Directional transport and random motion of particles in ALF ultrasonic cavitation structure

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\begin{abstract}
The motion of particles of different properties and sizes in ALF ultrasonic cavitation structure is investigated experimentally with high-speed photography. Particles tend to transport along the bubble chain and move towards the focus repeatedly and predictably in ALF cavitation structures. Particles at the focus aggregate and separate alternately over time. The separation of particles mainly occurs in the expansion process of cavitation bubbles, while the movement and aggregation of particles mostly take place during the collapse stage. The directional transport of particles along the bubble chain of ALF cavitation cloud and the random aggregation and dispersion at the focus of ALF are all related to the cavitation bubbles attached to the particles. The directional transportation (predictable, repeatable and pipeline-free) and aggregation of particles in ALF cavitation clouds may be used in special occasions, for example, drug delivery and targeted therapy.
\end{abstract}

1. Introduction

There are many phenomena or applications related to mass transport in ultrasonic field. For example, acoustic streaming \cite{1}, ultrasonic pump \cite{2} or piezoelectric pump \cite{3}, vibration induced transport of nano-abrasives \cite{4-6}, motion or manipulation of particles \cite{7,8}, droplets \cite{9,10}, or bubbles \cite{11,12} by acoustic levitation and ultrasonic tweezers. When the ultrasonic amplitude exceeds a certain threshold, cavitation will occur in the ultrasonic field. There are some phenomena or applications of mass transport which are related to cavitation bubble vibration. For example, microjet \cite{13}, microstreaming \cite{14}, the acceleration of solid particles subjected to cavitation nucleation \cite{15-17}, collapsing bubble-induced micropump in microfluidic devices \cite{18-20} and Sonoporation (transient enhancement of the cell membrane permeability induced by acoustic bubble) \cite{21-23}. In the ultrasonic field, the cavitation bubble not only vibrates radially, but also moves. It is generally known that cavitation bubble distribution is spatially inhomogeneous in the ultrasound field. These bubbles can form different structures during their motion \cite{24-26}. Acoustic Lichtenberg Figure (ALF) is a common cavitation structure far away from the radiant surface. Bubbles in the ALF structures move from outer regions (nodes of standing waves) towards the center (antinodes of standing waves) and finally reach a central point (focus) of high bubble concentration. The whole structure resembles roots of a plant, neurons, river system or electrical discharge patterns. Iskander S. Akhatov \cite{27} (1996), Ulrich Parlitz \cite{28} (1999), Robert Mettin \cite{29} (1999) and Zhang \cite{30} (2013) investigated the dynamics of ALF structure in acoustic cavitation fields. Most of the researches on the ALF cavitation structures are about the deformation, motion and distribution of cavitation bubbles. To our knowledge, there is no investigation of motion of solid particles in ALF structures. Unlike the material transport mentioned above, the transport of particles in the cavitation cloud is greatly influenced by the ultrasonic field, the cavitation bubbles and the cavitation structure simultaneously. In this paper, the directional transport and random motion of particles in ALF ultrasonic cavitation structures was investigated experimentally.

2. Experiment

Fig. 1 illustrates the experimental setup when the directional transport and random motion of particles in an ALF ultrasonic cavitation structure is recorded. The experimental setup consisted of the ultrasonic cavitation devices, the high-speed imaging and illumination system, particle throwing device, etc. The piezoceramic sandwich transducer is well enveloped and can be submerged in water completely. The
Ultrasonic horn is mounted horizontally in a transparent chamber (600 mm × 330 mm × 330 mm). Tap water (exposed to air for greater than 24 h) is used in the experiment. The ultrasound is produced by an ultrasonic processor (Jiuzhou Ultrasonic Technology Co., Ltd. China) with a frequency of 18.5 kHz (radiating surface diameter, \( d = 50 \) mm), and a maximum input electric power of 100 W. The radiating surface diameter of the sandwich piezoelectric ceramic ultrasonic transducer equals to the diameter of piezoelectric plate. Cavitation structure is recorded with a high-speed camera (Photron Fastcam SA-1, Photron Ltd., Japan) equipped with two long distance microscopes (Zoom 6000, Navitar, USA; LM50JCM, Kowa, Japan). The frames are illuminated with a high brightness Cree XHP70 LED and PI-LUMINOR high-light LED lamp.

3. Results and discussion

One of the most important characteristics of ALF cavitation structure is the existence of cavitation bubble chains. A large number of cavitation bubbles are arranged into a bubble chain structure in the process of radial vibration and translational motion. Bubbles line up and move along the bubble chains. These chains interweave with each other and assemble to form a branch structure (ALF). This is mainly due to the interaction of the standing wave sound field, the primary Bjerknes force and the secondary Bjerknes force. The ALF structure can be considered as a gas transport structure, although the gas content of cavitation bubbles is very small [28]. When a solid particle is put into the ALF cavitation structure (gas–liquid two-phase medium), it is found that the solid particle moved translationally (as shown in Fig. 2). The trajectory of the particle is almost coincident with that of cavitation bubbles. It can be seen that there is an abnormal change in the direction of motion at a point where the cavitation bubble chains converge. Without the ALF cavitation structure, the trajectory of the particle is inconceivable.

A relatively large bubble chain is used in the experiment to test particles of different sizes and physical properties (as shown in Table 1). The cavitation bubbles in the bubble chain move from left to right (as shown in Fig. 3A). Directional transportation along the bubble chain occurs in particles such as glass, NaCl, diamond sand, even a silicone particle whose size is much larger than the cavitation bubble (as shown in Fig. 3B,C,D,E). The transportation is directional, predictable and repeatable. The destination of this transportation, which is the antinode of the standing wave, namely the high amplitude region of acoustic pressure, is also the end point of cavitation bubble movement and the place where bubbles finally merge with the others.

Glass particles aggregate at the antinodes after being transported by the bubble chains (as shown in Fig. 4). Photos of particles with different acoustic phases after 20 acoustic periods can be taken by adjusting shooting frequency. The upper part of Fig. 4 is photographs of particles during bubble collapse, the frames are clean and have hardly any visible cavitation bubbles. The lower part is photographs of particles when cavitation bubbles expand to near their maximum volume. It can be seen that cavitation bubbles move towards the aggregation of particles, forming a ALF structure. The particle cluster is located in the focus of ALF cavitation cloud.

To study the motion of particles, we intercepted and enlarged the image of the glass particle aggregation at the focus (as shown in Fig. 5A). In order to filter out the interference of cavitation bubbles to the image

| Particle | Diameter/Length (mm) | Density (g/cm³) | Release method |
|----------|----------------------|----------------|---------------|
| Wax      | 0.2–1.5              | 0.9            | The particles are grasp with a small tweezers and released near the ALF cavitation structure. |
| Glass    | 0.15–0.8             | 2.5            | The particles are released on the liquid surface above the ALF cavitation structure through a small funnel |
| Salt     | 0.1–0.5              | 2.2            | The particles are released on the liquid surface above the ALF cavitation structure through a small funnel |
| Sand     | 0.02–0.04            | 2.7            | The particles are released on the liquid surface above the ALF cavitation structure through a small funnel |
| Silicone | 1.0–3.0              | 1.1            | The particles are grasp with a small tweezers and released near the ALF cavitation structure. |
| Fibre    | 0.3–2.0              | 1.4            | The particles are grasp with a small tweezers and released near the ALF cavitation structure. |

Fig. 1. Experimental setup to visualize the motion of particles and bubbles in an ALF cavitation structure.

Fig. 2. The transportation of a wax particle in an ALF structure. (A) Snapshots from a video of this experiment. (B) A composite image of superimposing the snapshots from the video sequence.
(the cavitation bubbles appear as black spots under the illumination of transmitted light), the video snapshot of cavitation bubble collapse is selected (as shown in Fig. 5B). It can be found in the experiments that particles are not fixed at the focus, they aggregate and separate alternately over time, and their relative position changes at any time.

Nearly 500 images taken in about 0.5 s in the experiment were analyzed in Fig. 5. The diameter of the smallest circle capable of enclosing all particles is defined as the equivalent diameter (as shown the blue dotted circle in Fig. 6). The equivalent diameter of particle aggregation in each image was measured. It can be found that the diameter of the particle cluster does not appear to have an obvious periodicity with time, the aggregation and dispersion of particles seem to be more like a random event (as shown in Fig. 6). However, careful observation of the diameter change curve reveals that the curve is flatter at the peak and sharper at the trough. The residence time of particles

Fig. 3. The transportation of various solid particles.

Fig. 4. Particle cluster aggregation in an ALF structure.

Fig. 5. The motion of particle cluster at the focus. The supplementary video can be accessed with the online version of the article as Video 1.

Fig. 6. The change of particle cluster diameter with time at the focus.
shown the subfigure (as shown the subfigure (τ = 0 ms) and the subfigure (τ = 6.58 ms) in Fig. 7 (A)). It can be seen that when τ = 0.08–1.08 ms, both particles are adhered with cavitation bubbles, and the bubbles are on the side towards the opposite particle (the velocity at which the two particles approach each other is 0.2 m/s). After that, the two particles get closer at a greater speed (0.8 m/s), cavitation bubbles are merged and the two particles (almost) contact (we believe the cavitation nuclei after collapsing are still situated in the middle of the two particles). The two particles did not bounce off immediately. They change their attitude randomly until the cavitation bubble and the two particles are in the right position (as shown in Fig. 7(B)). Then the two particles separated rapidly under the action of a certain force. The separation speed of two particles (1.7 m/s) is much higher than that of two particles approaching each other (0.8 m/s), which explains the behavior of particle cluster at the focus in Fig. 6. It can be found that the particles rotate violently during the separation process, which indicates that this force is not a mass force, and its point of action is not the center of gravity. Photographs with a larger visual field at τ = 0 ms and τ = 6.58 ms show that the velocity of the particle decreases rapidly with the collapse of the cavitation bubble, and shock wave and microjet occur at this time. Obviously, the shock wave and micro jet acting on the two particles do not separate the particles from each other. When the cavitation bubble rebound, the particle velocity rises again. The size of the cavitation bubble remains basically the same as the distance between particles increases. However, the moving speed of the particle is significantly reduced as the distance between particles increases, which is due to the weakening of the cavitation bubble interaction. This shows that the main mechanism of particles separation is particles bounce off each other because of cavitation expansion (the rapid expansion of the cavitation bubble volume produces an outward force), independent of shock wave or microjet induced by the cavitation collapse.

In order to study the role of cavitation bubbles in the process of particle dispersion, a higher shooting frequency and a larger magnification were used in the experiment of particle separation (as shown in Fig. 8). When τ = 0 μs, the two glass particles almost contact, but we think that there should be cavitation nuclei between them. When τ = 48 μs, the cavitation nuclei expands to form a cavitation bubble. Distance between particles increases with the expansion of the bubble. The cavitation bubble then grow and collapse for several times, meanwhile the particle distance becomes larger.

The diameter of the cavitation bubble and the moving speed of the particle on the right side in the experiment in Fig. 8(A) were measured. It turns out that the diameter of the bubble and the moving speed of the particle are periodic with time and moving distance, and there is a good correlation between them (as shown in Fig. 8(B)), the bubble diameter is the average of the maximum (triangles) and minimum (squares) values of multiple measurements.). In the bubble expansion stage, the particle velocity reaches the maximum value, and the translational motion of the particle mainly occurs in the bubble expansion stage. On the contrary, the velocity of the particle decreases rapidly with the collapse of the cavitation bubble, and shock wave and micro jet occur at this time. Obviously, the shock wave and micro jet acting on the two particles do not separate the particles from each other. When the cavitation bubble rebound, the particle velocity rises again. The size of the cavitation bubble remains basically the same as the distance between particles increases. However, the moving speed of the particle is significantly reduced as the distance between particles increases, which is due to the weakening of the cavitation bubble interaction. This shows that the main mechanism of particles separation is particles bounce off each other because of cavitation expansion (the rapid expansion of the cavitation bubble volume produces an outward force), independent of shock wave or microjet induced by the cavitation collapse.

In order to explore the role and mechanical mechanism of cavitation bubbles in the process of particle aggregation, some experiments on fibers (their mass is very small, so the velocity and acceleration change more obviously) in a ALF structure were performed (as shown in Fig. 9A). A small segment of fiber maintains uniform motion in water from τ = 0 ms to 3 ms. At τ = 4 ms, a bubble suddenly attaches to the middle of the fiber. The velocity and acceleration of the fiber increases (the acceleration increases from 0 m/s² to 48.8 m/s² and then to 222.2 m/s² until the fiber aggregates with a fiber cluster (as shown in Fig. 9C), the error is about 0.02 m/s). If the resistance of water is not taken into account, the force on the fiber is increased by more than four times with the decrease of distance. If the resistance of water is considered (the resistance increases rapidly with the increase of speed), the force on the fiber should be greater. Because the acceleration process of the fiber occurred just after the appearance of bubble adhesion, and the force is sensitive to distance, which is consistent with the secondary Bjerknes force, so it’s probably the secondary Bjerknes force between the
Fig. 8. Particles bounce off each other due to cavitation bubbles. (A) Snapshots from a video of the experiment. (B) Variation of bubble diameter and particle velocity with position. The supplementary video can be accessed with the online version of the article as Video 3.

Fig. 9. The motion of fibres in an ALF structure. (A) Snapshots from a video of the experiment. (B) The force and deformation of fibre in motion. (C) Variation of fibre velocity with time. The supplementary video can be accessed with the online version of the article as Video 4.

Fig. 10. The movement of a grain of diamond sand in ALF structure. (A) Snapshots from a video of the experiment. (B) Variation of velocity and distance of the particle with time. (C) A composite image of superimposing the snapshots from the translation line in subfigure A. The supplementary video can be accessed with the online version of the article as Video 5.
cavitation bubble on the fiber and cavitation bubbles in the cluster (cavitation bubbles in the fiber cluster are not shown in the picture) that plays a major role in the acceleration process. Owing to the resistance of water, the fiber deforms obviously during the acceleration process, which is shown in a more intuitive way (as shown in Fig. 9B).

To further explore the role and mechanical mechanism of cavitation bubble in particle aggregation, the diamond sand particle (its mass is not negligible but the size is much smaller than cavitation bubble) was used in the experiment (as shown in Fig. 10). Fig. 10A shows the motion of a grain of diamond sand and an attached bubble in 7 acoustic periods. The position of the particle and the attached cavitation bubble barely changes during the expansion stage. A significant motion of the particle occurs during the collapse stage (the image is blurry because of the rapid movement of cavitation bubble wall during the collapse), while the inertial movement after that is very small. This is quiet different from the situation when particles are separated, in which motion mainly occurs during the expansion stage. After 7 periods, the particle eventually converge with a particle cluster and its attached bubble. Fig. 10B depicts the variation of particle velocity and moving distance with time. The velocity of the particle presents a periodic change, it increases dramatically to nearly 4 m/s and then quickly declines to zero. Such change is synchronous with the acoustic period. This kind of intermittent movement of the particle makes the moving distance curves look like steps. Fig. 10C is a composite image of 7 pictures in the translation line in Fig. 10A, which shows the real trajectory of particle movement. It can be found that the trajectory of particle movement has an evident deflection when it is close to the particle cluster. Therefore, before the moving trajectory is changed, the main force that may drive the particle and attached bubbles is the primary Bjerknes force. The secondary Bjerknes force only works when the two particles with cavitation bubbles attached are relatively close. It also explains the behavior of particle aggregation and dispersion in Fig. 6. When particles disperse, the primary Bjerknes force drives the particles to slowly decelerate outward and slowly aggregate inward, so that the curve is smooth during dispersion stage (∩); when the distance between particles is relatively short, particles accelerate to aggregate. When particles contact each other, they bounce off rapidly under the action of cavitation bubbles, the curve is sharp during the aggregation (∪).

In fact, the aggregation process of particles with cavitation bubbles attached also takes on the characteristics of ALF structure obviously. Fig. 11 shows the process and trajectory of the aggregation of multiple particles. The orange part is the movement and aggregation of particles that will happen after the picture was taken. The blue part represents the movement and aggregation of particles that happened before the picture was taken. It can be seen that the moving and merging trajectory of particles have the characteristics of ALF structure. The moving direction of cavitation bubbles tend to be from the bottom left to the top right, which is controlled by the primary Bjerknes force, while the merging of cavitation bubbles is mostly affected by the secondary Bjerknes force. Due to the randomness of the position of particles, the merging also has some randomness. Accordingly, before particles enter an obvious ALF cavitation structure, the trajectory of particle aggregation also shows certain randomness. However, as particles and attached cavitation bubbles are in the vicinity of the antinode of a standing wave, the randomness of the spatial distribution of cavitation bubbles decreases, the branch structure of the cavitation cloud becomes clearer. Randomness of this aggregation of particles and attached cavitation bubbles also decreases. The motion of particles is more directional, controllable and predictable. But even in a very clear ALF structure, occasional randomness of particle movement still exists. For example, two particles in short-distance bounce off each other under the action of cavitation bubbles (as shown in Figs. 7 and 8), the particles deviate from the bubble chain, or even go backwards driven by cavitation bubbles (as shown in Fig. 12). The bubbles attached to the particle are subjected to the same force towards the focus as other bubbles in ALF cavitation structure, which determines the attitude of the particles. The attached bubbles face the focus and the particle deviate from the focus. The growth and collapse of the attached bubbles push the particles forward like a propeller, which would be an interesting field for future work.); particles break away from the constraint of bubble chains or the focus and then

Fig. 11. The trajectory of particle convergence.
rise or sink due to the gravity or buoyancy, etc. For the macro translational motion of particles on the scale of cavitation cloud, more attention is paid to the relationship between the trajectories of particles and the trajectories of cavitation bubbles (bubble chain), as well as the long-term routine behavior of particles. For the micro motion of particle interaction, particles present short-term disorder behavior, and more attention is paid to the role of cavitation bubble in particle movement. There is no contradiction between micro disorder and macro order.

4. Conclusion

The directional transport and random motion of particles in ALF ultrasonic cavitation structure was investigated experimentally. Particles with different physical properties and sizes may be transported along the bubble chain repeatedly and predictably in ALF cavitation structures. Particles tend to move towards the focus. Particles are not propelled particle accelerator, Phys. Rev. Lett. 92 (17) (2004), https://doi.org/10.1103/PhysRevLett.92.174501.
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CRediT authorship contribution statement

Ma Yuhang: Investigation, Visualization, Writing - original draft.
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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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