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# Influence of particle addition on sonochemiluminescence intensity under pulsed ultrasound

# パルス超音波照射下における粒子添加が音響化学発光強度に与える影響

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## 1. Introduction

A violent collapse of ultrasonic cavitation bubble can provide extreme conditions at the interior of the bubble, including temperatures of several thousand degrees Kelvin, pressures of several hundred atmospheres, and heating and cooling rates greater than $10^9$ K/s.[1,2] Under such conditions, water is easily decomposed and oxidants such as hydroxyl radicals, hydrogen peroxide and ozone are generated.[3] At the interface of the bubbles, these oxidants react with chemicals such as luminol, and light is emitted in a process known as sonochemiluminescence (SCL).[4] Chemical reactions involving ultrasonic cavitation bubbles are referred to as sonochemical reactions.[2,5]

Addition of particles to such a sonochemical system has the potential to enhance the efficiency of sonochemical reaction as the particles provide nucleation sites for the initiation of cavitation bubbles that are effective for sonochemical reaction.[6-9] The mechanism is as follows. The gas pocket is stabilized due to an action of surface tension in a concave shape at the gas-liquid interface in a crevice on a particle surface surrounded by liquid when external pressure such as ultrasound is not applied. The volume of gas increases under reduced pressure at a cycle of rarefaction phase when an ultrasound is applied. The expanded gas becomes convex to release a free and tiny cavitation bubble from the crevice. Such a tiny cavitation bubble is not only provided repeatedly but also has a potential to pulsate. Accordingly, the addition of particle can contribute to an increase in the number of bubbles active for sonochemical reaction, which leads to the enhancement in the reaction efficiency.

Pulsing ultrasound operation is useful to obtain highly efficient sonochemical reaction,[11-13] because it inhibits the generation of large degassing bubbles that restrict the efficient spatial region for sonochemical reaction. We have previously demonstrated that the residual sound pressure during the pulse-off time, in addition to the spatial enlargement, contribute to the increase in the sonochemical-reaction efficiency.[14] Little is known on the sonochemical effects of pulsed ultrasound under particle addition.[15]

In this study, an influence of particle addition on the sonochemical reaction by pulsed ultrasound was investigated through measurements of the sonochemiluminescence intensity.

## 2. Experiment

A continuous (CW) or pulsed wave sinusoidal signal of 151 kHz was generated by a function generator (NF Electronic Instruments, 1942), and amplified with a power amplifier (ENI, 1140LA) to drive a Langevin-type transducer (Kouwa, 45 mm OD). The transducer was attached to a circular stainless steel plate with a diameter of 110 mm (1 mm thick) set at the bottom of a rectangular glass vessel. The inner dimensions of the glass vessel were 56×56×80 mm, with a 2-mm-thick side wall. Pulsed sonication involved the repetition of 1000 acoustic cycles-ON and 1000 cycles-OFF. Luminol (3-aminophthalhydrazide) solution was used for measurement of the SCL intensity. Luminol reacts with OH radicals generated in the cavitation bubbles to yield aminophthalate anions which cause a blue fluorescence when intense ultrasound passes through the luminol solution.[16] A solution consisting of 0.33 M NaOH (Wako) and 1.9 mM luminol (Wako) was prepared using distilled water and was saturated with air. The air-saturated liquid was poured into the vessel to a liquid height of 64 mm (the liquid volume was 200 mL). The liquid temperature was 25 ºC. The intensity of SCL from the air-saturated liquid in the vessel was measured with a photomultiplier tube (PMT; Hamamatsu, R928). Data were recorded using a microcomputer (NEC, PC-9821 Xc16) through a digital multimeter (Advantest, TR6847) which measures the output voltage of the PMT. The sonication time for each set of data was 1 min. Alumina particles (Adomafine; Adomatechs, AO-809) of 10 μm in mean diameter were added to the solution.

## 3. Results and Discussion

Figure 1 shows the dependence of the measured SCL intensity on the power applied to the transducer. Note

that each set of data is normalized by the maximum at CW in the absence of particles. At relatively low power up to 17 W, pulsing in the presence of particles (for both 0.6 and 1.2 g/L) provides higher intensity than that in the absence of particles at that power. It is significant that the effect of particle addition on the sonochemical reaction efficiency can be obtained with pulsing, and more specifically at relatively low power, compared with that with CW. The maximum SCL intensity in the presence of particles obtained with pulsing was approximately 2 times higher than that without the addition of particles. It is interesting that the increase (2 times) in the sonochemical reaction efficiency with the addition of particles for pulsing is close to that previously obtained for CW, where the increase in efficiency was investigated for various particle sizes and amounts.[9] The peak-out of the intensity with pulsing appeared at a lower power than that with CW, which may be due to the increase in the acoustic amplitude with pulsing by the suppression of large degassing bubble generation due to the pulse-off time, compared with that for CW. As the power increased, the intensity with CW increased; the intensity with CW in the presence of particles (for both 0.6 and 1.2 g/L) at 43 W was higher than the maximum with pulsing in the presence of 0.6 g/L particles at 13 W. Therefore, it should be noted that at higher power with particle addition, the SCL intensity with CW can be higher than that with pulsing. At 28 to 30 W, the intensity with pulsing in the absence of particles approached almost zero, while the intensity with pulsing in the presence of particles and for all cases of CW did not change.

Thus, particle addition can lead to an enhancement of the sonochemical-reaction efficiency under both pulsing operation and CW.

Particle addition has another role, in that it suppresses the liquid-surface vibration which prevents high efficiency of sonochemical reactions. At high acoustic amplitude, liquid-surface vibration occurs frequently by the action of acoustic radiation force, at which time the establishment of the resonant standing wave field is disturbed and the sonochemical reaction efficiency is decreased. We previously reported that by weakening the vibration with a hydrophobic powder covering the liquid surface at relatively high acoustic amplitude, SCL intensity higher than that in the absence of the covering could be obtained.[10] Recently, the authors investigated the liquid-surface vibration under CW or pulsed ultrasound through optical measurements of the surface displacement with a laser displacement sensor and showed that the addition of particles inhibits the increase in the surface vibration amplitude both for CW and pulsed sonication.[15]

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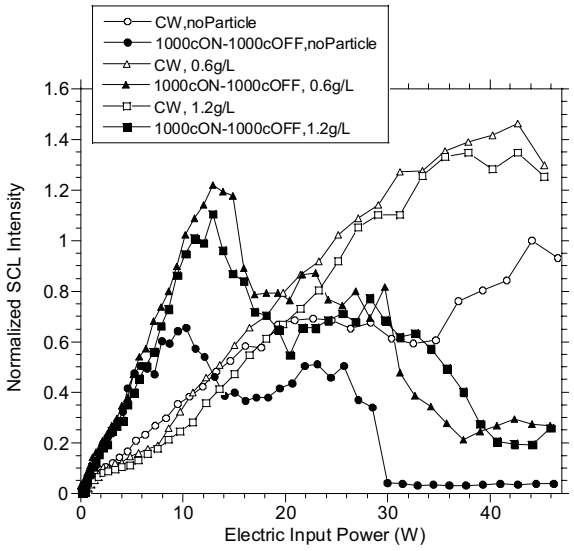


**Figure 1**. Dependence of the measured SCL intensity on the power applied to the transducer. (taken from ref 15 and reproduced by permission of © 2010 American Chemical Society)