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Acoustic separation of oil droplets, colloidal particles and their mixtures in a microfluidic cell

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Graphical Abstract

Research Highlights:

- Simultaneous separation of oil droplets and solid particles in aqueous mixtures of the two using acoustic standing wave patterns was demonstrated
- In-situ macroscopic and microscopic visualization of acoustic separation in a microfluidic cell using dodecane emulsion droplets, fine silica particle and their mixture as model systems
- Proof of concept experiments of continuous fluid flow acoustic separation

Abstract

Here we report direct macroscopic and microscopic observations of acoustic driven separation of dodecane oil droplets in water in the presence and absence of colloidal silica particles suspended in the water phase. The experiments were conducted in a simple rectangular channel glass microfluidic cell in which an ultrasound standing wave pattern was generated at 300 KHz frequency. The separation process of both oil droplets and colloidal particles inside the cell was recorded using a high-speed video camera equipped with a macro-objective lens for macroscopic observation or with a high-speed camera attached to an inverted optical microscope for a higher resolution microscopic observation. We characterize the clustering process in the case of emulsion droplets or solid colloidal particles and ultimately demonstrate the emulsion droplets separation from the solid particles in the mixtures based on their different acoustic contrast factors. Finally, we conduct proof of concept experiment to show that the same approach can be used in a continuous fluid flow process.

Keywords: Acoustic separation, Microfluidics, Emulsion droplets, Silica colloids, High-speed imaging

1. Introduction

The use of ultrasonic waves to separate particulates from solutions is a promising alternative to established separation processes, such as distillation, filtration or centrifugation, with a range of potential applications in food processing, pharmaceutical, oil production and biomedicine industries [1-4]. Some of the advantages of the high frequencies acoustic separation compared to traditional separation methods are the fast action, robustness of the process and low mechanical impact on the particulates to be separated. The last two decades have also seen rapid developments of microfluidic devices with integrated miniaturized acoustic piezo-electric-transducer (PET), known as acoustofluidics [5-17]. The majority of the acoustofluidics applications are related to the manipulation of biological samples using acoustophoresis [15], acoustic trapping [16] or acoustic streaming [17].

Acoustic separation of particulates dispensed in a fluid is attained by exciting a standing wave pattern in the dispersion. The effectiveness of acoustic separation is determined by the acoustic contrast factor, which is a dimensionless combination of the particulate and medium density and compressibility (see Eq. 2). In a standing acoustic wave, particulates that have a positive acoustic contrast factor, e.g. solid particles in water, are drawn towards the pressure node planes. On the other hand, particulates of negative acoustic factor, e.g. oil droplets in water, are drawn towards the pressure antinode planes. Most of the studies reported in the literature so far have used only one type of dispersed particulates, such as solid particles, biological cells, or emulsion droplets. The objective of the present study is to use a simple microfluidic flow cell experimental setup to demonstrate the acoustic separation for a mixture of oil droplets and solid colloidal particles, due to the opposite acoustic contrast factor of the two components in the mixture.

As a model emulsion, we use dodecane oil droplets dispersed in water, and as model solid particles we use monodispersed colloidal silica microspheres ranging from 0.15 μm to 7.5 μm in diameter. Such a combination of light hydrocarbon oil droplets and solid silicate particles is representative of produced water streams encountered in crude oil production. Prior ultrasound separation studies have targeted a wide range of oil production related applications, such as tar recovery from oily sands, desalination and dehydration of crude oil, and oil separation from water [18-21]. The rapid development of acoustic microfluidics based test platforms, as targeted in the present work, will hopefully contribute to the faster implementation of ultrasound assisted industrial scale applications. The use of microfluidic system allows the fast and unexpansive scan of a large number of operational conditions. Such proof-of-concept small scale experiments are of particular importance for oil industry related applications due to the high cost of the modification of existing process flows.

In the present investigation, we use a simple flat parallel plate channel glass cell with an aperture of 100 μm , which allows direct microscopic and macroscopic visualization of the response of particulates to ultrasound. A combination of high-resolution microscopic and wide-view macroscopic techniques integrating a state-of-the-art high-speed video camera was utilized for the process visualization. First, we investigate the simpler case of a single component emulsion of droplets or a suspension of solid particles. For the case of solid particles, monodispersed silica particles were used to allow us to establish the minimum particulate size for effective acoustic separation in our experimental setup. Next, we investigate simultaneous acoustic separation of both emulsion droplets and solid particle in water. Finally, we conduct proof-of-concept experiments demonstrating the continuous fluid flow separation of the dodecane droplets and silica particles.

2. Experimental setup

Oil-in-water emulsions used in the current experiments were prepared by mixing between 1 to 5 wt % dodecane oil (99.0+%, Aldrich) with DI water. To stabilize the emulsion droplets, 3×10^{-3} wt % oil-soluble SPAN 80 surfactant (Aldrich) was added to the dodecane phase, resulting in water–oil interfacial tension of about 7 mN/m [22]. The use of surfactant that is only soluble in the oil phase minimizes the generation of acoustic bubbles in the emulsion [23]. Two mixing protocols were used to prepare coarse and fine oil droplets. The coarse droplets were prepared by stirring the mixture for 5 minutes at 15000 rpm with a propeller disperser (IKT T18), resulting in oil droplet sizes in the range 2 to 10 μm (see for example Fig. 3a). To prepare the finer droplet emulsion, the samples were further dispersed using a 20 KHz horn-type-transducer ultrasonic homogenizer (Cole-Parmer CPX 500). About 5 minutes of pulsating irradiation at 200 Watts resulted in finer emulsion droplet sizes in the range from 1- to 3- μm (see for example Fig. 4a). To minimize the effect of gravitational stratification, the emulsion samples were used within 15 minutes following preparation.

In a limited number of experiments we used as the oil phase, perfluorohexane in the commercial form known as PP1 (Flutec PP1; F2 Chemicals Ltd., U.K.). In this case, the emulsion was stabilized by adding to the water phase 1×10^{-3} wt % of Zonyl FSN (DuPont) which is a nonionic fluorosurfactant that modifies the interfacial energies at very low concentrations [24]. In preparation of the perfluorohexane emulsions, we used the same mixing protocol as that for the fine dodecane droplets emulsion given above. This resulted in a similar size range for the perfluorohexane droplets of 1 to 3 μm .

Monodisperse silica particle suspensions of sizes 0.15 μm , 0.5 μm , 1.2 μm and 7.5 μm were purchased from MicroParticles GmbH as 5 wt % aqueous suspensions. Before use, the particles were washed from the original suspension solution to remove surfactant traces. The concentration of silica particles in the DI water suspension used in all experiments was 1 wt %, if not otherwise specified.

Figure 1 is a schematic of the experimental setup. We use a parallel-plates fluid flow cell (Starna 48/Q/0.1) with channel internal dimensions of 100 μm thickness, 8 mm length and 38 mm width. A

radial-mode piezo-electric ceramic disc transducer (Steminc, SMD07T03R411) of 7 mm diameter and 3 mm thickness, having resonant frequency of about 300 kHz, was attached at the bottom of the cell using an epoxy resin (Fig. 2 pictures). The transducer is powered using an Agilent 33220A function generator, and HAS 4012 bipolar amplifier. In all experiments, we operated the transducer at 300 kHz, with a sinusoidal signal of 1 V amplitude from the signal generator, giving a 40 W amplified input signal that generates about 11 W/cm² intensity in the front of the transducer. The resonance frequency of the transducer was chosen so that a few sound wave wavelengths can fit in the channel's width, which makes it convenient for observations with our experimental setup.

The cell is mounted on the observation stage of a Carl Zeiss, Axio Observer inverted microscope. A high-speed video camera, Photron SA5, is attached to the side port of the microscope allowing recording of bottom view observations of the irradiation process using 10×, 20×, or 40× magnification objectives. Another high-speed camera equipped with an 85 mm Nikon AF NIKKOR lens is mounted on a vertical rail at a focal distance of about 20 cm above the cell, for the top-view macroscopic observation of the irradiation process over the entire cell. The cell inlet is connected by plastic tubing to a 10 ml syringe used to introduce the solutions to be studied into the cell. Most of the experiments were done at static flow conditions by closing the inlet and outlet tubing after filling the cell with the desired solution. Controlled flow experiments were done by mounting the syringe to an automated syringe pump (Fusion 200, Chemyx, Inc).

Experiments at each solution condition studied herein, e.g. concentration and size of the emulsion droplets, concentration and size of the silica particles as well as their mixture ratio were repeated for at least three independent runs, to confirm the reproducibility of the separation patterns.

3. Background

Here we make a brief reference to the physical background of the acoustic separation involving the primary and secondary acoustic radiation forces and acoustic streaming [1]. The primary acoustic radiation force arises from the spatial gradient of the acoustic wave pressure. In a standing wave field the force in the direction of the wave the primary radiation can be expressed as:

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

where R is particle radius, $k = 2\pi/\lambda$ is the wavenumber, λ is the wavelength of the sound, E_{ac} is the acoustic energy density, x the distance from the nodal point and ϕ is the acoustic contrast factor. The direction of the primary acoustic radiation force experienced by a particulate dispersed in a medium is determined from the sign of the acoustic contrast factor [25, 26]:

$$\phi = \frac{5\rho_p - 2\rho_m}{2\rho_p + \rho_m} - \frac{\beta_p}{\beta_m} \quad (2)$$

where ρ is the density, β is the compressibility and the subscripts p refers to the particulate (emulsion droplet or colloidal particle) and m to the medium. The compressibility is related to the speed of the sound, c , in the medium or in the particle:

$$\beta = \frac{1}{\rho c^2} \quad (3)$$

When subject to a standing acoustic wave field a particulate with positive acoustic contrast factor experiences a primary acoustic radiation force that drives it toward the pressure nodes in the acoustic field, e.g. zones with minimum pressure amplitude variation. Reversely, particulates with negative acoustic contrast factor are driven toward the antinodes in the acoustic field, which have maximum pressure variation. Table 1 gives the specific density and speed of sound for the particulates used in our study (dodecane droplets, perfluorohexane droplets, and silica colloidal particles) and their respective acoustic contrast factor when dispersed in water. Dodecane and perfluorohexane droplets have a negative acoustic contrast factor, and the silica particles have a positive acoustic contrast factor in water medium.

The secondary radiation force arises from the acoustic field radiative scattering from the suspended particles, due to the difference in the particle compressibility from that of the medium, and becomes significant once the particles are brought closer together. The secondary radiation force depends on the position of the particles with respect to the acoustic wave propagation direction. The force is repulsive between particles in the direction perpendicular to the wave front propagation but attractive in direction along the wave front propagation [1].

In addition to the acoustic radiation force, acoustic streaming is also induced on the suspended particles. Streaming arises because of the dissipation process in the fluid phase and in the fluid – solid interface. There are several types of streaming which are pronounced at different length scales: large scale Eckart streaming (scale $\gg \lambda$) and small scale Rayleigh (scale $\sim \lambda$) and Schlichting (scale $\ll \lambda$) streaming [17]. In our experiments, based on the lateral dimensions of the cell, and the acoustic wave frequency, the most significant effect is expected to be the Rayleigh streaming. However, because the cell is only 100 μm thick, Schlichting streaming occurring in the boundary layer of the upper and lower wall could also impact the streaming flows in the cell. The acoustic radiation forces tend to be dominant for larger particulate sizes ($> 1 \mu\text{m}$) and lower irradiation frequencies ($< 1 \text{ MHz}$), and the streaming force is more pronounced for smaller particle sizes ($< 1 \mu\text{m}$) but higher acoustic frequencies ($> 1 \text{ MHz}$) [27-29].

4. Results and discussions

4.1 Oil in water emulsion

First, we examine the acoustic separation of dodecane emulsion droplets in the microfluidic cell. Figure 2 shows snapshots from supplementary Video 1, which is a top-view macroscopic observation of the rearrangement of the fine dodecane oil droplets in water after 40 and 600 seconds from exciting a

standing wave pattern in the cell. Prior to acoustic excitation, the oil droplets were uniformly distributed in the water (Fig. 2a). A few seconds after exciting a standing wave pattern, we observe the formation of a network comprised of dark spots connected by thin circular lines (Fig. 2b). The dark spots represent clusters of the oil droplets, while the thin lines represent linear chains of the oil droplets. These thin lines gradually fade as the droplets migrate toward the dark spots and, about 5 minutes after the start of the irradiation mostly discrete dark spots are observed as seen in Fig. 2c. Figure 2c and 2d are comparing the result of two independent experimental run to demonstrate the reproducibility of the characteristic separation patterns. The characteristic distances between the dark spots seen in Fig. 2c and Fig. 2d is about 2.5 mm, closely matching half the wavelength of the acoustic wave of $f = 300$ kHz in water, which is $\lambda/2 = c/(2f) \sim 2.5$ mm. Because of the negative acoustic contrast factor of the dodecane in water droplets (Table 1) the oil droplets are expected to have migrated to the pressure antinodes of the standing wave pattern. The observed distribution of oil droplets within the standing wave pattern is the result of both direct radiation forces and Rayleigh streaming. Apparently, 2D or 3D standing wave patterns are generated in the channel, with the dark spots positioned at the interception of the pressure antinode planes. Circulatory Rayleigh streaming around the antinodes helps in directing the oil droplets toward these interception antinodes where they accumulate [30]. Due to the inhomogeneity of the test cell walls, the standing wave patterns are not exactly the same in all directions, and so are the regions around antinodes where Rayleigh streaming is taking place. In our configuration the pressure antinode positions are close to the cell side walls, with three more lines of antinodes fitting in the space between. The position of the antinodes is slightly disordered because the shape of the cell is not an exact resonant cavity, but having curved end walls. Nevertheless the expected characteristic spacing of $\lambda/2$ between the cluster structures can be clearly observed. We note that this type of experiment can be used as a simple method to visualize the acoustic field pressure distribution in the cell, as alternative to particle image velocimetry (PIV) imaging [29].

The macroscopic observations give the general picture of the acoustic field pressure distribution in the cell. The inverted microscope high-speed video recordings provide more detailed observations of the emulsion droplets aggregation process at higher magnifications. Supplemental Video 2 is an example of cluster formation in the case of coarser dodecane emulsion droplets, recorded using a 20× magnification objective. Video 3 shows another example for a higher concentration of finer dodecane droplets size emulsion taken using 10× magnification objectives. Snapshots from Video 2 and Video 3 are shown in Fig. 3 and Fig. 4 respectively. As predicted by Eq. 1, the larger droplets aggregate faster toward the antinode position, i.e. in seconds. In this case, we can also clearly observe strings of large droplets moving toward the cluster (Video 2, Fig. 3c). In the case of the higher-concentration fine emulsion, the process takes longer (minutes) (Video 3, Fig. 2 and Fig. 4). Following the interruption of the ultrasonication, the oil droplets aggregate relaxes on the upper surface of the fluid cell driven by buoyancy. One simple way to further manipulate the deposited aggregates is by switching on and off the acoustic field. Video 4 is one such example, showing a few cycles of cluster relaxation (Fig. 5a) and cluster aggregation in the pressure antinode (Fig. 5d) upon the switching on of the ultrasound.

4.2 Colloidal silica particles suspension

Here we discuss observations from experiments conducted with colloidal silica particle suspensions. These monodisperse particles allow a more precise observation of the effect of particle size on the efficiency of the acoustic separation. The experiments used suspensions of 7.5 μm , 1.2 μm , 0.5 μm and 0.15 μm monodisperse silica particles in water. Since the primary acoustic radiation force is strongly dependent on the particle size to the third power of the particle radius, R (Eq. 1) it is expected that the smaller particles will be more difficult to aggregate compared to the larger particles. Moreover, the aggregation of the larger particles is expected to be dominated by acoustic radiation forces, while for smaller particles the streaming force will be dominant. For spherical latex particle suspensions excited

at 1.4 MHz, the transition between radiation force and streaming driven separation was observed for a particle size of about 1.0 μm [27]. A different transition size is expected for silica particles manipulated at the lower resonant frequency used here.

Video 5 is a top-view macroscopic observation for the particle separation process in the case of 1 wt % of 7.5 μm silica particle suspension, and Fig. 6 shows representative snapshots at 15 seconds (Fig. 6a) and 150 seconds (Fig. 6b) after exciting the acoustic standing wave patterns in the cell. A comparison between Fig. 2c for the case of fine dodecane droplets (1 to 3 μm in diameter) and Fig. 6a for the 7.5 μm silica particles shortly after the excitation of the standing wave pattern reveals that the positions of the silica particle aggregates are shifted by approximately $\lambda/4$ from the neighboring positions of the emulsion droplet aggregates. Recall that the pressure nodes and antinodes are separated by $\lambda/4$, confirming that the silica particles, due to their positive acoustic contrast, are driven toward the pressure nodes of the standing wave pattern, while the dodecane oil droplets, due to their negative acoustic contrast, are driven toward the pressure antinode. This observation supports the concept of employing acoustic standing wave patterns to selectively separate oil droplets from solid fine particles in aqueous solutions, which can be of great importance to several industrial application, including oil production and food processing.

Comparison between Figure 6a and 6b (see also Video 5) reveals that some of the neighboring clusters tend to migrate toward each other and the final pattern is of elongated shape clusters that are more sparsely separated along the cell length. The final distance between the clusters positions is about 2.5 mm $\sim \lambda/2$ in the cell's transverse direction, but closer to 5.0 mm, or $\sim \lambda$ in the longitudinal direction. This rearrangement can be attributed to the combined effect of acoustic radiation forces and Rayleigh streaming in a complex 2D or 3D standing wave pattern in the cell as mentioned earlier.

At a higher concentration of ~ 5 wt % of the 7.5 μm silica particles, a more complex dynamic pattern of particle clustering was observed as demonstrated in Video 6 and Fig 6c which shows a

snapshot from this video. In this case, a network of silica particle clusters is formed with the clusters periodically shifting their orientation in a synchronized manner. The characteristic spacing of the cluster structure is about 2.5 mm or $\lambda/2$. A more detailed characterization and explanation of this dynamic clustering process is out of the scope of the present investigation but we believe that this preliminary observation will stimulate future investigation of this intriguing behavior.

Next, we investigate the particle size limit for effective acoustic separation using smaller size silica particles. Top view snapshots of the microfluidic cell 150 seconds after the start of acoustic irradiation for the case of 1 wt % of 1.2 μm , 0.5 μm and 0.15 μm of silica particles are shown in Fig. 7a, Fig. 7b and Fig. 7c, respectively. Compared with the case of 7.5 μm silica particles at 150 seconds (Fig. 6b), it is apparent that the acoustic separation is less effective and takes longer time with the decrease of particle size. In the case of 1.2 μm , while the cluster formation can be clearly observed, there is a lighter shadow around the darker central spots representing particles that are yet to reach the pressure nodes (Fig. 7a). For 0.5 μm , the aggregates pattern starts to barely emerge after 150 second (Fig. 7b), and it takes much longer (5 to 10 minutes) for the formation of particle clusters at the pressure nodes. For the case of 0.15 μm silica particle, no visible signs of cluster formation at the pressure nodes were observed and the color of the particles remained uniformly distributed in the cell (Fig. 7c) even after 15 minutes of continuous irradiation.

Figure 8 shows microscopic high-speed observation for the various silica particles sizes. Figs. 8a and 6b are examples of the 7.5 μm silica particle aggregates observed. Video 7 shows the dynamics of the 7.5 μm silica particle aggregates as they are levitated in the pressure node by the acoustic radiation force. The observed clusters are comprised of well pronounced parallel strings of the 7.5 μm silica particles. We explain the formation of these particle strings with the action of asymmetric secondary acoustic radiation forces. The secondary radiation force between two particles is attractive in the direction of the axis perpendicular to the wave propagation, but repulsive in the direction of the acoustic

wave propagation [1]. Since the standing wave patterns generated in the microfluidic cell used here are of 2D or 3D nature, it is expected that both repulsive and attractive components of the secondary radiation force will act on each particle pairs in the pressure nodes. The alignment of the particle strings in these nodes suggests that the resultant component of the secondary acoustic radiation forces is attractive in the direction of the strings main axes, and repulsive everywhere else. Similar tendency for particulates strings formation was also noticed in the case of larger emulsion droplets (Fig. 3c), but was more clearly observed for the 7.5 μm silica particle case, probably due to the monodispersity of the silica particles samples.

Figs. 8c and 8d are examples of 1.2 μm silica particles clusters. In the case shown in Fig. 8c a distinct string sub-structure of the cluster is observed similar to the case of 7.5 μm particles. However this order could be lost and a vortex type cluster develops as demonstrated in Fig. 8d. This is an indication that for this particle size both radiation and streaming forces are involved in the clustering process. The dynamics of 1.2 μm cluster transition from string structure to vortex is also demonstrated in Video 8. At the end of this video we show clusters which has opposite vortex rotation direction. Respectively, in Fig. 8e shows a vortex structure core observed after about 5 minutes of irradiation in the case of 0.5 μm silica particles, indicating that for that particle size the acoustic clustering is still possible but is now dominated by the streaming acoustic forces. Finally, in Fig. 8f is shown the case of 0.15 μm particle suspensions after 10 minutes of irradiation. In this case we also dispersed a trace amount of 7.5 microns silica particles to mark the position of the pressure nodes. The image does not show any change of color in accordance with the macroscopic observation, indicating that the acoustic irradiation under these experimental conditions is not an effective way to cluster the 0.15 μm silica samples.

4.3 Dodecane droplets - silica particles mixtures

We now investigate the case of a mixture of oil droplets and solid colloidal particles. We used 2 wt % of dodecane droplets mixed with 1 wt % of 7.5 μm or 1.2 μm silica particles in DI water. In both cases, it was possible to achieve acoustic separation of the initially homogeneous mixture of the two into discrete dodecane droplets and silica particle clusters. A macroscopic observation of the acoustic separation of a mixture of fine dodecane droplets and 7.5 μm silica particles is demonstrated in Video 9, and Fig. 9 shows snapshots from this video. After about 150 seconds, one observes the formation of circular-shaped dodecane droplet clusters (dark circular spots) separate from elongated silica particle clusters (light elongated spots) as shown in Fig. 9a. The oil droplet and silica particle clusters continued to grow until ultrasonication is interrupted. Fig. 9b is a snapshot taken 5 seconds after ultrasonication interruption showing the oil droplets clusters formed at the top inner surface of the cell (dark circular spots) and the silica colloids clusters at the inner bottom surface of the cell (lighter elongated spots). The approximate spacing between two consecutive droplets clusters or between two consecutive particle clusters is similar to that between clusters in the case of a single component solution. However, the positions of the clusters in the mixture case is not an exact superposition of the single components cluster positions (Fig. 2c, Fig. 6b), implying a certain degree of interplay between the emulsion droplets and silica particle clusters.

A detailed view of the coexistence and interplay between the dodecane droplets and silica particle clusters during the separation is given by the high-speed video microscopic observations. Fig. 10a is a snapshot from Video 10 for the case of coarse emulsion droplets and 7.5 μm silica particles cluster and Fig. 10b is a snapshot from Video 11 for the case of the fine emulsion droplets and 1.2 μm silica particles. The average distance between the silica particle and dodecane droplets cluster shown in these figures is about 1.1 mm close to, but not exactly equal to the expected $\lambda/4$ spacing between the node and antinodes positions, which is about 1.25 mm. This confirms the macroscopic observation of some degree of interplay between the clusters. Similarly, the emulsion droplet and silica particle clusters have similar sub-structures to the one observed for the single components cases. However, we observed a

more complex dynamical interaction with the particles and droplets circulating are reflected between the clusters, even in the case of larger particulates (Video 10, Fig. 10a) and more pronounced for smaller particulates (Video 11, Fig. 10b) due to the stronger involvement of the vortex forming streaming forces. We repeated the same experiment using the fine-size perfluorohexane emulsion droplets and observed a similar picture for the particle and droplet separation as shown in Fig. 10c for 1.2 μm silica particles in the mixture. This confirms that acoustic separation of solid particles and emulsion droplets can work effectively for emulsion droplets components which are heavier than water, and otherwise could not be separated from the solid component using gravitational stratification.

Finally we performed some proof of concept experiments to demonstrate the ability of acoustic standing wave patterns for separating oil droplets and solid particle in a continuous fluid flow process. In these experiments, a syringe pump was used to control the duration and speed of fluid flow pulses to be applied through the cell. Video 12 is an example of experiments in which fluid flow pulses were initiated during ultrasonication of pure dodecane emulsion. Following 30 seconds of ultrasonication at static flow, a 5 mm/second fluid speed flow is applied for 30 seconds, and then the same cycle is repeated again. As shown in Fig. 11a during the continuous-flow-phase three streams of concentrated emulsion droplets are formed along the pressure antinode positions along the cell and draining out through the cell outlet. Respectively in Fig. 11b we present the case of a mixture of oil droplets and silica particles. Here seven lines formed along the cell length streaming toward the outlet. Three of them are oil droplets streams going through the antinodes positions and four are concentrated silica particles streams passing through the respective pressure-node positions along the cell, as indicated in Fig. 11b. The microfluidic cell used here was not specifically designed for the collection of the concentrated emulsion droplet and silica particles streams at the cell outlet. Nevertheless these preliminary

experiments demonstrate the potential for the operation of microfluidic devices in constant flow mode to separate components from a mixture of oil and solid particulates with different acoustic-contrast.

5. Conclusions

We introduced a simple experimental setup that can be used to systematically study the acoustic separation process of complex particulate mixtures. Such studies are of importance to several industrial and technological applications including crude oil production and processing, algae harvesting for biofuel production, food processing. The experimental setup used a rectangular channel microfluidic cell combined with macroscopic and microscopic visualization by high-speed imaging. We studied the acoustic separation process of a system consisted of aqueous solutions of dodecane oil emulsion droplets and silica colloidal particles. The cell dimensions combined with the a radial mode 300 KHz resonant frequency PZE transducer allowed the generation of 2D or 3D acoustic standing wave patterns inside the cell.

The experiments demonstrated the ability of the acoustic standing waves to simultaneously separate dodecane emulsion droplets from colloidal silica particles in an aqueous mixture of the two. Due to their negative acoustic contrast factor, the oil droplets migrate and accumulate in the pressure antinode planes while the silica particles, due to their positive acoustic contrast factor, accumulate in the pressure node planes of the standing wave patterns. This was demonstrated in mixtures containing 7.5 μm and 1.2 μm silica particles, dodecane oil droplets and heavier perfluorohexane oil droplets. Experiments under continuous flow conditions provided a proof-of-concept that can be used in the development of more sophisticate small and large scale acoustic separation systems.

The use of particulates of opposite acoustic contrast factors also provided a direct visualization means for mapping the distribution of the acoustic field pressure nodes and antinodes in the cell. Experiments in which monodisperse silica particles were the only dispersed phase showed that the separation of 7.5 μm silica particles was driven mostly by acoustic radiation forces, the 1.2 μm particles by both radiation and Rayleigh streaming, and the 0.5 μm particles mostly by vortex-formed streaming.

The larger size silica particles formed parallel strings of particles in the pressure nodes of the acoustic wave pattern. These were attributed to attractive resultant component of the secondary acoustic radiation forces in the direction of the strings' main axes.

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Figure 1. A schematic diagram of the experimental setup.

Figure 2. Snapshots taken from Video 1 showing top-view macroscopic observation for the acoustic separation in the case of fine dodecane droplets in water emulsion: (a) before the start of the ultrasonic irradiation, (b) 40 seconds and (c), 300 seconds after the beginning of the irradiation. The snapshot shown in (d) was taken after 300 seconds of ultrasonication in an independent experimental run.

Figure 3. Microscopic observations high-speed video snapshots from Video 2 showing the cluster formation for the case of coarse dodecane emulsion droplets: (a) before the start of the ultrasonic irradiation, (b) 12 seconds (c), 25 seconds and, (d) 50 seconds after the beginning of the irradiation.

Figure 4. Microscopic observations high-speed video snapshots from Video 3 for a high concentration of fine dodecane droplets: (a) before the start of the ultrasonic irradiation, (b) 12 seconds (c), 50 seconds and (d), 150 seconds after the beginning of the irradiation. (c) and (d) show two different cluster.

Figure 5. Microscopic observations high-speed video snapshots from Video 4 of emulsion droplet cluster (a) deposited on the cell surface at switch-off, or (b) levitated in the pressure antinode upon switching-on of the ultrasound.

Figure 6. High-speed video snapshots from top-view macroscopic view for the separation process in the case of case 1 wt % 7.5 μm silica particles: (a) 15 seconds and (b) 150 seconds after the beginning of the ultrasonication (see also Video 5). (c) One snapshot from Video 6 illustrating the dynamic clustering pattern observed in the case of 5 wt % 7.5 μm silica particles case.

Figure 7. High-speed video snapshots of top-view observations of the microfluidic cell 150 seconds after the beginning of the acoustic irradiation for 1 wt % of (a) $1.2\ \mu\text{m}$ silica particles (b) $0.5\ \mu\text{m}$ silica particles (c) $0.15\ \mu\text{m}$ silica particles suspensions.

Figure 8. Microscopic observation of silica particles clusters: (a) and (b) 7.5 μm silica particles (Video 7). (c) and (d) 1.2 μm silica particles cluster (Video 8), (e) 0.5 μm silica particles cluster during the ultrasonic irradiation. (f) 0.15 μm silica particles suspension, a string of 7.5 μm silica particles marks the pressure node position.

Figure 9. Macroscopic observation of the acoustic separation of a mixture of dodecane droplets and 7.5 μm silica particles mixtures (Video 9). (a) After 150 second irradiation: dark circular spots are the dodecane droplet clusters and the light elongated spots are silica particle clusters. (b) 5 seconds after the irradiation has been stopped: the darker round spots are dodecane droplet clusters deposited on the top side of the cell and the lighter colored spots are silica particle clusters deposited on the bottom side of the cell.

Figure 10. Microscopic observation high-speed video snapshots taken using 10× objective showing separate clustering of silica particle and dodecane droplets during the mixtures irradiation. (a) 7.5 μm silica particle cluster on left and coarse dodecane droplets cluster on right (Video 10). (b) 1.2 μm silica particle cluster on left and fine dodecane droplets cluster on right (Video 11). (c) 1.2 μm silica particle on the left and fine perfluorohexane droplets cluster on the right.

Figure 11. Acoustic separation in a continuous-flow illustrating microfluidic system. (a) Dodecane droplets emulsion. Three streams of droplets passing along the antinode positions during the 5 mm/second fluid flow phase (Video12). (b) Dodecane droplets and 7.5 μm silica particle mixture. The four wider streams of silica particles and three lighter colored streams of dodecane droplets at the node and antinode planes respectively.

Table 1. Density, speed of sound and acoustic contrast factors of the studied medium and particulates.

	ρ [kg/m ³]	c [m/s]	ϕ (in water)
Water	1000	1497	0.00
Dodecane	800	1260	- 0.83
Perfluorohexane	1700	480	- 7.12
Silica	2400	5640	+ 2.20