

## Effect of alkyl chain length of added alcohol on sonoluminescence intensity decay

ソノルミネッセンス強度の低下に対する添加アルコール炭素鎖長の影響

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### 1. Introduction

Multibubble sonoluminescence (MBSL) spectra observed when ultrasound is irradiated to Ar-saturated water can be classified in to two categories; one is continuum spectrum fraction ranging over wide wavelength, and the other is the definite peak located at around 310 nm, assignable to OH. Observation of MBSL has been known to be an appropriate approach to obtain physicochemical information about the acoustic cavitation bubble collapse under sonication. Thus far, the effects of alcohol addition on MBSL quenching have been reported [1]. The spectral changes are presumed to be attributable to gaseous products generated by sonochemical decomposition of alcohols.

The aim of the present study is to systematically investigate the correlation between MBSL and sonochemical decomposition of a series of alcohols.

### 2. Experimental procedure

To obtain Ar-saturated ultrapure water, the water was degassed under the reduced pressure (0.1 MPa) for 2 h, and then placed at ambient pressure of Ar for 2h. The water was stirred while these treatments. After the water was additionally purged by Ar flow, alcohol (methanol, ethanol, 1-propanol, or 1-butanol, 1-pentanol, 1-hexanol) was added using a micro syringe into the closed reactor. The volume of the sample aqueous solution was 135

cm<sup>3</sup> and the concentration of the alcohol was 2 mmol/dm<sup>3</sup>. Fig.1 shows the schematic diagram of the experimental setup. The stain-less steel cylindrical reactor was equipped with a lead zirconate titanate (PZT) disk-type transducer having 410 kHz resonance frequency. The sonication time was 30 min. To keep the temperature of the sonicated sample solution at 13 ± 1 °C, cooling water was circulated through the water jacket covering the reactor. Acoustic power was measured by the calorimetric method to keep it constant at 10 W, corresponding to 3.2 W/cm<sup>2</sup> of ultrasound intensity, in each experiment. MBSL spectra were observed through the quartz glass window using a spectrometer (SP2300i, Princeton Instruments) and a CCD detector (Pixis100, Princeton Instruments). Spectra ranging from 257 to 540 nm could be obtained when a 300 g/mm grating and a 1.00 mm slit were employed. Exposure time was 2 min to acquire MBSL spectra.

Gaseous products in the head space of the reactor were analyzed using gas chromatographs (GCs) after the solutions stood for 30 min to become equilibrium state. H<sub>2</sub> was determined using a GC equipped with a thermal conductivity detector (GC-8A, Shimadzu). Other gases were determined by a GC equipped with a flame ionization detector (GC-2014, Shimadzu). H<sub>2</sub>O<sub>2</sub> concentration in the sonicated solutions was determined by the colorimetry [2].

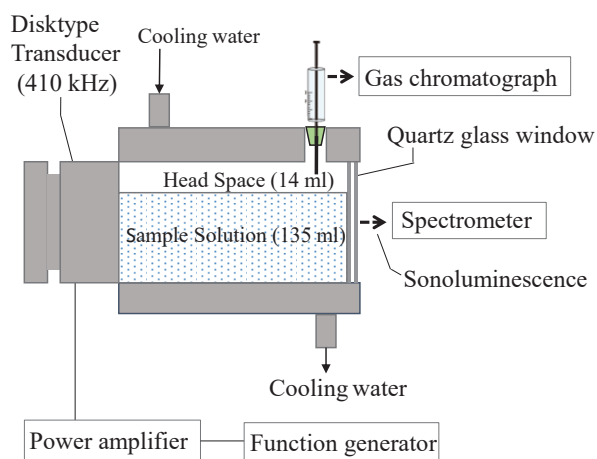


Fig.1. Experimental apparatus for sonication.

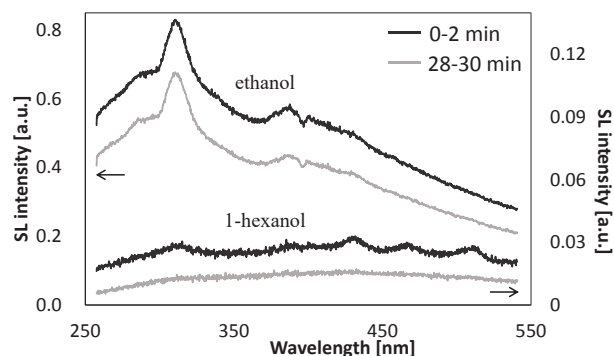


Fig.2. Sonoluminescence spectra from Ar-saturated ethanol, and 1-hexanol solutions.

### 3. Results and discussion

Fig. 2 shows the MBSL spectra immediately after the start (0-2 min) and just before the end (28-30 min) of the sonication when ethanol or 1-hexanol was added to Ar-saturated water. The measurements of the SL spectra were performed three times under each condition. The spectra were averaged and normalized using the intensity at 257 nm obtained from pure water at the start of the sonication (0-2 min) as the standard. The SL intensity was weakened by the addition of alcohols. The effect was emphasized when the alcohol with longer alkyl chain was added. This is considered to be related to the hydrophobicity of the added alcohols because long chained alcohol molecules with higher hydrophobicity seem to accumulate around bubbles generated by sonication, and they enter the bubbles as vapor [3]. When polyatomic molecules such as alcohols are in the bubbles, it is generally known that the specific heat ratio of the content in the bubble is reduced. And if the bubbles are filled with molecules such as alcohols, the cavitation collapse is cushioned. In addition, some energy generated during the collapse is consumed not only for SL, but for the decomposition of the alcohol molecules contained in the bubbles. These are considered to be attributable to weaken the SL intensity. Also, the concentration of generated H<sub>2</sub>O<sub>2</sub> was significantly lower than that of the sonicated pure water, meaning that alcohol molecules scavenge OH radicals to be oxidatively decomposed.

Fig. 3 shows the composition of the generated gaseous products at 2 min and 30 min of sonication when ethanol or 1-hexanol was added. When 1-hexanol was added, a large amount of gas was generated in 30 min sonication, so 1/10 of the actual generated amounts are shown in the figure. Gas generation originated from alcohol decomposition was observed even in the 2 min sonication. Compared with the case of ethanol

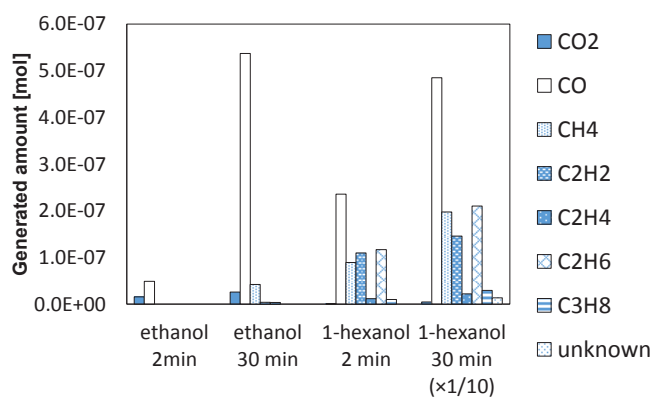


Fig.3. Distribution of carbon-contained gaseous products.

addition, a larger amount of gas was generated when long chain 1-hexanol was added. It is thought that some of the gaseous products are dissolved in the sample aqueous solution and enter the bubbles. As a result, they affect the temperature and the SL intensity. It is noticeable that considerable amounts of acetylene and propane are generated at the start of sonication from 1-hexanol aqueous solution. Because these polyatomic gases have smaller specific heat ratios and higher solubility in water than Ar [4], they are likely to significantly quench SL at the start of ultrasonic irradiation.

Fig. 4 shows the SL relative intensity of the peak attributable to the OH radical at 310 nm when 6 kinds of alcohols were added. The SL intensity was normalized using the SL intensity (0-2 min) of pure water as the standard.

The SL intensity systematically decayed with the alkyl chain length of the added alcohol and the duration of the sonication, suggesting that the SL intensity depends on the compositions and the amounts of the alcohol decomposition products.

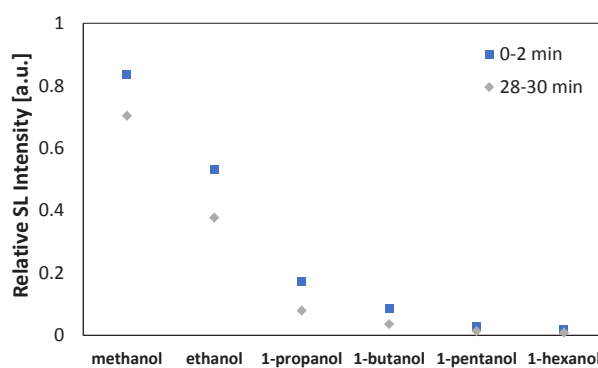


Fig.4. Relative sonoluminescence intensity at 310 nm at the beginning (0-2 min) and the ending (28-30 min) of the sonication.

### References

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