Study of the effects of ultrasound standing waves in an oil-in-water emulsion

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Phase separation in a resonant cavity due to the application of ultrasound is studied in this paper. A series of experiments are developed in order to characterize the wave transmitted into medium and to analyze the effects produced in an oil-in-water emulsion. An arrangement of oil droplets in antinodal planes is observed together with the consequent formation of oil aggregates in the medium, as predicted by the theory. A measure of the temperature variation is also carried out to determine the dependence of the attenuation of the wave on frequency.

I. INTRODUCTION

Phase separation is a widely studied process by the scientific community. Engineers are acquainted with separation technologies such as distillation columns, membrane filtration and centrifuges. These processes are usually highly invasive and can have significant effects on the physical properties of the particles which are being set apart. Nevertheless, there exists a rising separation method which relies on stationary acoustic waves at ultrasound frequencies. The fact of using frequencies above megahertz implies less energy absorption by the medium and, consequently, a lower risk of manipulating its physical features.

The main issue of the mentioned technique is its large scale application, as the forces involved have a marked nonlinear behaviour and they are second order forces produced by the mechanical wave. Therefore, other higher order forces can easily become dominant when instrumental scale is increased. Another reason of its difficult implementation is the fact that this technique depends on a large number of non controllable parameters (density, propagation speed, particle or droplets size, viscosity, compressibility...), which implies the need of more specific equipment depending on the samples.

Separation based on ultrasonic waves is being introduced in fields such as petrochemistry, pharmacology and biomedicine because of its non invasive nature, and also in microgravity setups, due to the fact that compressibility influence is prevalent over density. Another field of industrial interest is oil separation in an oil-in-water (O/W) emulsion.

This paper focuses on this latter application, proving the effects predicted by the theory when high frequency acoustic waves are applied to an emulsion of these characteristics. In the first place, the experimental setup is verified by using lower frequencies, of the order of kilohertz, which enables the characterization of stationary waves using an hydrophone. Once validated, high frequencies are used and effects are studied.

II. THEORY

Stationary waves can be generated by means of resonant cavities. These cavities take advantage of the reflection of a plane wave in order to obtain a second wave, with the same amplitude and frequency of the original one, which will interfere with it. The exact phase difference is only achieved for a value of the cavity length which satisfies the following equation:

$$L = n\frac{\lambda}{2} (n = 1, 2, 3...)$$  

being $L$ the length of the cavity in the direction of propagation of the wave and $\lambda$ the wavelength. The value of $n$ determines the number of nodes and antinodes inside the cavity. This mathematical model can be applied to pressure waves created inside a medium. The waves are generated by using transducers based on piezoelectric properties (the volume of the material is changed according to the received electric potential). When an electric signal with harmonic temporal dependence is applied to the transducer, it produces a pressure wave into the medium which is in contact with.

The application of ultrasonic waves ($f > 20kHz$) to a fluid might produce various effects, depending on the working frequency interval. The first one ($20 - 100kHz$) is responsible of the phenomenon known as acoustic cavitation. It relies on the creation of small gas bubbles due to the high pressures applied, producing collapses and releasing a large amount of energy in the form of heat. This heat transmission is predominant for frequencies lower than 100kHz, inasmuch as for higher frequencies bubbles are smaller and, as a result, less energy is released.

The following energy range is the one corresponding to high frequencies ($1 - 4MHz$), for which cavitation is negligible. When a stationary wave at these frequencies is applied in a medium, two main forces appear on particles and droplets inside the fluid. These are called ‘acoustic radiation forces’ and their effects depend on the size, density and compressibility of particles or droplets. A brief explanation of both of them and their effects is given below.
The 'Primary Radiation Force'\textsuperscript{11–12} is the first one that can be noticed and is shaped by the following equation:

$$F_{ac} = -\frac{4\pi}{3} R^3 k E_{ac} \phi \sin (2kx)$$  \hspace{1cm} (2)

where $R$ is the particle or droplet radius, $k = 2\pi/\lambda$ is the wave number, $E_{ac}$ is the specific energy density (which is proportional to the square of the wave amplitude), $x$ refers to the distance from the closest node and $\phi$ is the acoustic contrast factor\textsuperscript{12}.

The sign of the acoustic contrast factor determines whether the particles move towards the nodes or the antinodes. This factor presents a dependence on the density and compressibility of both media (the dispersed and the continuous phase) according to the equation:

$$\phi = \frac{5\rho_p - 2\rho_m}{2\rho_p + \rho_m} - \frac{\beta_p}{\beta_m}$$  \hspace{1cm} (3)

with $\rho_i$ being the density, $\beta_i$ the compressibility and the subscripts $m$ and $p$ refering to the medium and the particle or droplet, respectively.

Once particles are arranged in nodes or antinodes, a second force among them appears, known as the 'Secondary Radiation Force'\textsuperscript{13}. It produces an attractive force between particles or droplets regulated by the equation:

$$F_s = -\frac{\rho_m}{8\pi} (\omega P_a)^2 (\beta_m - \beta_p)^2 \frac{V_1 V_2}{d^2}$$  \hspace{1cm} (4)

where $d$ is the separation between two particles, $V_i$ is the volume of the $i$-th particle and $\omega$ and $P_a$ refer to the frequency and amplitude of the incident wave respectively. This force tends to generate aggregates of particles or droplets of size larger than the separation between lines previously produced.\textsuperscript{14}

When particles start moving, they are affected by viscous forces described in terms of the drag force\textsuperscript{15}, $F_d$:

$$F_d = -4\pi \left( \frac{1 + 2\mu_p/3}{1 + \mu_p} \right) \mu_m R \nu$$  \hspace{1cm} (5)

where $\mu_i$ refers to the viscosity of the medium and $\nu$ is the speed of the particle. Particle dynamics is the result of the final sum of the different forces. By looking at the equations, a tradeoff between particle size, applied frequency, media viscosity and wave amplitude can be observed\textsuperscript{16–17}. As a result, particles with different size and characteristics will be affected in a different way. Those with the same or higher size than $\lambda$ will be affected by getting divided into smaller particles, depending on the amount of nodal (or antinodal) planes that they cross. On the other hand, for very small particles or droplets the drag force will be larger than the acoustic forces and hence their dynamics will be slow enough so as not to produce any observable phase separation effect. However, the secondary radiation force presents a slender dependence on particle distance and size. Consequently, once aggregates are formed in the nodes or antinodes, smaller particles will be moved towards them and, therefore, will take part in the separation process.

Additional effects can be recognized when the cavity length is much larger than $\lambda$. For instance, the effect of 'acoustic streaming'\textsuperscript{18–20}, which refers to the flows generated in the whole fluid due to the applied wave. Therefore, particles are dragged through the fluid, assisting the union of aggregates from different nodal (or antinodal) planes or breaking them if they are large enough. It results in a net flow of aggregates in the direction of propagation of the wave. The existence of these flows can be explained by the attenuation of the wave inside the medium, which produces a velocity gradient due to the distortion of the standing wave. The acoustic attenuation coefficient for plane waves is given by the equation:

$$\alpha = \frac{2\mu (2\pi f)^2}{3\rho_m c^3}$$  \hspace{1cm} (6)

with $c$ being the speed of sound in the medium. The energy lost by the wave is mainly dissipated as heat. In spite of what it may seem more intuitive, the coefficient only depends on the frequency and viscosity of the medium and not on the amplitude of the wave.

### III. EXPERIMENTS AND RESULTS

#### A. Study of ultrasound waves in a resonant cavity

Due to the difficulties of working at high frequencies, the study of the propagation of acoustic waves and their characterization was carried out at lower frequencies. In this first part of the experiment a cylindrical piezo transducer with a nominal frequency of 130kHz and a 4.8cm x 2.5cm cavity were used. The function generator (Agilent 33210A) produced a 130kHz and 1V pk-pk signal which was amplified by an amplifier (Falco Systems WMA-300) with a 50x gain and applied to the piezo transducer. In order to characterize the standing wave originated into the cavity, a measure of the pressure amplitude at different points of the path between the walls was made by using an hydrophone (Onda HCT-0300).

<table>
<thead>
<tr>
<th>Media</th>
<th>Wavelength (mm)</th>
<th>Speed (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Distilled water</td>
<td>11.6</td>
<td>1508</td>
</tr>
<tr>
<td>Mineral oil</td>
<td>10.3</td>
<td>1335</td>
</tr>
<tr>
<td>Emulsion</td>
<td>11.2</td>
<td>1451</td>
</tr>
</tbody>
</table>

Table 1: Wavelength and velocity of sound waves in different media.
This procedure was repeated for three different media: distilled water, mineral oil (Nenuco body oil) and a 10% (v/v) mineral oil-in-water emulsion. The latter was prepared by using a magnetic stirrer (Iseline MS-H-S) and adding commercial soap as a surfactant in order to assist the emulsion process. In all cases a stationary wave could be observed from data treatment and wavelength could be determined. Results of wavelength and speed values in each medium were presented in table 1, with an error of 4% due to the limited precision while taking measures of the amplitude.

B. Separation of oil-in-water emulsion by ultrasound

As already mentioned, separation of an oil-in-water emulsion takes place at frequencies above 1MHz. Therefore, a higher nominal frequency piezoelectric transducer and a smaller cavity were needed. The piezo used presented a nominal frequency of 2.035MHz, which was determined by means of an impedance analyzer (Agilent 4291A). This frequency refers to the one that corresponds to a minimum of impedance, as in that way a larger amount of power is transmitted to the cavity. Figure 2 shows a plot of the impedance modulus as a function of the frequency. Nevertheless, when the transducer is attached to the cavity, the nominal frequency might vary. In order to correct the preceding value, an electric signal with varying frequency (starting from 1.9MHz and with $\Delta f = 0.001$MHz) was sent from the function generator to the amplifier, where it was divided into two signals: the first one was derived to the piezo, whereas the second one was connected to an oscilloscope (Agilent D50-X 2002A). A minimum in impedance of the piezo implies a lower rebound of power and, thus, a lower peak-to-peak voltage is measured in the oscilloscope. The new value of nominal frequency (2.012MHz) corresponded to that minimum in amplitude.

The experimental setup is shown in figure 1. The 2.012MHz piezo transducer (with a diameter of 2cm) was attached to a 5.4cm x 3.6cm methacrylate cavity. The 10% oil-water emulsion was introduced inside the cell and a 2.012MHz and 50V pk-pk signal was sent to the piezo. Macroscopic and microscopic effects were observed, the latter using a microscope, and were recorded by means of a high speed camera (Optronics CRx2 Camera Series).

It was first observed that after 2 or 3 seconds (depending on the amplitude applied) a pattern of lines perpendicular to the wave propagation direction appeared in the sample. By using the microscope it could be seen that lines were formed by white droplets joining together, which corresponded to a mixture of mineral oil and the soap used. This observation is in accordance with the primary force theory given in eq. 2, as it was expected that droplets from the dispersed phase (oil) would be arranged in pressure nodes or antinodes, depending on the acoustic contrast factor. Using eq. 3, an estimation of this factor for mineral oil/water can be made, which leads to a value of $\phi = -0.82$. Therefore, oil droplets are driven towards pressure antinodes and the observed lines corresponded to antinodal planes. An image of three antinodal planes taken with the microscope can be seen in figure 3, which has been treated (Adobe Photoshop) in order to make the lines more visible. Distance between planes, $d$, can be measured and it should correspond to half of the wavelength of the ultrasound. By image processing (Adobe Photoshop) and averaging results from different pictures an estimation of $d = 0.49$mm was obtained. The expected value can be determined from the speed calculated in the previous experiment (with the emulsion...
FIG. 3. Microscopic image of three antinodal planes. The image was taken with illumination coming from under the sample. Thus, the white aggregates forming the lines are now seen as black, as a less amount of light passes through it.

FIG. 4. Microscopic image for a frequency of 2.2 MHz. The appearance of the secondary radiation force causes the accumulation of oil droplets at antinodal planes.

as the medium). For a frequency of 2.012MHz it leads to a value of \( \lambda = 0.72 \text{mm} \), so distance between antinodes is 0.36mm. Even though there is an error of 36%, the order of magnitude corresponds to the one expected. There are different possible sources of the error, but it is mainly the difficulty of measuring the exact distance between planes, as there are not clear lines but aggregates of droplets in continuous movement among antinodes. Moreover, there was a limitation of precision in distance measuring and a previous error coming from the speed calculation.

The same procedure was repeated for higher frequencies (with increments of \( \Delta f = 0.01 \text{MHz} \)) in order to study the variations produced in the sample, considering that the primary force increases with frequency. Nevertheless, the arrangement of bubbles at antinodal planes diminished with increasing frequency, as it was getting further away from the piezo nominal frequency. However, for a frequency of 2.2MHz a clear effect was seen (figure 3). The arrangement of droplets in antinodal planes was more visible than for a 2.012MHz frequency (figure 4). Moreover, it could be more easily appreciated that droplets were forming aggregates as the time passed. This effect could be a consequence of the secondary radiation force, but it could also be produced by interferences in the acoustic wave that generate a pressure gradient in directions nonparallel from the propagation of the emitted wave\(^{21}\). This phenomenon at a frequency different from the nominal of the piezo could be explained by taking into account that it might be exactly the resonant frequency of the cavity. Moreover, by looking at the graphic of the piezo transducer impedance (figure 2) it can be seen that this frequency corresponds to one of the minimums of impedance. After 5 minutes, the signal coming from the function generator was stopped and aggregates which lined up started to spread over the whole cavity. In the final sample conglomerates of oil droplets could be distinguished from water.

This variation in frequency also enabled the observation of a displacement of the oil aggregates in the direction of propagation of the wave. It could be seen that the movement was done by steps between consecutive antinodal planes and they were produced because of the acoustic streaming phenomenon. In order to go a little bit deeper into this effect, a third experiment was carried out to determine the variation of the attenuation coefficient as a function of the frequency.

C. Study of the temperature variation due to ultrasound application

Using the same setup as in the previous experiment, variations of temperature inside the medium were determined. Hence, it could be studied the modification in the medium absorption as a function of the frequency (eq. 6). For that purpose, a thermocouple connected to a multimeter (Agilent U2131A) was placed in different points inside the cavity. A 2.012MHz wave was first applied and an averaged increase of 0.2°C was obtained after repeating the measure 20 times with a settling time of 4.2s. Before changing the frequency, the procedure was repeated with an amplitude and no change in temperature increase was observed, but settling time was
A new measure was carried out with a nominal frequency of 2.2MHz and an increase of 0.4°C was observed. Consequently, a significant relation between frequency and dissipated energy was evidenced, since this energy is mainly transferred as heat, thus producing an increase of temperature in the medium. It could be thought that this phenomenon is due to the fact that the piezo transducer becomes heated as it is not working at its nominal frequency, but heating in the piezo was always produced later than in the medium. Therefore, this results are consistent with eq. 6, as they show both the connection between frequency and medium absorption and the non dependence of the latter on the amplitude of the wave.

V. REFERENCES


IV. CONCLUSIONS

It has been proved that a rectangular methacrylate cavity attached to a piezoelectric transducer can produce stationary waves when an harmonic electric signal is applied. The effect of a 2.012MHz signal through the transducer to a 10% oil-in-water emulsion is a phase separation of the fluids, which leads to the arrangement of oil droplets at the antinodes. An increase of frequency up to 2.2MHz produces the creation of oil aggregates. Moreover, it has been detected a displacement of them towards the direction of propagation of the wave. Afterwards, the variation of temperature has been measured for both frequencies and it has been proved that a higher increase is produced for a frequency of 2.2MHz. The modification of the amplitude does not lead to a change in temperature but it does show a decrease in the settling time.

All the observed effects have been predicted by the theory. The existence of a primary radiation force produces the agglomeration of oil droplets at the antinodal planes. The secondary radiation force, which is proportional to the square of the frequency, can be macroscopically observed when an increase of frequency is produced and it is the responsible of the creation of oil aggregates. A frequency increment also produces a variation in temperature and droplet movement as a result of the dependence of the index of attenuation of the wave on the frequency. This coefficient is significantly related to the acoustic streaming phenomenon and to the amount of dissipated energy as heat.