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Relation between thresholds of free-radical generation and atomization under ultrasound exposure

超音波照射下におけるフリーラジカルと霧化の発生閾値の関連

Takeshi Aikawa[1†] and Nobuki Kudo[2] ([1]Graduate School of Information Science and Technology, Hokkaido Univ.; [2]Faculty of Information Science and Technology, Hokkaido Univ.)

相川　武司 [1†]，工藤　信樹 [2]　([1] 北海道大学　大学院情報科学院, [2] 北海道大学　大学院情報科学研究院)

1. Introduction

Atomization of liquids is one of characteristic phenomena induced at the liquid surface irradiated by a submerged ultrasound transducer, and the phenomena is widely used for home humidifiers. It is considered that atomization play an important role also in ultrasound therapies that utilize high intensity focused ultrasound. There are three theories on the mechanism of atomization: first is the theory based on cavitation phenomena[1)], the second is the theory based on surface acoustic waves called capillary waves[2)], and the last is combination of the first and second theories[3)]. However, still there is no conclusive evidence which mechanism is responsible for atomization.

In this study, free radicals generated by ultrasonically induced cavitation were quantified by iodine-starch test, and relations with a shape of protuberance generated at the water surface and with initiation of atomization were investigated.

2. Methods and Materials

Figure 1(a) shows an ultrasound atomization system used in this study, and Fig. 1(b) shows details of a water bath used to observe atomization. The atomizer (Honda Electronics, HMC-2400) uses a disk-shaped PZT transducer of 20 mm in diameter to generate continuous-wave ultrasound of 2.4 MHz. Ultrasound pressure was controlled in the range of 0.135−0.161 $MPa_{p\text{-}p}$ by changing a voltage amplitude to drive the transducer. Ultrasound pressure measurement was carried out using a needle-type hydrophone (Precision Acoustics, PAL HPM05-S) inside a sufficiently large water bath (not shown). The hydrophone was placed at the distance of about 25 mm away from the surface center of the transducer.

The transducer was installed at the bottom of the water bath with facing up, and the bath was then filled with a 12-mL iodine-starch solution (0.01 M KI, 0.1 M CCl_3CHO, 1 M NaCl and 0.3 g/L soluble starch)[4)]. The distance from the transducer surface to water surface was 25 mm in this condition. After 2-hour ultrasound exposure, free radical generation was quantified by evaluating absorbance of the solution. A iodine-starch solution of 300-μL in volume was evaluated using a microplate reader (Thermo Fisher Scientific, Multiskan FC) at a wavelength of 570 nm.

3. Results and Discussion

Figures 2(a)−2(c) show changes of a water surface shape with the increasing pressure of atomizer output. At a condition of 0.135 $MPa_{p\text{-}p}$, a protuberance created by an acoustic radiation force was already observed at the water surface above the transducer (Fig. 2(a)). Height and area of the protuberance were increased at the condition of 0.155 $MPa_{p\text{-}p}$ (Fig. 2(b)). At the condition of further output increase, a shape of the protuberance became unstable, and the protuberance showed a cyclic change in its height. At the condition of 0.161 $MPa_{p\text{-}p}$, the protuberance changed its shape to a fountain shape, and a large droplet of about 2 mm in diameter was occasionally pinched off from the

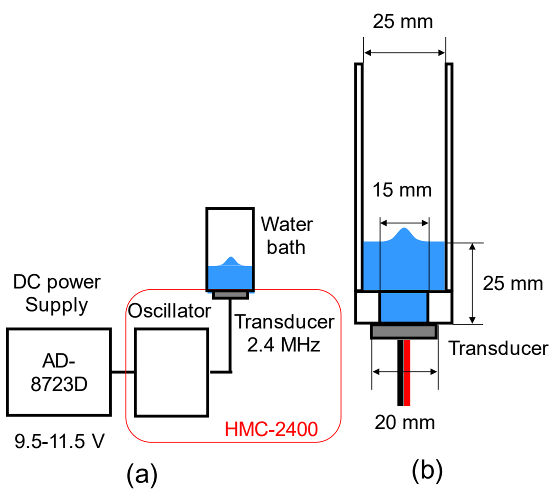

Fig. 1. An ultrasonic atomization system (a) entire system. (b) Details of a water bath used to observe atomization.

of the fountain top (Fig. 2(c)). At the same time, atomization was confirmed by naked-eye observation, suggesting that the fountain shape is essential to produce the capillary waves.

Figure 3 shows absorbance of the iodine-starch solution irradiated by ultrasound. Measurements were made twice at each acoustic pressure level, and the average was plotted in the graph. Absorbance of the solution without ultrasound irradiation was 0.041 in arbitrary unit. In the pressure range from 0.135 $MPa_{p\text{-}p}$ to 0.155 $MPa_{p\text{-}p}$, the absorbance was almost the same, and Mean (SD) of absorbance in this pressure range was 0.046 (0.005), and the mean was almost equal to that of the solution without ultrasound irradiation. In contrast, the absorbance at the acoustic pressure of 0.161 $MPa_{p\text{-}p}$ ($\simeq$1.7 W/cm^2) showed a sharp increase up to 0.41, indicating the presence of a cavitation threshold around this pressure.

Three phenomena observed were in this study, i.e. fountain formation, atomization, and sharp increase in absorbance had the same threshold pressure, suggesting the importance of inertial cavitation in atomization. Yasuda et al. also confirmed initiation of atomization under exposure to 2.4-MHz ultrasound of about 1 W/cm^2, and suggested the importance of non-cavitational mechanisms[5)], because the cavitation threshold of air-saturated water in 2.4 MHz is 1,700 W/cm^2 [6)]. In our experiment, atomization was initiated at 1.7 W/cm^2, and generation of inertial cavitation was confirmed by the sharp increase in absorbance, indicating that cavitation plays an important role in atomization. Focusing of ultrasound reflected by the protuberance[7)] can be one of the possible mechanisms to initiate cavitation at intensities 1,000 times lower than the typical threshold.

4. Conclusion

Free radical generation and atomization under ultrasound irradiation were investigated to elucidate the mechanisms of ultrasound atomization. Good agreement was found in the pressure thresholds for cavitation and atomization, suggesting that cavitation phenomena play an important role in the ultrasound atomization.

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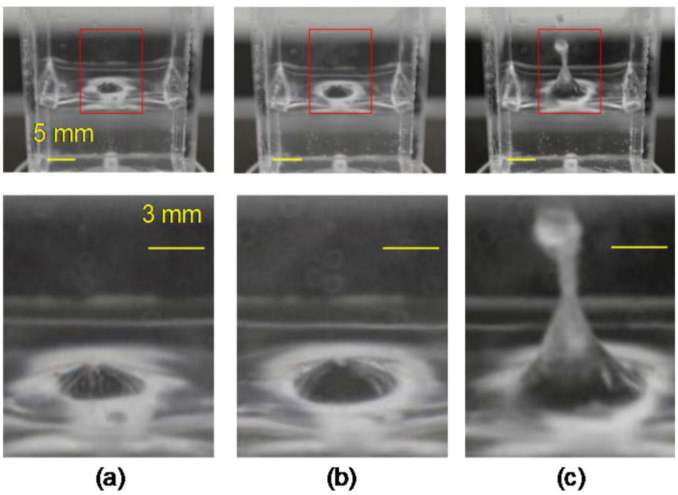

Fig. 2. Changes of a water surface shape with the increasing pressure of atomizer output. (a) 0.135 $MPa_{p\text{-}p}$, (b) 0.155 $MPa_{p\text{-}p}$, and (c) 0.161 $MPa_{p\text{-}p}$/

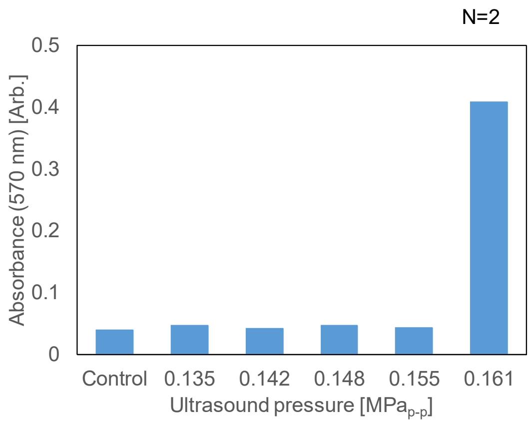

Fig. 3. Absorbance of an iodine-starch solution irradiated by ultrasound.