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Effect of Superposition of Ultrasonic Fields on Sonochemical Reaction Rate

ソノケミカル反応速度に及ぼす超音波場重ね合わせの影響

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1. Introduction

In order to apply the sonochemical methods to on-site wastewater treatment, the enhancement of sonochemical reaction is necessary. It has been reported that the superposition of ultrasonic fields[1] increased the sonochemical reaction rate. In this study, the effect of superposition place of ultrasonic fields on the sonochemical reaction is examined by experiment and numerical simulation.

2. Experiment

Fig. 1 shows the photograph of the experimental apparatus. The reactor was made of transparent acrylic resin. Four PZT ultrasonic transducers were attached at the bottom, lower, middle and upper of the reactor. The ultrasonic frequency was 486 kHz. The effective electric power applied to each transducer was 50 W. The sample was the potassium iodide aqueous solution. The initial concentration and volume were 0.1 mol/L and 4.5 L, respectively. The concentration of triiodide ion, I_3^- produced after 30 min sonication was measured by UV spectroscopy. The sonochemical reaction area was visualized by using sonochemical luminescence of luminol solution. The luminol image was recorded for the exposure time of 30 s with digital camera in dark condition.

3. Numerical simulation

The theory of acoustic field is based on inhomogeneous Helmholtz equation. The acoustic pressure area above the cavitation threshold was calculated to estimate the reaction area. The liquid velocity was calculated from the volume force due to acoustic pressure distributions by using the momentum transport equation. The detail of numerical simulation is described elsewhere.[2] The acoustic absorption coefficient of sample was set at 1 m^{-1}. The calculations were carried out by COMSOL Multiphysics™ (COMSOL AB, Stockholm, Sweden). To save the calculation time and memory, the reactor in two dimensions was calculated.

4. Results and discussion

Table I shows the I_3^- concentration and the enhancement amount for the irradiations from single and dual transducers. The enhancement amount was obtained from the concentration ratio as follows.

$$E = \frac{C_D}{C_B + C_S} - 1 \qquad (1)$$

where the subscripts D, B and S denote ultrasound irradiations from dual, bottom and, side (lower, middle and upper) transducers, respectively.

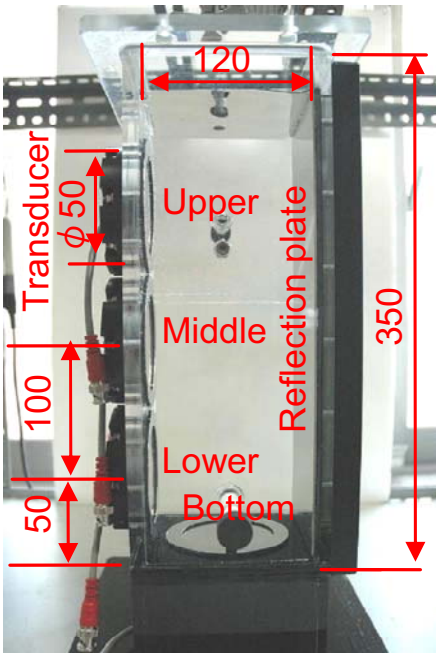

Fig. 1 Photograph of experimental apparatus (unit: mm)

Table I I_3^- concentration and enhancement amount for irradiations from single and dual transducer.

	Single C (mmol/m^3)	Dual* C_D (mmol/m^3)	Enhancement amount E (%)
Bottom	7.9	—	—
Lower	11.3	21.8	13.5
Middle	10.9	19.0	1.1
Upper	11.1	22.8	20.6

* For all dual conditions, bottom transducer was used.

In the case of single transducer, the I_3^- concentration for side transducer is higher than that for bottom transducer. For the case of dual transducer, the concentration for upper and bottom transducers is highest.

Fig. 2 shows luminol images for irradiations from upper (a), bottom (b) and dual transducers(c). The bright area is sonochemical luminescence, that is, reaction area. In the case of upper transducer, many stripes of luminescence are observed from the left-hand side wall to the right-hand wall. On the other hand, the case of bottom transducer, the reaction area is localized near liquid surface above transducer. For the case of dual transducers, the reaction area becomes extensive and intensive compared with that for single transducer.

Fig. 3 shows the acoustic pressure area above cavitation threshold by numerical simulations. In the case of upper transducer, the acoustic pressure area above cavitation threshold is similar to the reaction area of luminol image. For the case of dual transducer, the acoustic pressure area above cavitation threshold is enlarged and enhanced. This is because the pressure amplitude in reactor is increased due to the interference of dual ultrasonic fields. The increase of pressure amplitude induces the implosions of transient cavitation to become more violent. Hence, it is considered that the reaction becomes extensive and intensive. On the other hand, the case of bottom transducer, the acoustic amplitude was low and the acoustic pressure area above cavitation threshold is different from the reaction area of luminol image.

The liquid velocity distribution for irradiation from bottom transducer by numerical simulation is shown in **Fig. 4**. The relative high velocity area is assembled in the centre of the transducer and orientated towards acoustic propagation direction. The liquid velocity increases as the sample position becomes higher since the flow acceleration length is long. It is known that the liquid flow enhances the sonochemical reaction due to the supply of reactant and cavity nuclei to the reaction area and the prevention of cavity aggregation in the standing wave. Assuming that the cavity generates at all places with acoustic pressure above cavitation threshold and does not disappear in sample, the cavity density becomes highest near liquid surface by upward flow of cavity. It is considered that the reaction area of luminol image for bottom transducer is related to the acoustic pressure, liquid flow and cavity density. Among three side transducers, since the liquid flow in the acoustic field from upper transducer is high and wide, it is thought that the reaction rate becomes highest.

References

1. Yasuda et al.: Ultrason. Sonochem. **14**(2007) 699.
2. Xu et al.: Ultrason. Sonochem., (2012) in press.

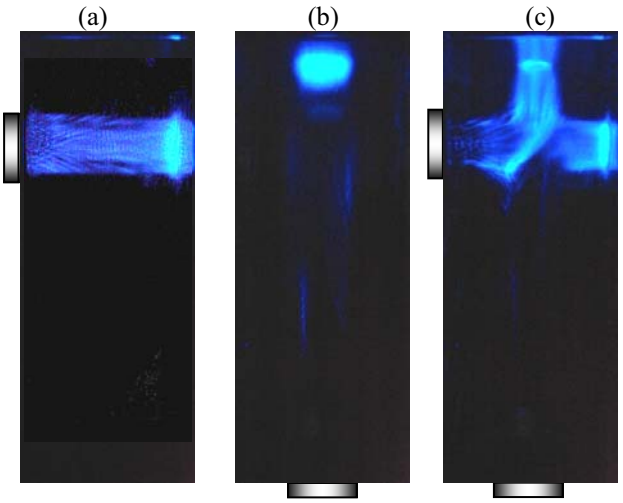

Fig. 2 Luminol images for irradiations from upper and bottom transducers.

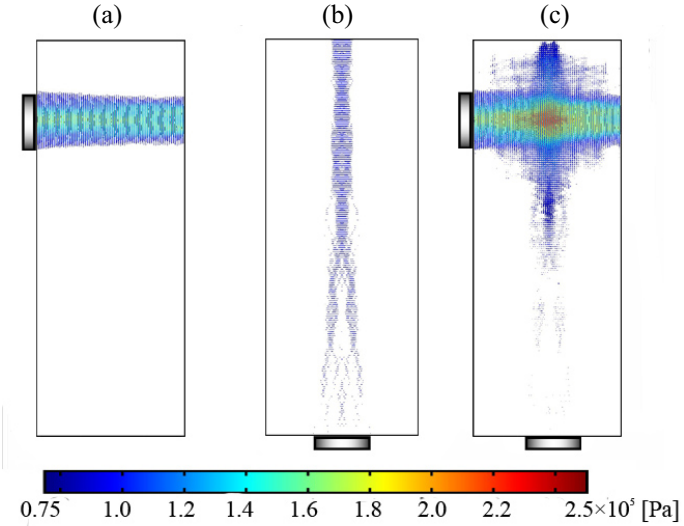

Fig. 3 Acoustic pressure area above cavitation threshold for irradiation from bottom and upper transducers by numerical simulations.

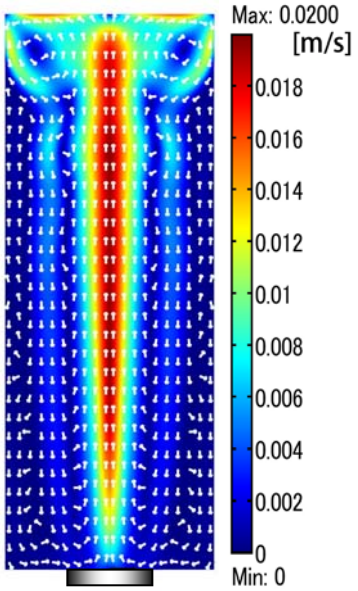

Fig. 4 Liquid velocity distribution for irradiation from bottom transducer by numerical simulation.