Development and Optimization of a Microbubble Generator with a Hollow Cylindrical Ultrasonic Horn

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Abstract
The microbubble generator with a hollow ultrasonic horn is a recent development that can easily generate large quantities of microbubbles with a diameter of less than 100 μm. However, it still presents some drawbacks such as not being able to produce a sufficiently large gas flow rate for industrial applications, whereas the ultrasonic oscillation consumes large amounts of power. In this study, we developed a microbubble generator with an interchangeable orifice, and we evaluated the effect of the orifice shape on optimizing the microbubble generation through the time evolution of dissolved oxygen in oxygen-bubbled water, as well as the distribution of the bubble diameters. We also evaluated whether the generator has the sonochemical qualities required for practical applications by investigating the degree of degradation of indigo carmine. Our results revealed that a multi-orifice horn with orifices that were sufficiently apart to prevent the generated bubbles was well suited for microbubble generation and sonochemical reactions.

Key words
Microbubble, Ultrasonic Horn, Sonochemistry, Dissolved Oxygen Concentration, Optimization

1. Introduction
Microbubbles with diameters of less than 100 μm are currently attracting considerable attention and are being widely used in various fields owing to their properties such as large surface area per unit volume, low rising velocity, and self-pressurization caused by surface tension. Conventionally, microbubbles are generated by releasing gas through a small orifice [1] or two-phase flow nozzle [2-4], or by the depressurization of supersaturated water [5]. However, the technique whereby gas is released through small holes such as needles or through porous media is not suitable for generating microbubbles of less than 100 μm or for producing large quantities of microbubbles. Other microbubble generation methods also offer little control over the microbubble size. Makuta et al. developed a new microbubble generator using a hollow cylindrical ultrasonic horn (HUSH), and they reported that it could easily generate microbubbles with diameters of less than 100 μm [6]. However, this HUSH-based generation cannot yet generate a gas flow rate that is suitable for industrial use, thus the available gas flow rate must be increased to make the technique commercially viable. In this study, we developed a HUSH-based microbubble generator with orifices that can be changed, and we evaluated the effect of the orifice shape on microbubble generation based on the time evolution of the dissolved oxygen concentration in bubbled water, as well as the bubble diameter distribution. In addition, Makuta et al. also reported that a HUSH-based microbubble generator can be applied to a sonochemical reactor [7], which relies on the high temperatures and pressures generated in the ultrasonically oscillating microbubbles [8]. We also evaluated the suitability of a HUSH-based generator with multiple orifices for sonochemical applications by studying the degree of degradation of indigo carmine. Our results revealed that a HUSH-based generator with multiple orifices that were sufficiently apart to prevent the generated bubbles from coalescing was well suited for microbubble generation and sonochemical reactions.

2. Experiment
2.1 Microbubble generator
A microbubble generator consists of an ultrasonic generator (Model 6271, KAIJO Co., Ltd., Japan), an ultrasonic transducer (Transducer 6281A, KAIJO Co., Ltd., Japan), a gas supply source, and the HUSH, as shown in Fig. 1. The generator can transmit a sine wave signal of 19.5 ± 0.5 kHz to the transducer. The oscillation amplitude at the end of the HUSH increases linearly with the electrical power being input to the transducer [6], which is measured using a wattmeter (Power Hitester 3332, HIOKI E. E. Co., Japan).

Fig. 1 Configuration of the microbubble generator

Fig. 2 The internal and external shape of the HUSH
Figure 2 shows a photograph of the HUSH. The HUSH, made of 6Al-4V titanium alloy, has a flow path with an inner diameter of 3 mm and a detachable tip. Gas is fed from the inlet located on the side of the HUSH to the end of the HUSH. The gas being emitted into the liquid through the HUSH forms a gas-liquid interface at the end of the HUSH. A large number of microbubbles form when the end oscillates ultrasonically in response to the electrical power input.

2.2 HUSH orifice types

Microbubble generation from the HUSH is a result of the fragmentation of a capillary wave on the gas-liquid interface formed near the orifices at the end of the HUSH [9]. Thus, microbubble generation is thought to be enhanced by increasing the area of contact between the water, gas, and HUSH, where the ultrasound oscillation is more intense. We therefore clarified the effect of the orifice shape on microbubble generation by using one tip with a single orifice and three other types of tips, each with three orifices. Regarding the multi-orifice configuration at the end of the HUSH, Aizawa et al. reported that the coalescence of bubbles is more likely to occur when the center-to-center distance (dc-c) between circular orifices in a multi-orifice configuration is relatively small [10]. We also clarified the effect of the dc-c value on microbubble generation for a 3-orifice configuration.

Figure 3 shows four types of HUSH tips attached to the horn end. Figure 3 (a) shows a circular orifice that is 6 mm in diameter. Figures 3 (b - d) show φ3.5-dc-c, 4.1 orifices (three orifices, each 3.5 mm in diameter, dc-c = 4.1 mm), φ3.5-dc-c, 5.3 orifices (three orifices, each 3.5 mm in diameter, dc-c = 5.3 mm) and φ3.5-dc-c, 8.1 orifices (three orifices, each 3.5 mm in diameter, dc-c = 8.1 mm) respectively. All these orifices have the same area, and the amplitude of the ultrasonic oscillation is maintained at 31 μm for each HUSH tip.

2.3 Experimental methodology

2.3.1 Measurement of dissolved oxygen

Microbubbles are excellent at dissolving gas because they have a large surface area per unit volume and a slow floating velocity, unlike bubbles with diameters of over 100 μm, which float to the surface quickly due to their rapid floating velocity. In this study, we evaluated the microbubble generation ability of the HUSH by measuring the oxygen dissolution concentration in water (DO) using oxygen microbubbles. For this DO measurement, we used a water tank (80 mm × 80 mm × 700 mm) containing 3.7 L of pure water as the test section. Using a pump (LF521402, ITT Industries Co., Ltd., US), we generated a downward flow with an average velocity of 6.1 mm/s in the water tank. Because the diameter of the bubbles with a floating terminal velocity of 6.1 mm/s is 100 μm, any bubbles with diameters of less than 100 μm are circulated by the downward flow until they either dissolve completely or coalesce, i.e., the rapid DO increase corresponds to the increase in the number of generated microbubbles in this measurement. The DO in the test section and its time evolution were measured by using a dissolved oxygen meter (SevenGo pro SG6, Mettler Toledo Inc., CHE) inserted through a sampling port at the bottom center of the test section. As for this DO observation, interference by the bubble attachment to DO probe was not confirmed because the DO probe having the fine mesh on the edge was horizontally inserted from side wall of test section and was located at the bottom of test section. We measured the dissolved oxygen concentration in the water containing the oxygen microbubbles generated by the HUSH at a gas flow rate of 50 mL/min every 20 s.

2.3.2 Indigo carmine degradation experiment

To evaluate the sonochemical efficiency, we degraded indigo carmine. Indigo carmine (WAKO Co., Ltd., Japan) is used for dying clothes and has a carbon-carbon double bond with a peak absorbance at 610 nm [10]. This double bond is degraded by heating to >300°C, either by UV irradiation or by ozone exposure [10, 11]. Therefore, indigo carmine can be degraded by high temperatures and pressures of a sonochemical reaction. We evaluated the sonochemical capability of the HUSH by measuring the degree of degradation of the indigo carmine.

Dissolved indigo carmine was degraded and quantified by the following processes. (1) 100 mL of 20-ppm indigo carmine aqueous solution was placed in a 150-mL glass beaker and maintained at 10 °C in a water bath (TM-1, AS-ONE Co., Ltd., Japan). (2) The HUSH, positioned 13 mm below the surface of the solution, was ultrasonically oscillated for 30 min, whereas argon gas was supplied at gas flow rates of 0, 10, 30, 50, and 100 mL/min. (3) The transmittance at 610 nm in the solution degraded by the previous process was measured using a UV-Vis spectrometer (SEC2000 Spectrometer, ALS Co., Ltd.,...
Japan) and then the indigo carmine concentration was calculated by the application of a standard curve.

3. Results and Discussion

3.1 Effect of orifice type on HUSH ability to generate microbubbles

Figure 5 shows the time evolution of $DO$ for the $\Phi 6$, $\Phi 3.5-d_{c-c} 4.1$, $\Phi 3.5-d_{c-c} 5.3$, and $\Phi 3.5-d_{c-c} 8.1$ outgassing orifices oscillating with $31 \mu m$. The rapid increase in $DO$ shown in Fig. 5 points to an increase in the quantity of generated microbubbles. These are carried down by the flow in the test section, and exhibit excellent solubility due to their large surface area per unit volume. Figure 6 shows microbubble generation under in same conditions as in Fig. 5, as captured by a high-speed camera (FASTCAM SA1.1, PHOTRON Co., Ltd., Japan) and then the indigo carmine concentration was calculated by the application of a standard curve.

Consequently, these results point to the fact that the use of multiple orifices is sufficient to suppress the coalescence of the generated bubbles, and is thus better suited to microbubble generation.

Fig. 5 Time evolution of dissolved oxygen concentration with oxygen microbubbles generated by HUSH with $\Phi 6$, $\Phi 3.5-d_{c-c} 4.1$, $\Phi 3.5-d_{c-c} 5.3$, and $\Phi 3.5-d_{c-c} 8.1$ orifices

Fig. 6 Photographs showing microbubble generation near the oscillating HUSH end with $\Phi 6$, $\Phi 3.5-d_{c-c} 4.1$, $\Phi 3.5-d_{c-c} 5.3$, and $\Phi 3.5-d_{c-c} 8.1$ orifices

Fig. 7 Bubble diameter distribution for $\Phi 6$, $\Phi 3.5-d_{c-c} 4.1$, $\Phi 3.5-d_{c-c} 5.3$, and $\Phi 3.5-d_{c-c} 8.1$ orifices
3.2 Effect of orifice type on sonochemical reaction

A microbubble generator with the HUSH can also be applied to a sonochemical reactor [7]. Thus, we evaluated the sonochemical capability when using the HUSH with the $\phi 3.5\times d_{c}=8.1$ orifice that had been shown to offer the best microbubble generation ability in the experiment that examined the degradation of indigo carmine. Figure 8 is a graph of the degradation rates of indigo carmine when using a HUSH with a $\phi 3.5\times d_{c}=8.1$ orifice and with the $\phi 6$ orifice both oscillating with 31 $\mu$m, versus the argon gas flow rate ($Q$). Figure 8 shows that the degradation rates of indigo carmine when using a HUSH with a $\phi 3.5\times d_{c}=8.1$ orifice were consistently greater than when using a HUSH with a $\phi 6$ orifice, exhibiting a similar microbubble generation capability as that shown in Fig. 5.

![Fig. 8 Degradation rate of indigo carmine versus argon gas flow rate ($Q$)](image)

Microbubbles in an ultrasonic field tend to spherically oscillate volumetrically due to their high surface tension, thus instantaneously raising the pressure and temperature inside the bubble at the minimum volume. The high pressure and high temperature field directly cleaved a carbon-carbon double bond in indigo carmine by the heat or indirectly cleaved it by the free radicals generated from water in this field [11, 12]. Therefore, the microbubble generation capability of a HUSH is almost the same as the sonochemical capability of a HUSH, given that a HUSH with multiple orifices can degrade indigo carmine at a faster rate than a HUSH with only a single orifice.

Regarding the gas flow rate, an argon gas supply in excess of 50 mL/min led to a decrease in the degree of degradation, as shown in Fig. 8. These decreases are also thought to be caused by an increase in the frequency of large-bubble generation because these relatively large bubbles whose surface tension is smaller utilize the ultrasonic pressure not only for compression and expansion of bubbles but also for the transformation of the non-spherical bubble shape, moreover, the large surface area of these bubbles muffled or reflected the pressure propagation [7, 10].

As a result, a HUSH with multiple orifices can degrade the indigo carmine degradation than the HUSH with single orifice and in thought to be applicable to widespread sonochemical reaction because it can generate the maximum possible number of microbubbles while simultaneously minimizing the generation of large bubbles.

4. Conclusion

We developed a microbubble generator with a HUSH with changeable orifices, and we evaluated the effect of the orifice type on microbubble generation according to the time evolution of the dissolved oxygen concentration in bubbled water and the bubble diameter distribution. These results revealed that the use of multiple orifices that were sufficiently far to suppress the coalescence of the generated bubbles could stably generate large amounts of microbubbles.

In addition, we also evaluated the sonochemical capability of a HUSH with multiple orifices by examining the degree of degradation of indigo carmine. These results also revealed that the sonochemical capability increases with an increase in the amount of microbubbles, because the microbubbles in the ultrasonic field tend to spherically oscillate volumetrically due to their considerable surface tension, thus instantaneously raising the pressure and temperature inside the bubble at the minimum volume.

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