

# On the influence of liquid-surface vibration on sonochemiluminescence

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## ABSTRACT

The influence of surface vibrations on the intensity of sonochemiluminescence (SCL) produced by pulsed ultrasound with a frequency of 151 kHz is investigated through optical measurement of the vibration amplitude. Pulsed ultrasound inhibits the generation of large degassing bubbles that restrict the efficient spatial region for sonochemical reaction. The vibration amplitude of the liquid surface becomes gradually significant with pulsed ultrasound as the power applied to the transducer increases. At this time the SCL intensity increases and then decreases after displaying a peak. The SCL intensity during high-amplitude pulsing becomes almost zero if the distance between the highest position of the liquid surface and its position in the absence of ultrasound becomes close to or larger than one quarter of the ultrasound wavelength. This condition for the liquid surface vibration provides a limit for establishment of a resonant standing wave that is effective for sonochemical reaction.

## INTRODUCTION

The ultrasonic cavitation bubble can provide the extreme conditions at the interior, including temperatures of several thousand Kelvin, pressures of several hundred atmospheres, and heating and cooling rates greater than  $10^9$  K/s [1,2]. Under the condition, water is easily decomposed and oxidants such as hydroxyl radicals, hydrogen peroxide and ozone are created [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 acoustic bubbles are referred to as sonochemical reactions [2,5].

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 was obtained [6].

Pulsing operation of ultrasound is useful to obtain high efficient sonochemical reaction [7-9]. The pulsing operation inhibits the generation of degassing large bubbles that restrict the efficient spatial region for sonochemical reaction. The authors have shown that the residual sound pressure during pulse-off time as well as the spatial enlargement contributes to the enhancement in the sonochemical-reaction effi-

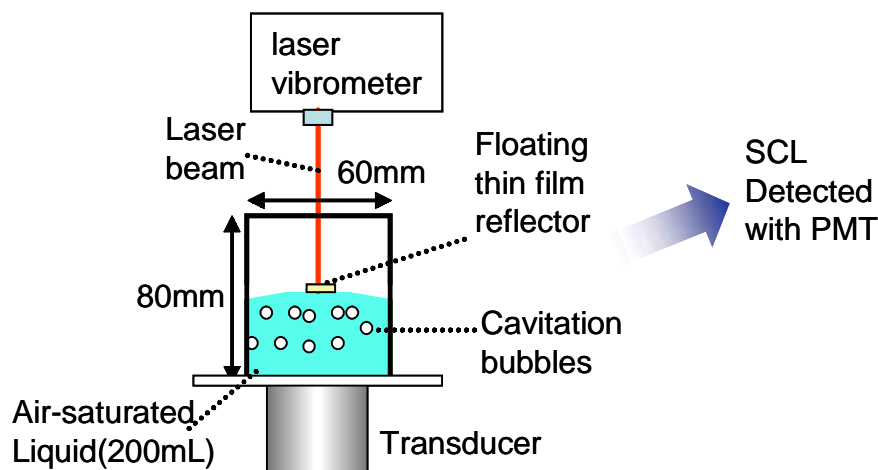


Figure 1. Experimental apparatus.

ciency[10].

To the best of our knowledge, a quantitative study on the effect of the liquid-surface vibration amplitude on the sonochemical-reaction efficiency has not yet been reported. In this study, the influence of liquid surface vibration on the SCL intensity by pulsed ultrasound was investigated through optical measurements of the vibration amplitude.

## EXPERIMENTAL DETAILS

Figure 1 shows an experimental apparatus. 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 and a blue fluorescence when intense ultrasound passes through the luminol solution [11]. 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 a larger vessel to a liquid height of 64 mm (the liquid volume was 200 mL). The liquid temperature was 25 °C. The intensity of SCL from the air-saturated liquid in the large vessel was measured with a photomultiplier tube (PMT; Hamamatsu, R928). Data were recorded using a computer (NEC, PC-9821 Xc16) through a digital multimeter (Advantest, TR6847) that reads the output voltage from the PMT. The sonication time for each set of data was 1 min.

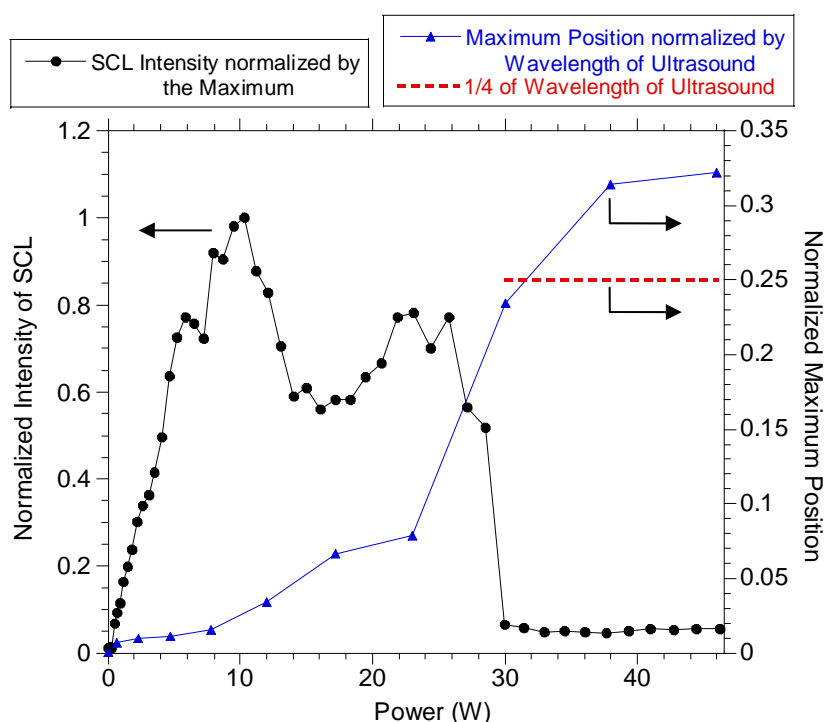
In order to evaluate the amplitude of liquid surface vibration, the time variation in the longitudinal position of the liquid was measured with a laser displacement sensor (OPTEx-FA, CD4), where a laser beam was introduced to a plastic thin film reflector (0.4 mm thickness, 9 mm OD) floating on the liquid surface. The horizontal position of the film was limited loosely within a region surrounded by 6 thin wires in order not to disturb the motion of the film and the liquid. The waveform was observed with an oscilloscope (Yokogawa, DL1540C) to determine the vibration amplitude and the position of the liquid surface.

## RESULTS AND DISCUSSION

A relationship was investigated between SCL intensity at pulsing (repetition of 1000 cycles ON and 1000 cycles OFF) normalized by the maximum intensity and measured maximum position of liquid surface as a function of applied power to the transducer (Figure 2). As the power increased, the SCL increased and showed the maximum. At higher power, the intensity decreased suddenly up to around zero. A longitudinal vibration of the liquid surface became remarkable and the maximum position gradually increased as the applied power increased.

It is noteworthy that at that time when the sudden decrease in SCL intensity in the above occurred, the maximum position of the liquid surface was close to the forth of the wavelength of the ultrasound. This condition for the liquid surface vibration provides a limit for the establishment of a resonant standing wave that is effective for sonochemical reaction. A pressure node is present in the air-liquid interface near the side wall; however, the above condition provides a pressure antinode at the central region in the same horizontal plane. This means that it is impossible to establish a stable horizontal stripe specific to the resonant standing wave.

At further power, the maximum position was over the forth of the wavelength of the ultrasound. At this time SCL inten-



**Figure 2.** Relationship between SCL intensity and the maximum longitudinal position of liquid surface as a function of applied power to the transducer.

sity remained around zero.

Liquid surface vibration is possible to show modal motion by a superposition of surface wave propagating from the center of the surface to side wall and the reflective wave [12]. Hashimoto and Sudo [12] have shown from an observation of ripple stripes on the surface that subharmonic wave is excited through an experiment of longitudinal vibration of a cylindrical vessel in which water is filled up to a certain height in a range of 1 to 3500Hz in the driving frequency, although no ultrasound is applied into the liquid. Modal analysis for liquid surface vibration of the present study is complex because of the presence of ultrasound or particles and accordingly, left to future study.

## CONCLUSION

The quantitative measurement of the amplitude and displacement of liquid surface vibration clarified that under the condition where the highest position of liquid surface becomes close to or overcomes the fourth of wavelength of an ultrasound, a remarkable decrease in sonochemical reaction efficiency occurs.

## ACKNOWLEDGMENT

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## REFERENCES

- 1 Leighton, T. G. "5. Effects and Mechanisms," in *The Acoustic Bubble*, Academic, London, 1996.
- 2 Suslick, K. S. *Science* **247**, 1439 (1990)
- 3 Yasui, K.; Tuziuti, T.; Sivakumar, M.; Iida, Y. *J. Chem. Phys.* **122**, 224706 (2005)
- 4 Walton, A.J.; Reynolds, G.T. *Adv. Phys.* **33**, 595 (1984)
- 5 Mason, T.J. "1. An introduction to the uses of power ultrasound in chemistry," in *Sonochemistry*, Oxford University Press: New York, 1999.
- 6 Tuziuti, T.; Yasui, K.; Kozuka, T.; Towata, A.; Iida, Y. *J. Phys. Chem. A* **111**, 12093 (2007)
- 7 Flynn, H. G.; Church, C. C.; *J. Acoust. Soc. Am.* **76**, 505 (1984)
- 8 Henglein, A.; Ulrich, R.; Lilie, J. *J. Am. Chem. Soc.* **111**, 1974 (1989)
- 9 Casadonte, D. J.; Flores, M.; Pétrier, C. *Ultrason. Sonochem.* **12**, 147 (2005)
- 10 Tuziuti, T.; Yasui, K.; Lee, J.; Kozuka, T.; Towata, A.; Iida, Y. *J. Phys. Chem. A* **112**, 4875 (2008)
- 11 Pétrier, C.; Lamy, M-F.; Francony, A.; Benahcene, A.; David, B.; Renaudin, V.; Gondrexon, N. *J. Phys. Chem. A* **98**, 10514 (1994)
- 12 Hashimoto, H.; Sudo, S. *Transact. Jpn. Soc. Mech. Eng. B* **49**, 1841 (1983)