RELATIONSHIP AMONG SOUND FIELD, BUBBLE BEHAVIOR AND SONOLUMINESCENCE

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Abstract

To evaluate efficiency of sonochemical reactors, intensity of sonoluminescence was measured under various conditions. Quenching phenomenon of sonoluminescence was observed at excessive sound pressure, which suggests that high power ultrasound does not necessarily attain high efficiency in sonochemistry. Visualization of sound fields showed no differences in the patterns of sound pressure distributions at different power levels. On the other hand, behavior of cavitation bubbles changed very much at high sound pressure levels showing formation of bubble clusters. It was also found that the clustering was affected by fluid motion such as stirring operation and circulating flow in the sound field. Hence, fluid motion changes the intensity of sonoluminescence and efficiency of sonochemical reactions.

Introduction

Two kinds of apparatus are used in sonochemistry; one is an ultrasonic horn type and the other one is an ultrasonic cleaner type (Fig.1). Although both apparatuses utilize action at the collapse of cavitation bubbles, their sound fields are very different and their sonochemical reaction fields are different, accordingly [1]. When an ultrasonic horn is immersed in a liquid, fast motion of cavitation bubbles is observed at the tip of the horn. The motion of bubbles attains 2 m/s near the tip but the speed decreases rapidly apart from the tip, because the ultrasound diffuses spherically and its amplitude decreases inversely proportional to the distance from the tip. If the horn is immersed in a luminol solution, we can see sonochemical luminescence but the luminescing region is limited to a very small region near the tip [1].

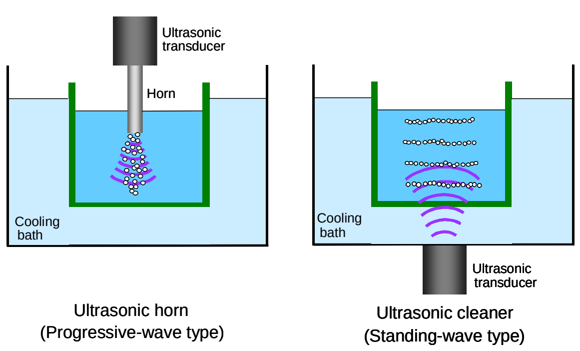

Figure 1: Typical apparatus used in sonochemistry.

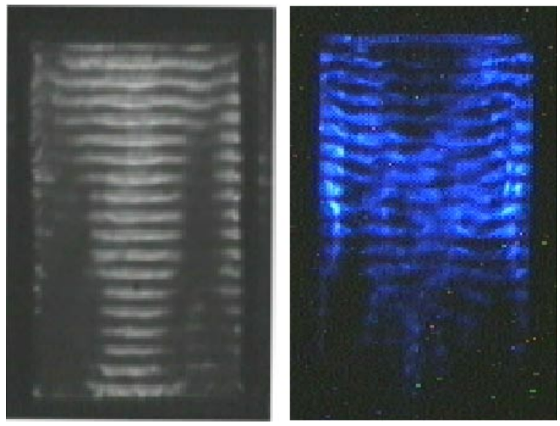

Figure 2: Sound field visualized by the schlieren method (left) and observed sonoluminescence (right) in a rectangular glass cell driven at 140-kHz ultrasound.

On the other hand, the apparatus of ultrasonic cleaner type generates a standing-wave field between the ultrasonic transducer and liquid surface. Figure 2 shows the sound field visualized by the schlieren method (left) and sonochemical luminescing field (right) in a rectangular glass cell driven at 140-kHz ultrasound [2]. A clear standing-wave field with layered structure is observed and the luminescing region corresponds to the standing-wave field very well. Since cavitation bubbles generated by intense ultrasound are exerted by acoustic radiation pressure or Bjerknes force, tiny bubbles go to antinodes of sound pressure distribution in a standing-wave field and they emit light there due to abrupt quasi-adiabatic compression and resulting high temperature.

To make a sonochemical reactor effective, larger luminescing region is preferable and higher sound pressure may be applied to this end. The present paper discusses the change of sonoluminescence at excessive sound pressure in relation to the sound field and bubble behavior in the field.

Quenching of sonoluminescence

Sonoluminescence from distilled water filled in a rectangular glass cell was measured using a photomultiplier tube (PMT) as shown in Fig.3 [3]. The reactor was a standing-wave type driven at 132.2 kHz. The bubble motion was recorded with a still camera and a high-speed video camera.

As shown in Fig.4, input power to the ultrasonic transducer increased in a quadratic function of the function generator (FG) output or input to the power amplifier. But multibubble sonoluminescence (MBSL) intensity decreased suddenly beyond a certain level of FG output; namely, quenching of sonoluminescence occurred [3].

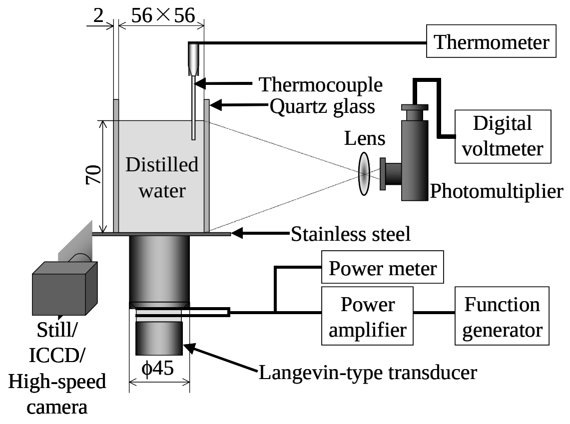

Figure 3: Experimental setup to measure changes in sonoluminescence intensity.

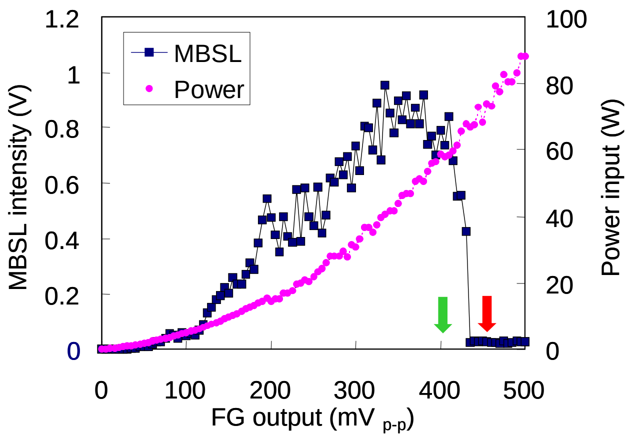

Figure 4: Changes of input power to the ultrasonic transducer and the corresponding intensity of multi-bubble sonoluminescence (MBSL) at 132.2 kHz.

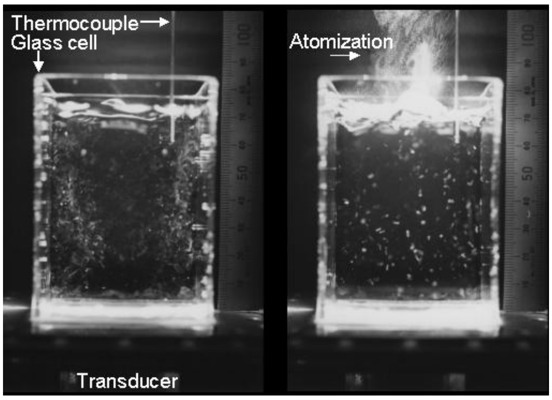

Figure 5: Changes of behavior of cavitation bubbles before (left, 400 mV) and after (right, 450 mV) quenching of sonoluminescence at 132.2 kHz.

Formation of bubble clusters

Figure 5 shows the changes of bubble behavior before (left, 400 mV) and after (right, 450 mV) quenching occurred [3]. Before quenching, we observed foggy bubble cloud. But after quenching, cavitation noise changed apparently and small clusters of bubbles were observed. These observations suggest that, once bubbles form clusters, sonoluminescence intensity decreases. Or one may say that excessive sound pressure induces clustering of cavitation bubbles and diminishes sonochemical efficiency.

Formation of bubble clusters is due to the secondary Bjerknes force acting between oscillating bubbles (Fig.6). If bubbles oscillate in phase, they are attracted each other to form a cluster. Once these clusters are formed, they become large to be expelled from pressure antinodes to nodes, and ultrasound cannot reach inside the cluster because of impedance shielding at the outer surface of the cluster [4]. Although the secondary Bjerknes force is weak, which is almost inversely proportional to the second power of distance between bubbles and the forth power of ultrasonic frequency, it becomes apparent beyond a certain amplitude level to form clusters and diminishes sonoluminescence.

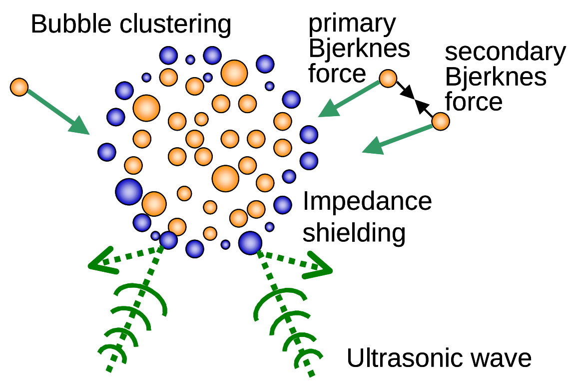

Figure 6: Clustering of bubbles due to the secondary Bjerknes force in a sound field.

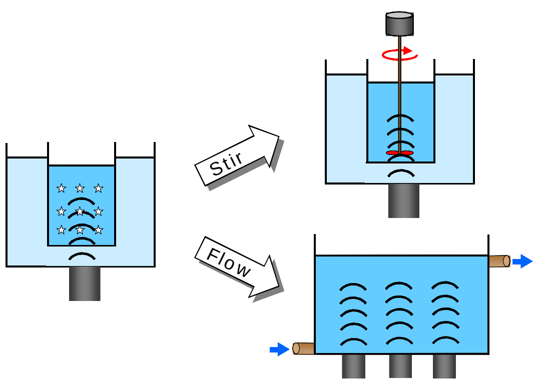

Figure 7: Probable fluid motion in a sound field of an actual sonochemical reactor.

Influence of fluid motion

In practical applications of sonochemistry, fluid motion such as stirring operation or flow of sample liquid in a cell is probable (Fig.7). Although the time scale of fluid motion is much slower than that of bubble collapse and luminescence phenomenon, fluid motion will disturb formation of bubble clusters because the secondary Bjerknes force is weak [5].

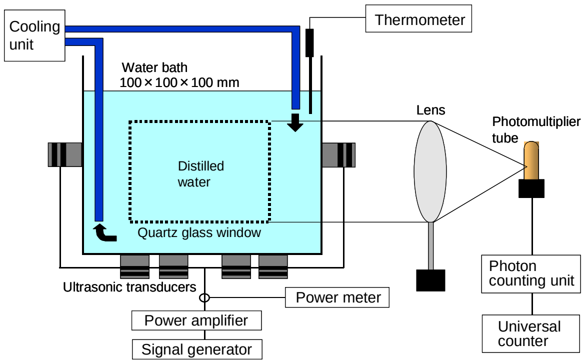

Figure 8: Experimental setup to study influence of fluid motion on sonoluminescence intensity.

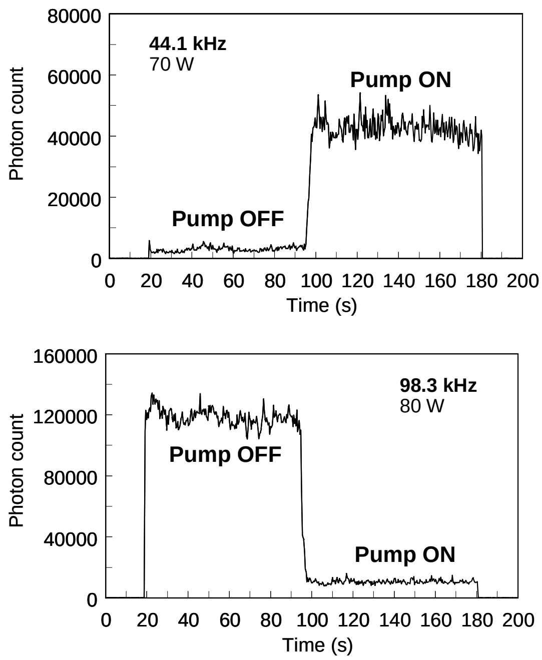

Figure 9: Changes of sonoluminescence intensity with and without fluid motion generated by the pump of cooling unit.

Using an apparatus shown in Fig.8, influence of fluid motion on sonoluminescence from distilled water was studied at various sound pressure levels and ultrasonic frequency. The sonoluminescence intensity was measured with a photomultiplier tube and a photon counter. Fluid motion was generated using a pump of cooling unit to keep the water temperature constant and was about 4 L/min into a 1 L reactor cell. The results are shown in Fig.9. The upper part shows the case at 44.1 kHz and 70 W in input power to the ultrasonic transducers. The sonoluminescence is intense when the pump is on or fluid motion exists. On the contrary, sonoluminescence intensity is low with fluid motion at 98.3 kHz and 80 W as shown in the lower part.

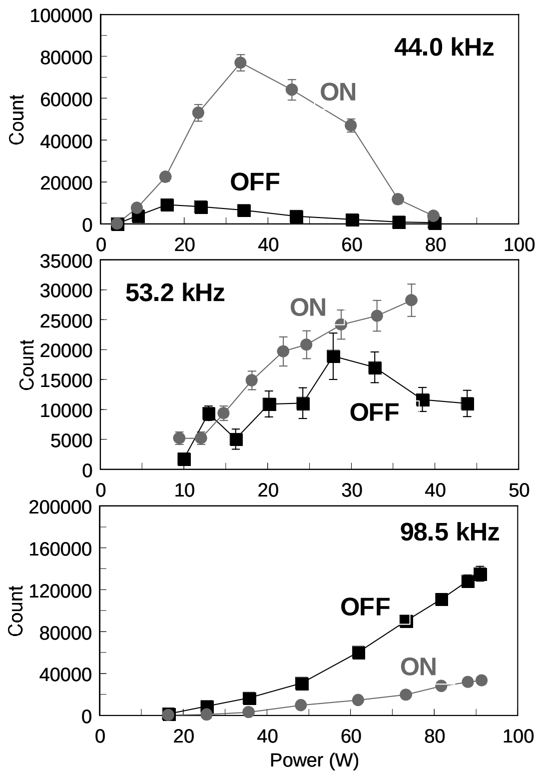

Figure 10: Amplitude and frequency dependence of sonoluminescence intensity with and without fluid motion.

This confusing phenomenon becomes clearer in Fig.10, in which amplitude dependence at different frequency is shown comparing the cases with and without fluid motion. At lower frequency of 44.0 kHz, fluid motion (pump on) enhances sonoluminescence intensity. Quenching phenomena at excessive sound pressure (or excessive input power) are seen for both cases of pump on and pump off. At medium frequency of 53.2 kHz, the difference between pump on and off becomes less. At higher frequency of 98.5 kHz, the relation is inverted; namely, the case of pump off gives higher sonoluminescence than that of pump on.

Bubble behavior taken with a still camera at lower frequency of 23 kHz is shown in Fig.11 comparing the cases of pump on and off [3]. The center of each photograph corresponds to the pressure antinode of the standing wave field in the reactor cell. In the case of pump off, the bubbles are expelled from the pressure antinode. When the pump is on or there exists fluid motion, the bubbles gather around the antinode. The difference in bubble behavior becomes clear in Fig.12, which was taken with a high-speed video camera [3]. If there is no fluid motion (upper), tiny cavitation bubbles form clusters and they are expelled from the pressure antinode because they are too larger to go to antinode. But if there exists fluid motion (lower), the bubble clusters are scattered into tiny bubbles and they go to pressure antinode to emit sonoluminescence. This is the explanation why fluid motion enhances sonoluminescence intensity at low frequency.

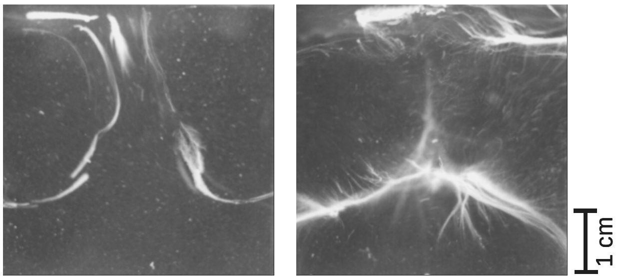

Figure 11: Changes of bubble behavior without (left) and with (right) fluid motion at 23 kHz. The center of each photograph corresponds to the antinode of sound pressure distribution.

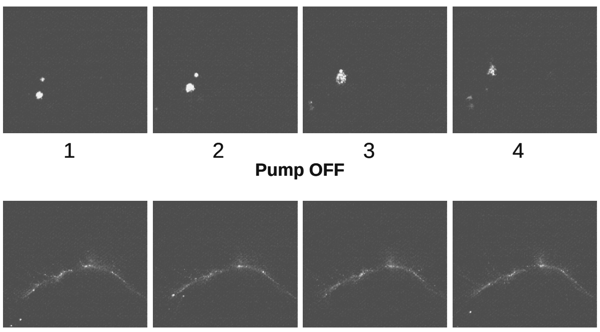

Figure 12: Behavior of cavitation bubbles taken with a high-speed video camera at 200 frames per second. Upper: without fluid motion, lower: with fluid motion, corresponding to Fig.11.

The situation is different at higher frequency. Although too large bubbles are expelled from pressure antinodes, a bubble should be large enough to some extent to attain high compression ratio at the collapse phase of the bubble oscillation. Otherwise, the temperature in the bubble is not sufficiently high to emit sonoluminescence. If sound pressure amplitude is the same as in the case of lower frequency, the bubble cannot grow large enough at higher frequency. Then clustering of bubbles helps to make larger bubbles. But the secondary Bjerknes force to form bubble clusters decreases almost inversely proportional to the forth power of frequency. So, the clustering phenomenon works in a favorable manner for sonoluminescence without making too large bubbles at higher frequencies. Hence, the fluid motion prevents growth of bubbles and brings less sonoluminescence.

Conclusions

Ultrasonic cleaner type reactors have wider region for sonochemical reactions than ultrasonic horn type reactors. Intense ultrasound does not necessarily mean higher efficiency in sonochemistry. The relation between ultrasonic intensity and sonoluminescence intensity was discussed showing occurrence of quenching phenomenon in sonoluminescence at excessive sound pressure. Frequency dependence was observed in this phenomenon. High-speed video images of bubble behavior showed that this quenching phenomenon was intimately connected with growth and clustering of cavitation bubbles. The frequency dependence in the quenching phenomenon can be explained by the effect of the secondary Bjerknes force, which forms clusters of bubbles. Large bubbles and bubble clusters are expelled from antinodes to nodes of sound pressure distribution in the standing-wave field. It is also shown that fluid motion under ultrasonic irradiation affect the intensity of sonoluminescence. At low frequency or high intensity ultrasound, fluid motion enhances sonoluminescence, because it prevents bubbles from forming clusters. Otherwise bubbles will grow too large to be effective in the sound field. At high frequency or moderate intensity ultrasound, the bubbles are too small to collapse violently. Then, the clustering phenomenon promotes growth of bubbles, and fluid motion is not favorable for sonoluminescence.

Acknowledgments

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