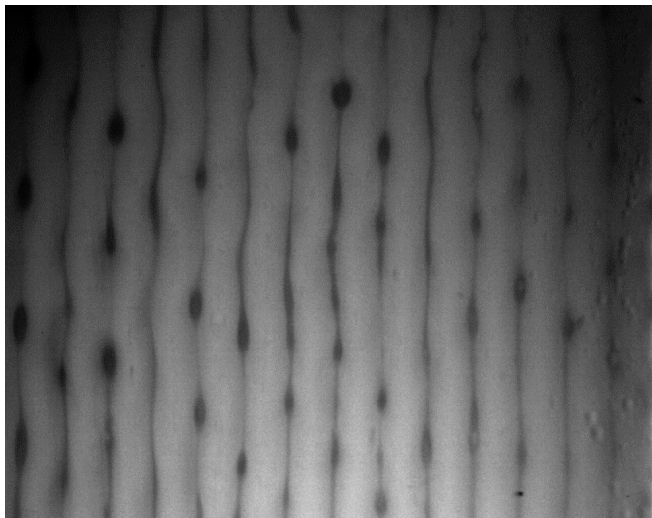

Fig. (1). Oil droplets captured in standing waves at 1.345MHz in quartz Starna cuvette.

Fig. (2). Oil droplets captured in standing waves concentrically at 1.67 kHz frequency modulated in cylindrical piezo transducer.
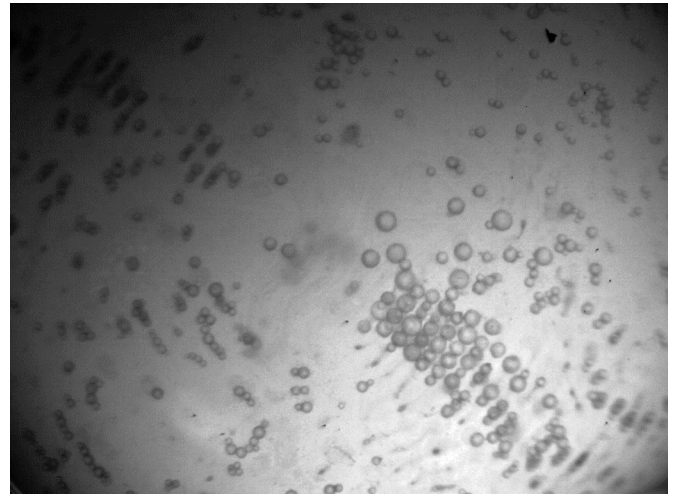

Fig. (3). Large oil droplets forming at 1.84MHz in cylindrical piezo transducer.

Following the experiments in containers, a hollow piezoelectric cylindrical transducer of dimensions: 20 mm height, 18 mm diameter, and 1.09 mm thickness with both inner and outer surface coated with an electrode material was tested to determine if direct contact with the transducer instead of the sound going through the wall of a cuvette enhanced the separation of an oil-water emulsion. The cylindrical transducer was converted into a container by placing a glass slide on the bottom side. The emulsion was poured into this cylindrical container and the signal from the function generator at 2.152 MHz and 6V pk-pk was applied directly to the outer and inner electrodes of the piezo-cylinder. This frequency was in the neighborhood of the thickness mode resonance frequency of the piezo-cylinder and corresponded to one of the cavity resonances. Standing waves of oil in circular patterns as concentric rings were observed because of the circular geometry of the container. Both frequency and amplitude modulations of the excitation signal were used in this setup as a means of controlling the power output and for forcing oil droplets to coalesce more efficiently. The effect of amplitude modulation was studied first; here clear images of concentric standing waves were formed at 1.841MHz (one of the cavity resonances) at 1kHz modulating frequency, as shown in Fig. (**2**). Frequency modulation was tested at 2.122 MHz with a 6 kHz carrier wave at 30V pk-pk. The frequency modulation showed an increase of oil droplet coalescence at the surface of the emulsion and while forming concentric standing waves (Fig. **3**).

The standing waves produced previously were spaced evenly with the spacing determined by the resonant frequency. To produce standing waves at a focal point, and thus intensify the acoustic field, a cuvette was constructed using half of a hollow cylindrical piezo transducer. This U-shaped piezo with its axis horizontal was used as the bottom of the cuvette and glass rectangles were adhered to make the walls, resulting the dimensions of 24 mm x 25 mm x 35 mm. The air-water interface was used as a reflecting surface for producing acoustic standing waves. Standing wave patterns of oil were visibly produced in the 10% mineral oil emulsion at 2.173 MHz and 2.25MHz set to 15.8V pk-pk (Fig. **4**).
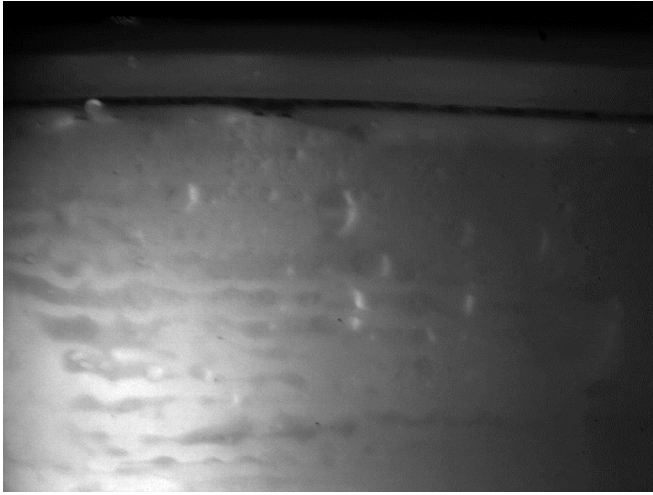
Fig. (4). Trapped oil in 2.173MHz curved cuvette

MOTOR OIL-WATER EMULSION EXPERIMENTS

In recovering oil from an oil-water emulsion, different viscosities of oil need to be considered. Motor oil 10W40–water emulsions were made using 1% by volume of surfactant in water. To determine the effectiveness of the surfactant and the state of these emulsions; the speed of sound was measured for varying amounts of oil in water. The sound speed was determined using a frequency sweep technique under ambient conditions in a rectangular cuvette [22].

Measurements were made at 5%, 10%, 15%, 20%, and 25% motor oil by volume emulsions as well as pure water and motor oil. The speed of sound was compared to the Urick model,

$$v = \frac{1}{\sqrt{\rho_m^2 \kappa_m}}$$
$$\rho_m = \phi \rho' + (1 - \phi) \rho \tag{6}$$
$$\kappa_m = \phi \kappa' + (1 - \phi) \kappa$$

where κ' is the compressibility of oil and κ is the compressibility of water, and ϕ the volume fraction. Meng *et al*. [23] showed that Urick Model is adequate for prediction of speed of sound in crude oil-water emulsion. The Urick model assumes the suspended particles are infinitesimally small compared to the wavelength of sound, and the effects of scattering neglected. Density and compressibility are averages weighted by phase concentration. As expected, the emulsion experiment values closely follow the Urick model, establishing that the starting condition is an emulsion instead of a stirred mixture (Fig. **5**).

Measurements with motor-oil emulsions were very similar to that of mineral oil and the results were identical. To enhance the strength of the standing waves, an aluminum reflector was used instead of the previous air-water interface and it indeed produced a very strong standing wave pattern (Fig. **6**).

The cylindrical container used in the mineral oil experiments was modified for the motor oil emulsion for visibility. An acrylic cylindrical container was fabricated and a cylin-

drical transducer was placed inside the acrylic holder, as shown in Fig. (**7**). The emulsion was poured into the cylinder where some of the emulsion was in the concentric region between the container and transducer. The emulsion was held at the resonance frequency (1.93MHz at 36V pk-pk) for 2 minutes. Standing waves were seen forming during the insonification process (Fig. **8**). Once the power was turned off and the transducer removed, an oil layer could be seen forming at the top of the emulsion immediately.
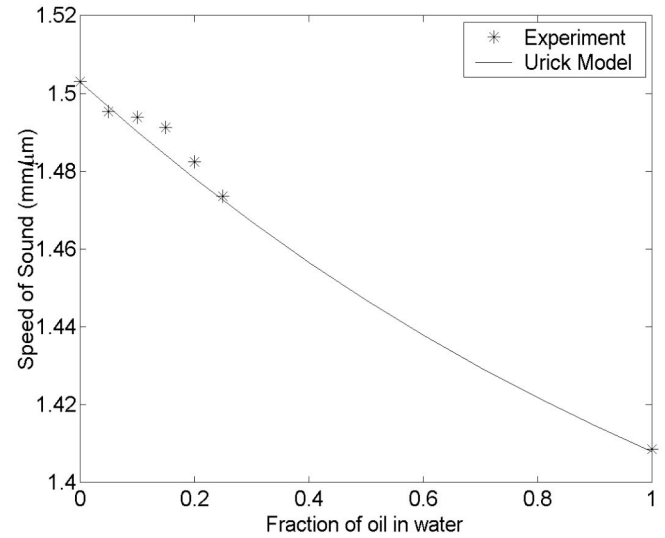

Fig. (5). Fraction of Motor oil in water *vs* speed of sound.

Fig. (6). Motor oil emulsions; lines are motor oil trapped in standing waves at 2.253MHz in curved cuvette.

Following this experiment a separate two acrylic container holder was made to study the difference in oil accumulation with and without acoustic exposure. Equal amounts of motor oil emulsion were placed into each of the containers and the piezo transducer was placed inside one of the containers. A downward ramped frequency sweep at 22V pk-pk was sustained for 1, 2, 3 and 4 minutes. The results showed that over the prolonged exposure the oil collected at the top of the liquid was significantly more for the acoustically treated emulsion than without (Fig. **9**). Comparing amplitude modulated, ramped frequency and static frequency at residence time of 4 min with the same conditions as before; all
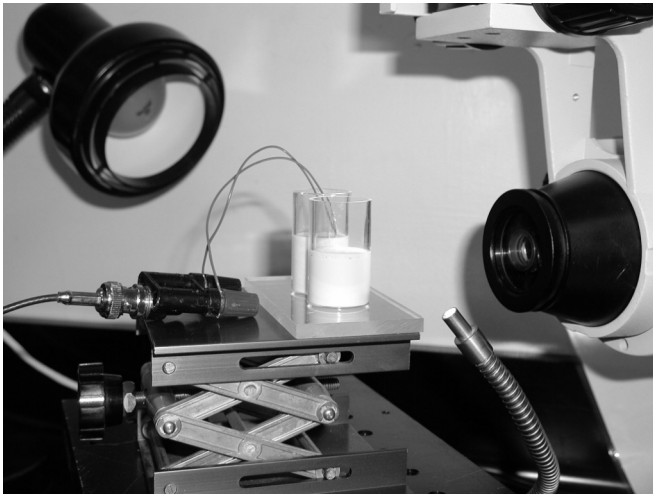
Fig. (7). Experimental setup.

Fig. (8). Motor oil trapped in standing waves in cylindrical holder sandwiched by piezo cylinder transducer.
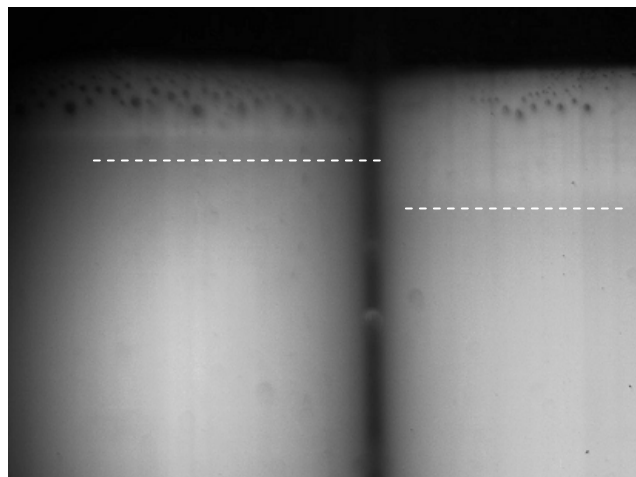

Fig. (9). Control on the left and experiment on the right showing motor oil levels recovered after 4 min of having applied acoustic field.

the results showed a significant increase in oil accumulation compared to the control, however when compared to one another all three experiments looked the same in terms of oil layer thickness. A power level experiment was conducted

where for 4 min the frequency was held at 1.94MHz and the power levels were varied at 18V, 32V, and 52V pk-pk. The results of this experiment showed a large increase in the thickness of oil layer on the emulsion when treated acoustically compared to the control and compared to the other experiments conducted.

CONCLUSIONS

Oil-water emulsions were made using mineral oil and 10W40 motor oil. Several transducer configurations were explored for producing standing waves in the emulsions. In all transducer geometries and configurations it was observed that oil droplets could be trapped and coalesced in standing waves produced to separate oil from emulsions. In a side-by-side experiment, the acoustically stimulated sample showed a significant difference in oil separation compared to the control sample. In the case of motor oil-water emulsion, the contrast factor is low and the viscosity is significantly higher than that of mineral oil. However, it was still possible to separate this emulsion in simple resonant cavities without having to employ any porous media or other artifact in the cavity and at reasonable powers. Too much power would have induced both streaming and heating.

In regards to practical implementation of acoustic for liquid-liquid separation, direct contact of transducer to liquid was observed to be most effective in separating emulsions. Surface area in relation to volume appears to have a more dominant role rather than shape in effectively separating oil water emulsions, however shape can play a part in the practical implementation of acoustics for liquid-liquid separation. Variation of frequency, power, and wave function showed a positive effect in coalescing oil droplets in standing waves.

ACKNOWLEDGEMENT

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