

On the influence of degree of gas saturation on multibubble sonoluminescence intensity

溶存気体飽和度が音響発光強度に与える影響

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1. Introduction

Cavitation bubble in a liquid under intense ultrasound can provide extreme condition of high pressure and high temperature inside the bubble at violent collapse^{1,2}. When a bubble collapses it emits light; this phenomenon is known as sonoluminescence (SL)³.

The addition of volatile solutes to an aqueous solution greatly reduces the SL intensity relative to that in pure water by lowering the cavitation bubble temperature^{4,5}. Alcohol molecules adsorb on the surfaces of bubbles and then enter the bubbles during expansion caused by an ultrasonic standing wave that has the potential to realize stable pulsation of bubbles^{4,6}. Hydrocarbon products^{5,7} are then created inside the bubbles. Decomposition of hydrocarbon products that occurs remarkably at high alcohol concentration when the bubble collapses reduces the temperature inside the bubble and quenches the SL.

The degree of saturation (DOS) of a gas dissolved in a solution is one of the main parameters that determines the sonochemical reaction efficiency. The DOS generally decreases as sonication proceeds. This is due to a release of bubbles from a sonicated system through the growth of a bubble by both rectified diffusion and coalescence between bubbles by the action of secondary Bjerknes force. Rectified diffusion increases the bubble volume since more gas diffuses into a bubble during expansion than diffuses out of the bubble during compression since the bubble has a larger surface area during expansion. The secondary Bjerknes force is an attractive force between bubbles that are smaller than the resonant size at an antinode in an ultrasonic standing wave.

The present study investigates the influence of adding ethanol on the SL intensity for 261-kHz ultrasound irradiation at different DOS of air in solution⁸.

2. Experiment

A continuous sinusoidal signal with a frequency of 261 kHz was generated by a function generator and amplified by a power amplifier to drive a plane transducer of 50 mm diameter. The transducer was

attached to a circular stainless-steel plate of 100 mm diameter and 1 mm thickness set at the bottom of a rectangular glass vessel (inner dimensions: 56×56×80 mm; side wall thickness: 2 mm). The ultrasonic power absorbed by the liquid was determined by calorimetry. The present experiment was conducted at 0.34 W/cm².

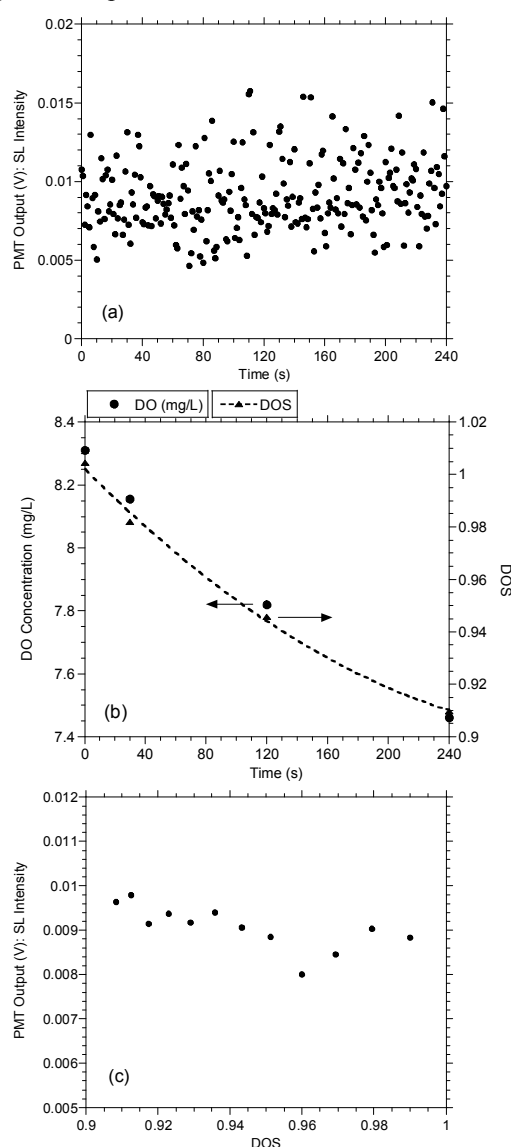


Figure 1. Conversion from time dependence to DOS dependence of SL intensity by curve fitting the time-dependent DOS estimated from measured DO data. (a) Example of time dependence of SL intensity. (b) DOS as a function of time determined from a curve fitted to DOS data, which was estimated from the measured time dependence of the DO concentration. (c) Example of DOS dependence of SL intensity. (from Ref. 8 under © 2011 American Chemical Society)

The ethanol used had a purity of $\geq 99.5\%$ and the solutions were made up using distilled water. 200 mL of an air-saturated solution was poured into the glass vessel. The solution temperature was initially 23 °C. Temperature rise was within a few degrees on ultrasonic irradiation. The SL intensity from the solution in the vessel was measured using a photomultiplier tube. The output voltage from the photomultiplier tube was measured using a digital multimeter and recorded on a computer. Sonication was performed for 2 min, stopped for 2 min, and then performed for a further 2 min. Thus, the total sonication time was 4 min. The present experiment was conducted using ethanol concentrations of 12, 25, 37, 50, 75, 100, and 130 mM. The dissolved oxygen concentration of each solution was measured relative to the dissolved air concentration using a dissolved oxygen (DO) meter. The DOS is defined as the relative DO concentration divided by the saturated DO concentration at the same temperature.

3. Results and Discussion

We describe here the procedure used to estimate the DOS dependence of the SL intensity. Figure 1(a) shows an example of the time dependence of the SL intensity. Figure 1(b) shows the time dependence of the DO concentration for the same conditions as Fig. 1(a); the DOS was determined from the DO concentration. Curve fitting was carried out on the DOS data to allow the DOS to be estimated at any sonication time. The DOS dependence of the SL intensity is estimated by eliminating the time parameter from the time dependences of both the SL intensity and the DOS (Fig. 1(c)). Note that the SL and DOS data in Fig. 1(c) represent averages over 20 s.

Figure 2 shows the DOS dependence of the SL intensity at various ethanol concentrations. The sonication proceeds from right to left, since the DOS generally decreases with bubble degassing. It is found

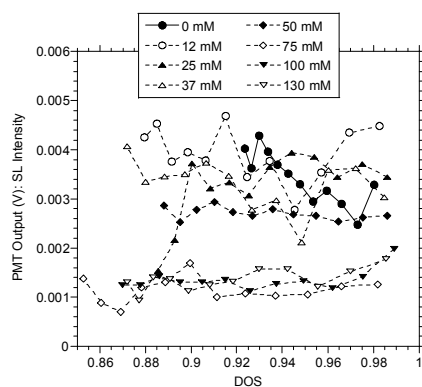


Figure 2. DOS dependence of SL intensity for various ethanol concentrations. (from Ref. 8 under © 2011 American Chemical Society)

that solutions containing ethanol had a lower SL intensity than pure water (0 mM). The SL intensity at 0 mM seems to increase as the DOS decreases, whereas the SL intensity from the solution containing ethanol seems to be almost constant or decreases, although the data are rather scattered. At a high DOS (i.e., ~ 0.94 - 0.99), the SL intensity at relatively low ethanol concentrations (12, 25, and 37 mM) is higher than that for pure water.

From Fig.2, the DOS for solutions containing ethanol seems to be much lower than that for pure water. This reduction in the DOS is considered to be caused by bubble degassing in the solution due to bubble growth by rectified diffusion, since adding ethanol inhibits bubble coalescence⁹. Lee et al.⁹ suggested that rectified diffusion is responsible for an increase in the total bubble volume at higher alcohol concentrations. A higher bubble volume is thought to promote degassing from the solution and reduce the DOS. For pure water, bubble degassing mainly occurs due to bubbles coalescing. As the DOS of pure water decreases as sonication proceeds, the number of large bubbles decreases and tiny active bubbles are distributed over a wider region¹⁰. In this case, little coalescence of bubbles occurs and the degassing rate of pure water is lower than that of solutions containing ethanol. A relatively high DOS is maintained for pure water and this results in more bubbles than for solutions containing ethanol. Thus, pure water has a higher SL intensity than solutions containing ethanol in suitably degassed conditions. The enhancement of SL intensity at relatively low ethanol concentrations at high DOS comes from both little decomposition of hydrocarbon products and the number of active bubbles which is larger than that at pure water.

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