Relationship between Sonoluminescence, Radical Production and Bubble Dynamics during Single-Bubble Cavitation

単泡性キャビテーションのソノルミネッセンス, ラジカル生成 と気泡ダイナミクスの関係

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1. 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⁾ pressures of about 500 atm,³⁾ and usually emit bluish light called sonoluminescence (SL).⁴⁾ Inside cavitation bubbles in an aqueous solution, water vapor is decomposed to form H atoms and OH radicals,⁵⁾ part of which is dissolved into liquid. Therefore, there are mainly two different reaction sites, which are pyrolysis inside gas bubbles and oxidation by OH radicals in liquid. Since many sonochemical reactions are associated with OH radicals in liquid phase, higher efficiency of OH radical production is one of the most 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.⁶⁾ Then, Didenko and Suslick reported the amount of OH radicals during single-bubble cavitation,⁷⁾ but they did not discuss with the corresponding bubble dynamics in detail.

In this study, the amount of OH radicals produced by a single bubble is measured, while the corresponding bubble dynamics is monitored by stroboscopic observation, including the condition of an unstable bubble known as "dancing" bubble.

2. Experimental

The Experimental setup is shown in **Fig. 1**. A continuous sinusoidal signal generated by a function generator (NF, WF1946) was amplified by a power amplifier (Yokogawa, 7058-10) and fed to a bolt-clamped Langevin-type transducer of 45 mm



Fig.1. Experimental setup.

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 or 33.3 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 а 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. By the difference of 0.5 Hz between ultrasonic and stroboscopic frequencies, the bubble dynamics during one period was observed apparently in slow motion for 2 s.⁸⁾

3. Results and Discussion

Figure 2 shows radii of single bubbles as a function of time during one cycle of the ultrasound at 24.5 kHz for acoustic pressure amplitudes of 1.40, 1.30, 1.13, and 1.07 atm. These radius curves were obtained by laser-light scattering method with

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scaling using the stroboscopic images of the bubbles at maximum and ambient sizes in Fig. 3. In Fig. 2, relative intensities of SL for the bubbles and measured acoustic pressures at the position of the bubble in the ultrasonic field are also shown. The maximum radius of single bubble decreases with a decrease in acoustic pressure amplitude. The height of SL pulse at collapse also decreases remarkably. In contrast, at 1.07 atm the maximum radius does not significantly decrease and the ambient radius increases, where the SL pulse is not detected. Under this condition the bubble was no longer stable and "dancing." It is known that in the region lower SL threshold of acoustic pressure amplitude a single bubble is pinching off micro-bubbles called "daughter bubbles" and "dancing" by the recoil of the remaining bubble.⁹⁾ Splitting bubbles are confirmed by the stroboscopic images in Fig. 3 (d).

Figure 4 shows the amount of OH radicals per cycle produced by the single bubbles for 24.5 and 33.3 kHz, the dynamics of which are shown in Figs. 2 and 3 for 24.5 kHz, as a function of acoustic pressure amplitude. Under the stable bubble conditions, the amount of OH radicals increases with pressure amplitude, which is consistent with results reported by Didenko and Suslick.⁷⁾ Under the dancing bubble condition, however, the amount of OH radicals increases, in spite of a decrease in pressure amplitude. The amounts of OH radicals produced by the dancing bubbles are more than three times as high as those produced by the stable bubbles, according to the extrapolation value of the amount of OH radicals for the stable bubbles.

4. Conclusion

The amount of OH radicals produced by a stable sonoluminescing bubble was proportional to acoustic pressure amplitude, where the maximum bubble radius and the intensity of SL also increased with the acoustic pressure amplitude. The amount of OH radicals produced by a dancing bubble, however, was higher than that produced by the stable bubble, in spite of lower pressure amplitude, where no SL was detected from the dancing bubble.

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Fig. 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.



Fig. 3. Stroboscopic images of the single bubbles of maximum (left) and ambient (right) sizes at 24.5 kHz for the amplitudes of (a) 1.40, (b) 1.30, (c) 1.13 and (d) 1.07 atm. Bubble fragmentation of the dancing bubble is seen in (d).



Fig. 4. Acoustic pressure dependence of the amount of OH radicals per cycle produced by the single bubbles shown in Figs. 2 and 3.

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