

Enhancement of sonochemical reaction by dual-pulse ultrasound

デュアルパルス型超音波による超音波化学反応の促進

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1. Introduction

Application of ultrasound in chemical reaction, i.e., sonochemistry, has been extensively studied for various field. In the procedure of degrading refractory materials in wastewater, the sonochemical method is easy to operate and hardly to make harmful by-products comparing with other methods. However, reaction performance is still not high enough for practical use. For the purpose of enhancing reaction performance, various methods, such as utilizing pulsed wave¹⁾ and superposition of ultrasonic field²⁾ have been reported.

In the present study, dual-pulse has been investigated and superposition of pulsed wave in sonochemical reaction have been performed experimentally in various operation conditions.

2. Experimental

The experimental apparatus of dual-pulse sonochemical reaction is described as **Fig.1**. The reactor was made of transparent acrylic resin. The dimensions were 120 mm in length, 120 mm in width and 350 mm in height. Two transducers were stuck on the bottom and side wall of rectangular vessel and operated at 492 kHz. A signal generator was used to modulate pulsed on time ranging from 0.001 s to 0.400 s. The generator output was connected to power amplifier so that the pulse signal was amplified to drive transducers. The digitizing oscilloscope was used to detect the pulse signal received by the transducer. In the case of pulsed wave, the input power of transducer was 50 W. The ratio of pulse on time to pulse off time was 1. For the case of continuous wave, the input power to transducer was 25 W in order to become the same energy as continuous wave. The 3 L of potassium iodide (KI) solution was used and the concentration was fixed at 0.1 mol/L. The irradiation time was 30 min. The I_3^- ion concentration was measured by UV spectrometer.

3. Results and discussion

Fig.2 shows the effect of pulse on time (t_{on}) on I_3^- ion concentration (C) for irradiation from side transducer. The concentration for continuous wave

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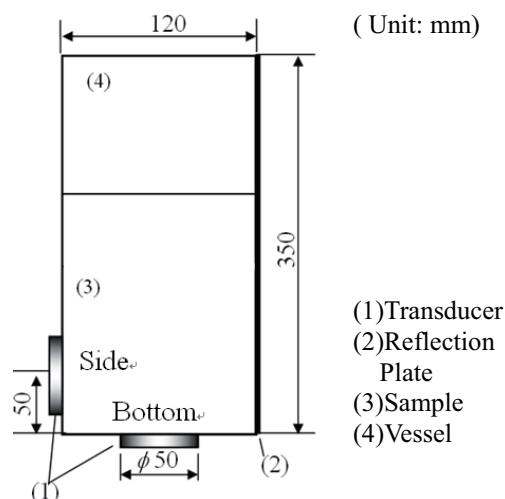


Fig.1 Outline of experimental apparatus

is also represented in **Fig.2** as horizontal line. In the case of $t_{on} = 0.001$ s, the I_3^- ion concentration for pulsed wave is lower than that for continuous wave. It is considered that the formation of transient cavity site requires the certain energy per one pulsed wave. For $t_{on} > 0.01$ s, the I_3^- ion concentration for pulsed wave becomes higher than that for continuous wave. This is because the power-modulated pulsed ultrasound increases cavity nuclei, decreases large stable cavity and enhances sound pressure due to reverberation in reactor. After the I_3^- ion concentration reaches a maximum at $t_{on} = 0.2$ s, it decreases gradually by the increasing of pulse off time. It is considered that long pulse off time decreases the residual sound pressure due to reverberation in reaction field.

Fig.3 shows the effect of pulse on time on I_3^- ion concentration for irradiation from bottom transducer. The concentration for continuous wave is also represented in **Fig.3** as horizontal line. In the case of $t_{on} = 0.001$ s, the I_3^- ion concentration for pulsed wave is lower than that for continuous wave. For $t_{on} > 0.01$ s, the I_3^- ion concentration for pulsed wave is higher than that for continuous wave. Compared with pulsed wave irradiation from side transducer (**Fig.2**), the increasement of I_3^- ion concentration for pulsed wave is small. When the power-modulated pulsed ultrasound was utilized, a fountain on the liquid surface was observed. From these results it is considered that the fountain on liquid surface distorts the reflective wave and

decreases the residual sound pressure due to reverberation in reaction field and the number of standing wave.

The I_3^- ion concentration and enhancement amount for dual transducer are shown in **Tab.1**. The pulse on time was 0.24 s. The pulsed wave irradiation increases the sonochemical reaction performance as previously stated in single transducer case. When dual-pulse ultrasound was utilized, the reaction performance was higher than those for any other irradiation conditions.

In order to consider the mechanism of reaction enhancement, the calculation of enhancement amount is separated into two parts. The dual transducer enhancement (E_{dual}) and pulse wave enhancement (E_{pw}) are defined as follows,

$$E_{dual} = C(S\&B) / [C(S) + C(B)] - 1 \quad (2)$$

$$E_{pw} = C(S\&B) / C(S_{cw}\&B_{cw}) - 1 \quad (3)$$

where S, B and S&B are ultrasound irradiations from side, bottom and both transducers, respectively. The subscripts of CW and PW are continuous and pulsed waves, respectively. The enhancements of dual-transducer are approximately at 0.1. This is because the sonochemical reaction fields become more intensive and extensive²⁾. The pulsed wave enhancement is responsive to the position of transducer. In the case of pulsed wave irradiation for side transducer, the amount of pulse wave enhancement is much higher than that for bottom transducer. Especially, the greatest value of pulse enhancement can be obtained at dual-pulse wave.

The different dual-pulse mode was also experimentally studied. The trigger is modulated, so that two kinds of superposition mode in the center of vessel. One is superposition of pulsed wave on cycle and that on cycle (same phase). The other is superposition of pulsed wave on cycle and that off cycle (reverse phase). As shown in **Tab.2**, the superposition mode also has influence on reaction performance. The superposition of pulse waves with same phase generates much I_3^- ions compared with reverse phase. This is because superposition of pulse waves with same phase creates high ultrasonic pressure fields.

Conclusion

The dual-pulse ultrasound enhances largely sonochemical reaction performance.

References

- 1) D. J. Casadonte Jr. *et al*: Ultrason. Sonochem., **12** (2005) 147.
- 2) K. Yasuda *et al*.: Ultrason. Sonochem., **14** (2007) 699.

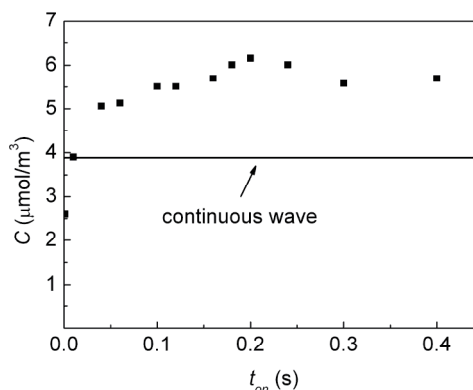


Fig.2 Effect of pulsed on time on I_3^- ion concentration for irradiation from side transducer.

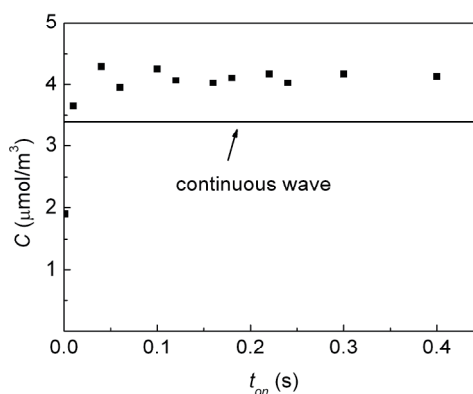


Fig.3 Effect of pulsed on time on I_3^- ion concentration for irradiation from bottom transducer.

Tab.1 I_3^- ion concentration and enhancement amount for dual transducer. ($t_{on} = 0.24$ s)

| | C ($\mu\text{mol}/\text{m}^3$) | E_{dual} (-) | E_{pw} (-) |
|---------------------|------------------------------------|----------------|--------------|
| S_{cw} & B_{cw} | 7.83 | 0.079 | - |
| S_{cw} & B_{pw} | 8.63 | 0.091 | 0.102 |
| S_{pw} & B_{cw} | 10.65 | 0.085 | 0.359 |
| S_{pw} & B_{pw} | 11.60 | 0.130 | 0.481 |

Tab.2 I_3^- molecular concentration for different superposition mode. ($t_{on} = 0.24$ s)

| | C ($\mu\text{mol}/\text{m}^3$) |
|---------------|------------------------------------|
| Same phase | 11.60 |
| Reverse phase | 9.31 |