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Short communication

Development of a large sonochemical reactor at a high frequency

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Abstract

The large sonochemical reactor was developed by using 12 PZT transducers. The frequency was 500 kHz and the total effective electric power applied to transducers was 620 W. The sample and volume were aqueous solution of potassium iodide and 112 dm^3, respectively. The ultrasonic power dissipated into solutions was measured by a calorimetric method.

The energy conversion efficiency from electricity to ultrasound was 70%. When the liquid height was from 400 to 435 mm, the I_3^- production rate has a maximum value. The I_3^- production rate increased with increasing ultrasonic power. In the case of high ultrasonic power, the I_3^- production rate for transducers located at the side wall was higher than that at the bottom wall. The sonochemical efficiency for a large sonochemical reactor operated at 500 kHz was close in value to those for laboratory scale reactors at 500 kHz.

Keywords: Sonochemical reactor; Large scale; Liquid height; Transducer position; Sonochemical efficiency

1. Introduction

The ultrasound irradiation in liquids gives rise to cavitation, which causes various physical and chemical effects. The physical effects of ultrasonic cavitation have been already put to practical use in ultrasonic cleaner, homogenizer, and so on. Some large-sized ultrasonic cleaners and homogenizers have developed, whose volume was more than 100 dm^3.

In two-decade years, many researchers have investigated chemical effects of ultrasonic cavitation such as the degradation of chemical contaminants and synthesis of chemical compounds in solution [1–3]. Koda et al. [4] have shown that the generation rate of hydroxyl radical due to sonolysis of water has a maximum value in the frequency range from 200 to 600 kHz. However, the studies of chemical effects have been mostly conducted by using laboratory-scale sonochemical reactors, whose volume was less than 1 dm^3. In order to put the sonochemical processes of wastewater treatment and material processing in practical use, it is necessary to design and develop a large sonochemical reactor on a pilot-scale (more than 100 dm^3) or an industrial-scale (more than 1 m^3).

The large-scale sonochemical reactors have been developed for low frequency at 20–50 kHz [5–7]. Recently, some large-scale sonochemical reactors for high frequency at 200–600 kHz have been reported [8,9]. Gonze et al. [8] have developed the sonochemical reactor with the capacity of 28 dm^3. Their reactor was attached three 500 kHz transducers at the reactor bottom. Destaillants et al. [9] have used four transducers at 612 kHz and developed the sonochemical reactor with the volume of 7 dm^3. However, for practical use, the further increase of volume of sonochemical reactor is necessary. In order to develop the larger sonochemical reactor,

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Nomenclature

C_I	I_3^- concentration (mol dm^{-3})
C_{PUS}	specific heat at constant pressure of water (J kg^{-1} K^{-1})
E_{US}	ultrasonic energy (J)
h	distance between transducer units and reflection surface (m)
m_I	number of reacted molecules (mol)
M_{US}	solution mass (kg)
P_E	total effective electric power applied to two transducer units (W)
P_{US}	ultrasonic power dissipated into liquid (W)
r	I_3^- production rate (mol s^{-1})
SE	sonochemical efficiency (mol J^{-1})
t_{US}	ultrasonic irradiation time (s)
T	temperature (K)
V_I	sample volume (dm^3)

optimization of liquid height and transducer position is important.

In this study, a large sonochemical reactor with the capacity of 100 dm^3 operated at 500 kHz was developed. The effects of liquid height and transducer position on KI oxidation by hydroxyl radical were examined. The sonochemical efficiency of the large sonochemical reactors was compared with those of sonochemical reactors on a laboratory scale.

2. Experimental

2.1. Sonochemical reactor

Fig. 1 shows the outline of experimental apparatus. The rectangular vessel was made of stainless steel and the dimensions were 508 mm in length, 508 mm in width and 672 mm in height. The transducer unit was developed by Honda Electronic Co. Ltd. and the dimensions were 236 mm in length, 161 mm in width and 73 mm in height. One transducer unit consisted of six PZT transducers with 50 mm in the diameter. In this study, two transducer units were used. The driving frequency was 500 kHz. The total effective electric power applied to two transducer units was changed from 0 to 620 W which was measured by using an oscilloscope (TDS3012B, Tektronix Inc.) and a current probe (TCP202, Tektronix Inc.). In order to investigate the effect of reflection surface, two transducer units (12 PZT transducers) located at the vessel side (Fig. 1, positions 1 and 2) or the vessel bottom (Fig. 1, positions 3 and 4). In the case of two transducer units located at the vessel bottom, the liquid height, which was the distance between the transducer unit and the reflection surface, was changed from 300 to 500 mm (sample volume: 77–131 dm^3).

2.2. Quantification of chemical reaction

The oxidation reaction of the aqueous solution of potassium iodide (KI) was used in order to know the sonochemical efficiency of the reactor [4]. When ultrasound is irradiated into the aqueous solution of KI, I^- ions are oxidized to give I_2. When excess I^- ions are present in solutions, I_2 reacts with an excess I^- ion to form an I_3^- ion in the following equation:

$$I_2 + I^- \rightleftarrows I_3^- \quad (1)$$

The absorption peak of the I_3^- ion appeared at 355 nm (ε = 26,303 dm^3 mol^{-1} cm^{-1}). After the ultrasonic irradiation of 90 min, the liquid in the reactor was agitated so that the I_3^- concentration might be uniform in the reactor. The I_3^- concentration was measured by using a UV spectrometer (UV-1600, Shimadzu Co.). In this study, the KI concentration in the aqueous solution before ultrasonic irradiation was 0.1 mol dm^{-3}. The KI was special grade chemicals and purchased from Wako Pure Chemical Industries, Ltd. All samples were used without purification.

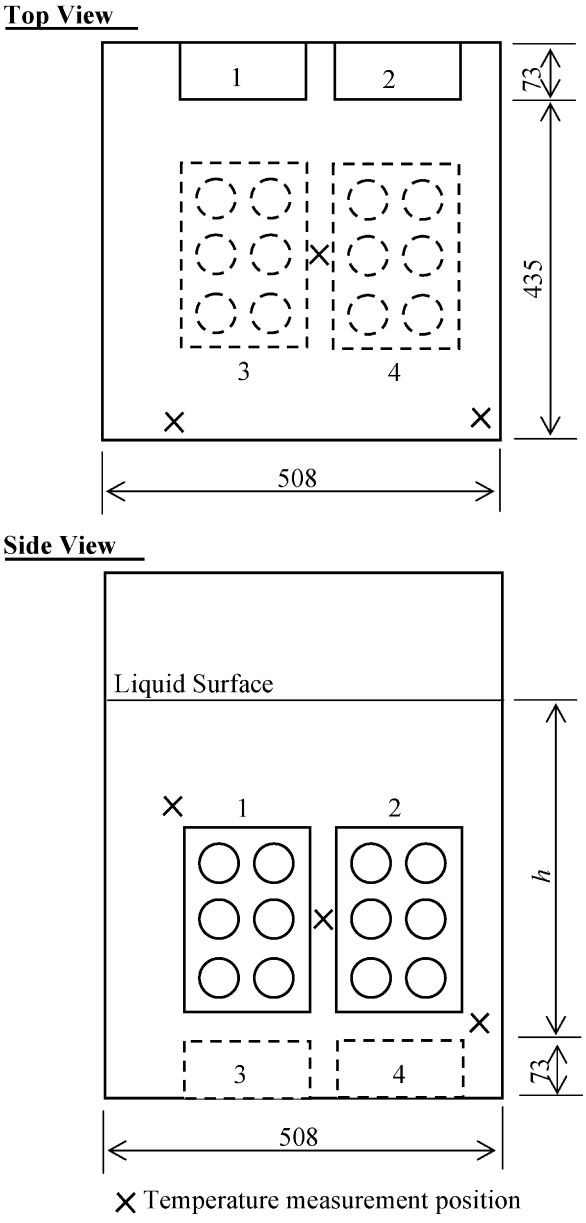

Fig. 1. Outline of experimental apparatus.

2.3. Quantification of ultrasonic power

The sonochemical reactor was equipped with three thermocouples (Copper and constantan, Omega Engineering Inc.) which are connected with a temperature recorder (NR-500, NR-TH08, Keyence Corp.) and PC. Three thermocouples were located at different positions as shown in Fig. 1. The thermocouples were used to measure temperature of the solution in the sonochemical reactor. The ultrasonic power dissipated into liquid, P_{US}, was calculated from the following equation:

$$P_{US} = M_{US} C_{PUS} \frac{\Delta T}{\Delta t_{US}} \quad (2)$$

where M_{US}, C_{PUS} and $\Delta T/\Delta t_{US}$ denote the solution mass, the specific heat at constant pressure and the rate of temperature increase, respectively. The ultrasonic power was estimated from the average value of initial temperature rises at three positions.

3. Results and discussion

Fig. 2 shows the plot of ultrasonic power against the electric power for transducer units located at positions 1 and 2, and the distance between transducer units and reflection surface was 435 mm. The ultrasonic power increases linearly with the electric power. The slope in Fig. 2 is around 0.7 and this result indicates that the energy conversion efficiency of this large sonochemical reactor is about 70%. The energy conversion efficiency of sonochemical reactors reported by Destaillants et al. was about 40% [9]. The reactor investigated here presents high-energy conversion efficiency. This is because we optimized the ratio of transducer diameter to transducer thickness and the thickness of vibration plates attached with transducer. The energy conversion efficiency was little dependent on the position of transducer units and liquid height.

Fig. 3 shows the effect of liquid height on I_3^- production rate for the transducer units located at positions 3 and 4. The electric power for transducer units was 520 W. The I_3^- production rate has a maximum value when the liquid height was from 400 to 435 mm.

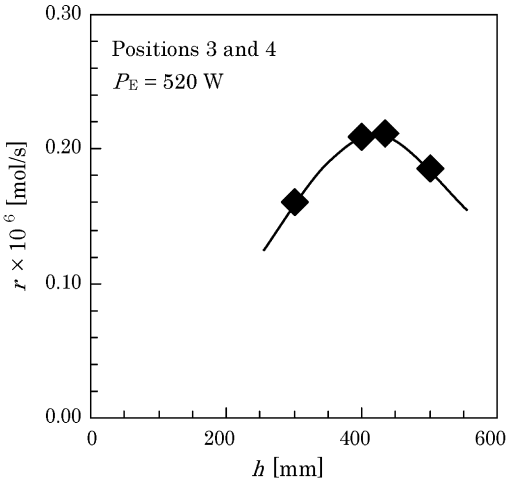

Fig. 3. Effect of liquid height on I_3^- production rate for transducer units located at positions 3 and 4.

Fig. 4 shows the effect of electric power on I_3^- production rate for different positions of transducer units. The distance between transducer units and reflection surface was fixed at 435 mm, which was determined from data in Fig. 3. The I_3^- production rate increases with increasing electric power. In the case of electric power at 620 W, the I_3^- production rate for transducer units located at positions 1 and 2 is larger than that for transducer units located at positions 3 and 4. The rises of liquid surface at just above 12 PZT transducers were observed when the transducer units were located at positions 3 and 4, and applied electric power at 620 W. The height of these rises of liquid surface was about 2 mm. Recently, we have [11] reported that the standing wave was formed near the liquid surface and the chemical reaction effectively proceeded. From these results, when the transducer units were located at positions 3 and 4, were operated with the applied electric power of 620 W, it is considered that the rise of liquid surface may disturb the formation of standing wave

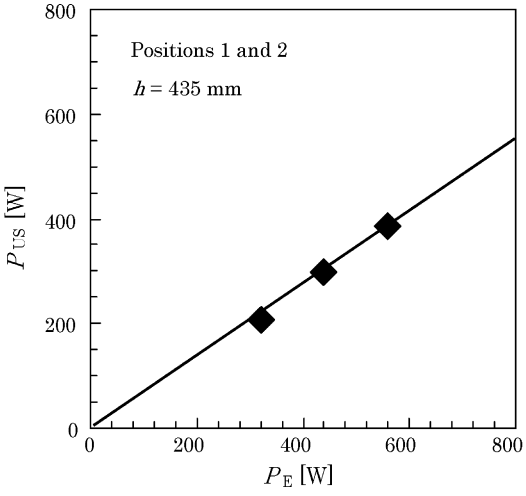

Fig. 2. Plot of ultrasonic power against electric power for transducer units located at positions 1 and 2.

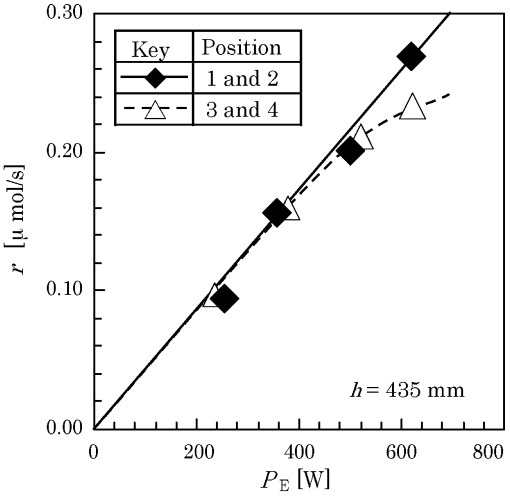

Fig. 4. Effect of electric power on I_3^- production rate for different positions of transducer units.

and consequently the sonochemical reaction rate for transducer position with a free end is lower than that with a fixed end.

The sonochemical efficiency is used to evaluate the performance of sonochemical reactors. The sonochemical efficiency SE – the ratio of the number of reacted molecules m_I to the ultrasound energy E_{US} – is given by [4]:

$$SE = \frac{m_I}{E_{US}} = \frac{C_I}{P_{US} t_{US}/V_I} = \frac{r}{P_{US}} \quad (3)$$

where C_I and r denote the I_3^- concentration and I_3^- production rate of the 0.1 mol dm^{-3} KI solution, respectively. And V_I and P_{US} are sample volume and ultrasonic power, respectively. The *SE* value corresponded to the I_3^- ion concentration normalised by the ultrasonic energy absorbed by unit-volume solutions for the irradiation time t_{US}. When the transducer units are located at positions 1 and 2, and the distance between transducer units and reflection surface is 435 mm, the average value of SE is 6.3×10^{-10} mol J^{-1}. Koda et al. [4] have reported that the average value of SE for laboratory scale reactors (under 0.2 dm^3 in volume) is 7.1×10^{-10} mol J^{-1}. The sonochemical efficiency for a large sonochemical reactor operated at 500 kHz is close in value to those for laboratory scale reactors at 500 kHz. Recently, we [10] have investigated the effect of liquid height on the SE value for a cylindrical sonochemical reactor. The reactor was made of a cylindrical acrylic pipe with 70 mm in diameter and a transducer with 490 kHz was attached at the bottom of the acrylic pipe. The SE values for the cylindrical reactor had two maximum peaks at 29 and 400 mm, and the SE values at 29 and 400 mm were 7.1×10^{-10} and 6.2×10^{-10} mol J^{-1}, respectively. From these results, it is considered that the sonochemical reaction performances for laboratory reactors and this large scale reactor correspond to those for the cylindrical reactor at 29 and 400 mm in heights, respectively. That is, the SE values for 500 kHz depend on liquid height but that values little depend on reactor width and length.

Fig. 5 shows photographs of sonochemical luminescence for different positions of transducer units. The distance between transducer units and reflection surface was fixed at 435 mm and the electric power for transducer units was 520 W. The photographs were shoot from the reactor top. In both positions of transducer units, the 12 high-intensity sonochemical luminescences from 12 transducers were observed near the reflection surface and these sonochemical reaction fields were independent each other. This is because that the ultrasound propagation at 500 kHz has high directivity and the sonochemical reaction fields little spread in a direction perpendicular to ultrasonic propagation.

Our results show that the liquid height is one of very important factors to optimize the sonochemical reaction efficiency to design a large scale sonochemical reactor for high frequency. It is considered that the formation of standing wave is one of important factor to develop the sonochemical reactors since the liquid height or volume influence on the formation of standing wave. Further works will be continued to discuss optimization of large-scaled sonoreactor in detail.

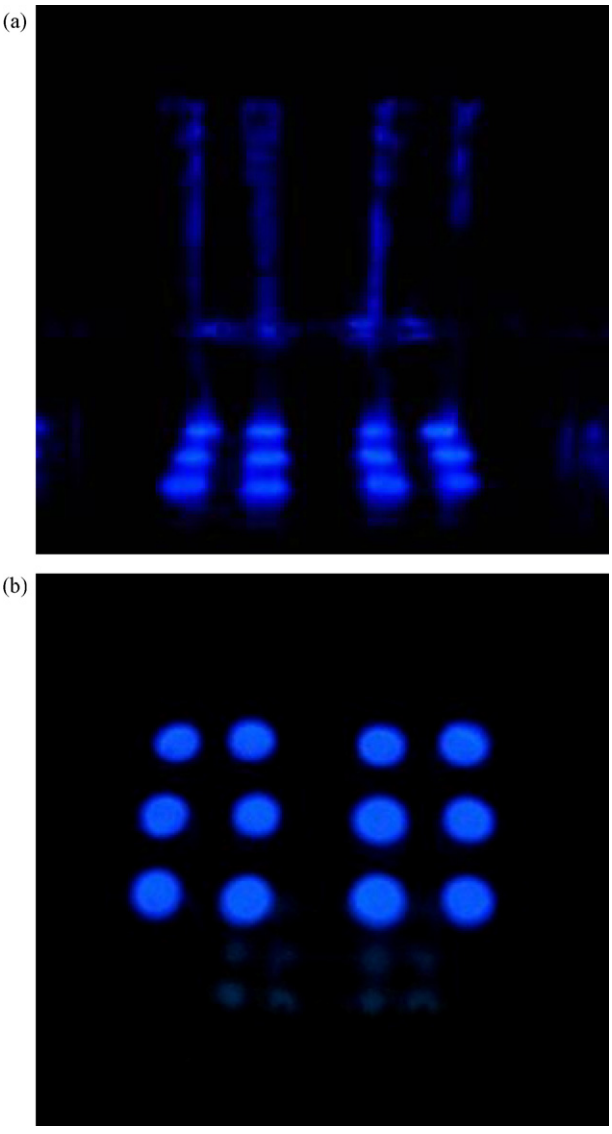

Fig. 5. Photographs of sonochemical luminescence: (a) transducer units located at positions 1 and 2; (b) 3 and 4.

4. Conclusions

Large sonochemical reactor operated at 500 kHz was developed. The effects of liquid height and transducer position on KI oxidation rate were examined. The following results were found:

1. The energy conversion efficiency from electricity to ultrasound was about 70%.
2. The I_3^- production rate had maximum value in the liquid height region from 400 to 435 mm.
3. In the case of high ultrasonic power, the I_3^- production rate for transducers located at the side wall was higher than that at the bottom wall.
4. The sonochemical efficiency for a large sonochemical reactor was close in value to those for laboratory scale reactors.

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