5aPA8. Effect of gases on radical production rates during single-bubble cavitation

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The yields of hydroxyl radicals and nitrite ions produced from single-bubble cavitation were quantified while the corresponding dynamics of the single bubble was observed by stroboscopic and laser-light scattering methods. The numbers of both hydroxyl radicals and nitrite ions per cycle were proportional to pressure amplitude at 25 kHz under the conditions of stable sonoluminescing single-bubbles. Under the dancing single-bubbles without sonoluminescence below sonoluminescence threshold for air bubbles, however, the number of hydroxyl radicals was larger than that for the stable single-bubble and nitrite ions were not detected in contrast to hydroxyl radicals. For argon bubbles, the numbers of hydroxyl radicals were larger than those for air bubbles and the number of hydroxyl radical was significantly increased with the dancing bubble by argon bubbling. The results imply that the shape instability may promote the dissolution and the diffusion of hydroxyl radicals from the bubble into the liquid and the part of hydroxyl radicals may convert into nitrite ions in the case of the air bubble above the threshold of sonoluminescence.

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INTRODUCTION

Intense ultrasound in liquids provides a unique environment where high-energy chemical reactions occur [1]. The chemical effects of ultrasound originate from hot spots formed during the collapse of acoustic cavitation bubbles. These hot spots have temperatures of roughly 5000 K [2, 3], pressures of about 300 atm [4], and usually emit bluish light called sonoluminescence [5, 6]. Inside cavitation bubbles in an aqueous solution, water vapor is decomposed to form hydrogen atoms and hydroxyl radicals (OH•) [7], part of which dissolves into liquid. Therefore, there are mainly two different reaction paths and sites: pyrolysis inside gas bubbles and radical reactions in liquid around the gas bubbles. Since many reactions are considered to be associated with OH radicals in liquid phase, higher efficiency of OH radicals dissolving into liquid is one of the important keys in sonochemistry.

However, it is difficult to study what parameters can influence the OH radical production in multibubble cavitation. Multibubble cavitation is complicated phenomena because of uncontrollable bubble dynamics, which involves formation, growth, coalescence, fragmentation of a large number of cavitation bubbles. The discovery of single-bubble sonoluminescence has helped greatly in the study of cavitation bubble dynamics in a highly controlled and repeatable fashion [8]. Then, Didenko and Suslick reported the amount of OH radicals during single-bubble cavitation [9], but they did not measure the amount of OH radicals under an unstable bubble condition.

In this study, the amount of OH radicals produced by a single cavitation bubble including an unstable single bubble known as a “dancing” bubble is quantified, while the corresponding bubble dynamics is always monitored by stroboscopic observation to confirm whether the bubble is stable or not. Furthermore, in order to clarify the mechanism for the enhancement of OH radicals by bubble dancing, OH radicals are measured in a pure argon bubble and nitrite ions are measured in a dancing air bubble.

EXPERIMENT

The Experimental setup is shown in Fig. 1. A continuous sinusoidal signal generated by a function generator (NF Electronic Instruments, WF1946) was amplified by a power amplifier (Yokogawa, 7058-10) and fed to a bolted Langevin-type transducer of 45 mm diameter (Honda Electronics). The transducer was fixed to a stainless steel plate at the bottom of a rectangular cell, which was 56×56×80 mm³ internal dimensions and made of quartz glass of 2 mm thickness. Aqueous 2 mM terephthalic acid solution, which was partially degassed to 20% saturation at 293 K, was filled to 70 mm depth in the cell. The free surface of the solution was covered with paraffin film to suppress the dissolution of air. A bubble was inserted with a syringe and trapped at a pressure antinode in a standing wave field at a resonant frequency of 24.5 kHz. By adjusting pressure amplitude, SBSL was obtained. The acoustic pressure at the bubble position was measured with a needle hydrophone (DAPCO, NP10-3) and corrected with a calibrated hydrophone (Brüel & Kjær, 8103). The dynamics of the single bubble was observed with a CCD video camera through a zoom lens by stroboscopic backlight of 90 ns pulse width (Sugawara, NP1A-U1) and 30 Hz flash as same as video rate. By the difference of 0.5 Hz between ultrasonic and stroboscopic frequencies, the bubble dynamics during one period was observed apparently for 2 s [10].

FIGURE 1. Experimental setup.
RESULTS AND DISCUSSION

Figure 2 shows radii of single bubbles as a function of time during one period at 24.5 kHz for acoustic pressures of 1.40, 1.30, 1.13, and 1.07 atm. These radius curves were obtained by laser-light scattering method with scaling using the stroboscopic images of the bubbles at maximum and ambient sizes (Fig. 3). In Fig. 2, relative intensities of SL and measured acoustic pressures at the position of the bubble are also shown. The maximum radius of single bubble and the height of SL pulse decrease with a decrease in acoustic pressure. In contrast, at 1.07 atm the maximum radius does not significantly decrease, where the SL pulse is not detected. Under this condition the bubble was no longer stable and “dancing.” It is known that under the condition below the SL threshold a single bubble pinches off daughter bubbles and dances by the recoil of the remaining bubble [11, 12]. Splitting bubbles are confirmed in Fig. 3 (d).

Figure 4 shows the amount of OH radicals per cycle produced by the single bubbles whose dynamics at 24.5 kHz is shown in Figs. 2 and 3, as a function of acoustic pressure amplitude. Under the stable bubble conditions, the amount of OH radicals increases with pressure amplitude. Under the dancing bubble condition, however, the amount of OH radicals increases with a decrease in pressure amplitude.

![Graph showing bubble radius, sonoluminescence (SL) and acoustic pressure amplitude vs. time curves at 24.5 kHz for various sound pressure amplitudes. At 1.07 atm the single bubble was unstably dancing and was accompanied by no SL.](image)

FIGURE 2. Bubble radius, sonoluminescence (SL) and acoustic pressure amplitude vs. time curves at 24.5 kHz for various sound pressure amplitudes. At 1.07 atm the single bubble was unstably dancing and was accompanied by no SL.

![Stroboscopic images of the single bubbles of maximum (left) and ambient (right) sizes at 24.5 kHz for pressure amplitudes of (a) 1.40 atm, (b) 1.30 atm, (c) 1.13 atm and (d) 1.07 atm. Selected images of bubble fragmentation of the dancing bubble is shown in (d).](image)

FIGURE 3. Stroboscopic images of the single bubbles of maximum (left) and ambient (right) sizes at 24.5 kHz for pressure amplitudes of (a) 1.40 atm, (b) 1.30 atm, (c) 1.13 atm and (d) 1.07 atm. Selected images of bubble fragmentation of the dancing bubble is shown in (d).
FIGURE 4. Dependence of the amount of OH radicals produced by the single bubbles per cycle on acoustic pressure amplitude. The dynamics of the single bubbles at 24.5 kHz corresponds to the bubble dynamics shown in Figs. 2 and 3.

FIGURE 5. The number of nitrite ions and hydroxyl radicals per acoustic cycle at 25 kHz. At 1.07 atm the bubble was dancing without sonoluminescence.

FIGURE 6. The number of hydroxyl radicals per acoustic cycle at 25 kHz. In Ar bubble case the dancing bubble was obtained by Ar gas bubbling.
FIGURE 7. Selected images of bubble fragmentation of the argon dancing bubble.

FIGURE 8. Radius-time curves of the stable and dancing bubbles for Ar bubbles at 1.3 atm in Fig. 6.

CONCLUSION

The numbers of both hydroxyl radicals and nitrite ions per cycle were proportional to pressure amplitude under the conditions of stable sonoluminescing single-bubbles. Under the dancing bubbles without sonoluminescence below sonoluminescence threshold for air bubbles, however, the number of hydroxyl radicals was larger than that for the stable single-bubble and nitrite ions were not detected. For argon bubbles, the numbers of hydroxyl radicals were larger than those for air bubbles and the number of hydroxyl radical was significantly increased with the dancing bubble by argon bubbling. The results imply that the shape instability may promote the dissolution and the diffusion of hydroxyl radicals from the bubble into the liquid and the part of hydroxyl radicals may convert into nitrite ions in the case of the air bubble above the threshold of sonoluminescence.

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