INFLUENCE OF SURFACTANTS ON INSTABILITY OF A SONOLUMINESCING BUBBLE

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Abstract

Influence of a surfactant additive on instability of a sonoluminescing single bubble (SB) is studied through a directly stroboscopic observation of the bubble. The surfactant additives of sodium dodecyl sulfate (SDS) enhance the instability of the bubble. Therefore. upper stable bubble region with sonoluminescence (SL) became narrower as the concentration of SDS increased. Instead, the dancing bubble region became broader, where a bubble ejects tiny bubbles, making it dance by counteraction. Since the instability of bubble collapse is a key parameter for sonochemistry in the bulk liquid region, surfactant additives have potential for promoting sonochemical reactions. We confirmed it by sonochemical oxidation of I⁻ in the presence of CCl₄. The quantity of I₃⁻ due to a dancing SB with SDS was more than two times larger than that due to the stable SB without SDS.

Introduction

Effects of surfactants on sonoluminescence (SL) have been intensively investigated, especially by Grieser and his colleagues [1]. For single-bubble (SB) cases, there are also some literatures since Stottlemyer and Apfel first reported [2]. They reported that Triton X-100 reduced the maximum radius of the SB, thereby decreasing the SBSL intensity. On the other hand, Ashokkumar et al. reported that SDS in low concentration range did not significantly affect the radial dynamics nor the SL intensity of a SB [3]. Yanagita et al. reported that SDS accelerated the breathing motion of a SB but had no effect on its maximum radius, while the SBSL was quenched [4]. Thus the effects of surfactants are complicated and then are worthy to be studied further. In this study, we focus on the influence of SDS on the instability of SB dynamics.

Experimental Methods

Figure 1 shows experimental setup. A continuous sinusoidal wave at 24.5105 kHz generated with a function generator (NF Electronic Instruments, 1946) was amplified with a 50 dB power amplifier (ENI, 240L) and fed to a bolted Langevin-type transducer (Honda Electronics) at the bottom of a cell. The rectangular glass cell had $56 \times 56 \times 80$ mm³ internal dimensions. Partially degassed distilled water of 220 mL at 22°C and about 2 mg/L dissolved oxygen was filled to 70 mm depth. By adjusting the function generator outputs, a bubble inserted with a syringe



Fig. 1 Experimental setup.

was trapped at a pressure antinode of the standing wave and SBSL was emitted We observed the SB dynamics with a charge-coupled device (CCD) camera through a zoom lens by stroboscopic backlight of 90 ns pulse width (Sugawara, NP1A-U1), while decreasing the function generator output. The function generator outputs were considered as relative acoustic pressure amplitudes and in some cases the acoustic pressure amplitude was measured with a calibrated hydrophone (RESON, TC4038) at the position of the bubble. The stroboscopic frequency was set 30 Hz as same as video rate. By the phase difference of 0.5 Hz between ultrasonic and stroboscopic frequencies, we observed the bubble dynamics during one period apparently for 2 s [5].

Results and Discussion

Figure 2 shows stroboscopic images as examples, which were selected from the video every five frame at 810 mV_{p-p} function generator output for 0, 150 and 300 μ M SDS solutions. It is seen that the bubble is unstable at 300 μ M SDS. In the video movie we can confirm more clearly whether the bubble is stable or not. From the observations of SB dynamics for various SDS concentrations, we obtained the phase diagram for bubble instability as shown in Figure 3 for SDS concentration vs. the function generator outputs. In Figure 3 the dancing bubble region extends as the SDS concentration increases. In the dancing bubble region a new phase of moving bubble region and the function generator outputs.

At first, in the upper stable region, we show the dependence of bubble dynamics on sound pressure amplitude in the absence and the presence of SDS.





Figure 3 : Effect of SDS concentration on bubble instability threshold for acoustic pressure.

Figure 4 shows the radius-time curves by laser scattering method. Figure 4 (a) is the case of distilled water and Fig. 4 (b) is the case of 100 µM SDS aqueous solution. Each curve was the averaging 32 data. As well known, the maximum bubble radius gradually decreases with decreasing sound pressure in the absence of SDS. In contrast, the maximum bubble radius suddenly decreases at 1.06 atm in the presence of SDS, where we confirmed the single bubbles were stable and spherical by stroboscopic observation. We expected that the SB expands easier in the presence of surfactants because of lower surface tension during the expansion period [6], but the result is contrary. By the way, the acoustic pressure amplitudes seem too low. It may be due to extrapolated calibration data used. The practical pressure amplitudes may be higher.



Figure 4 : Dependence of the radius-time curves on acoustic pressure amplitude for (a) 0 μ M and (b) 100 μ M SDS solutions.



Figure 5 : Light emission pulse of the SBSL under the condition of spherical bubble oscillation for 0, 50 and 100 μ M SDS solutions.

Figure 5 shows the SL pulse at relatively high pressure of 1.33 atm for 0, 50 and 100 μ M SDS solutions. The same timing of SBSL pulse means that all the bubble dynamics were almost the same because of the same collapse timing. It is clear that the height of SBSL pulse for 100 μ M SDS is the smallest and about half as high as that for 0 μ M. The pulse height for 50 μ M SDS is more scattered relative to that for 0 μ M. The height changes about half and twofold by turns. One of the authors, Yasui, have suggested by computational simulation that the surfactant effect is

caused by the inhibition of condensation of water vapor at the bubble wall during collapse, which results in lowering the achieved temperature inside a bubble due to the increase in amount of vapor that undergoes endothermal chemical reactions [7].

Next, the lower stable thresholds in Fig. 3 are almost the same for various SDS concentrations. But we observed peculiar bubble behavior in this region in the presence of SDS. Figure 6 shows the selected video frames of the bubbles in the lower stable region for 150 μ M SDS. Twin bubble oscillation continued for more than 4 s, which means that this twin bubble state kept for more than ten thousands ultrasonic cycles.

Finally, we compared the sonochemical yield due to a stable sonoluminescing bubble in the absence of SDS with that due to an unstable dancing bubble in the presence of SDS using Weisler reaction, which is the sonication of iodide ion in the presence of tetrachloride. We performed the experiment referred to that by Lepoint et al. [8]. In this experiment we covered the free surface with Parafilm $M^{\text{(R)}}$ to suppress the dissolution of air. Figure 7 shows UV-vis spectra of 1 M aqueous sodium chloride solutions after 2.5 h sonications. The sonochemical product of I_3^- has absorbance at 350 nm. It is seen that the sonochemical product with 230 μ M SDS is 2.5 times as large as that without SDS. Since the unstable dancing bubble can cause sonochemical reaction in the bulk liquid region



Figure 6 : Selected stroboscopic images at 590 mV_{p-p} for 150 μ M SDS solution. Two-mode surface wave oscillation continued more than 4 s.



Figure 7 : UV-vis spectra of 1 M aqueous NaI solutions in the presence of CCl_4 after 2.5 h sonications for the stable bubble without SDS and the dancing bubble with 230 μ M SDS.

more efficiently than the stable bubble [9,10], the instability of bubble with SDS may enhance sonochemical reactions. More amounts of OH radicals produced from the more amounts of vapor trapped inside the bubble by SDS may contribute to the enhancement [7]. The other radicals produced from the dissociation of SDS in the interfacial region may contribute to the enhancement.

Conclusions

The surfactant additive of SDS enhanced the instability of single bubbles. Therefore, upper stable bubble region with SL became narrower as the concentration of SDS increased. Instead, the dancing bubble region became broader. In the dancing bubble region with SDS, sonochemical reaction for the oxidation of iodide ion was enhanced more than two times. The SDS additive also changed the dynamics of single bubbles that still remained stable. In the upper stable region, the maximum bubble radius suddenly decreased with decreasing acoustic pressure amplitude. In the lower stable region, the surface oscillation of bubble continued for more than ten thousands acoustic cycles. On the other hand, in the case of almost the same dynamics of the upper stable bubble, the SBSL intensity decreased with SDS.

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