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Effects of Pressure, Temperature and Frequency on the Sonochemical Reaction in a Flow-Type Reactor

流通式反応器のソノケミカル反応に及ぼす圧力，温度，周波数の影響

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1. Introduction

When a liquid is irradiated by a high power ultrasound, local spots with extreme high pressures and temperatures “so-called hotspots” are formed in the liquid via a sequence of process of formation, growth and collapse of cavitation bubbles. Hence, physical and chemical effects are induced in a liquid by ultrasonication. Recently, high power ultrasonic technique has attracted attention as one of applicable technologies to wastewater treatment, synthesis of chemicals and preparation of materials.

On designing a sonochemical reactor, it is significant to know how a variety of physical or chemical parameters influence acoustic power or cavitation intensity[1-7]. Frequency and hydrostatic pressure is significant factors for determining the behaviors (threshold, growth and collapse etc.) of the cavitation bubbles. Since temperature affects the physical properties of liquid, the behaviors of the cavitation bubbles would depend on temperature of the bulk liquid or solution. In addition, pressure and temperature are fundamental factors affecting chemical reaction rates and chemical equilibrium.

In the present study, the effects of pressure temperature and frequency on sonochemical reaction were investigated using a flow-type sonochemical reactor equipped with PZT transducers which effectively oscillates at the frequencies of 500 kHz and 1 MHz. Potassium iodide dosimetry was used in order to evaluate the effects of the pressure, temperature and frequency on the sonochemical reaction rate.

2. Experimental

Fig. 1 shows the entire experimental setup. The reactor of a stainless-steel with an effective volumetric capacity of ca. 2.75× 10^{-2} dm^3 was used. The frequency range of ultrasound employed in the present work was 500-1000 kHz. The solution pressurized by a pump (PU-2086, Jasco Corp.) was fed into the sonochemical reactor at the flow rate of 10^{-3} $dm^3 \cdot min^{-1}$. The temperature was controlled using a water bath or a hot plate.The pressure was controlled by a back pressure regulator (TESCOM Corp.). All the experiments were carried out at the frequency determined on the basis of the frequency-transducer impedance curve which was obtained under each pressure and temperature condition. The ultrasound power, P_{US} [W], was estimated by calorimetry.

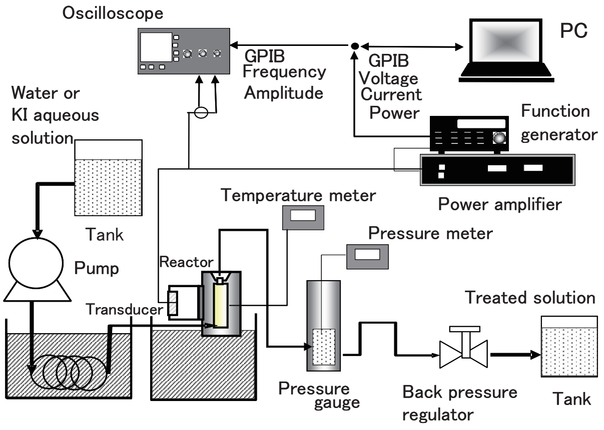

Fig. 1 A schematic diagram of experimental set-up

The oxidation reaction of KI in aqueous solution was used in order to evaluate the sonochemical effect in the reactor[8-9]. The ion-exchange water saturated with oxygen in the atmosphere at 298K was used to prepare the aqueous solution of KI with concentration 0.1 $mol \cdot dm^{-3}$. After ultrasonication, the absorption peak of the I_3^- ion at 355 nm (ε = 26,303 $dm^3 \cdot mol^{-1} \cdot cm^{-1}$) was measured by a visible-ultraviolet spectrophotometer (V-630, Jasco Corp.). KI oxidation under ultrasonication was the zero-order reaction and the oxidation reaction rate constant k_e [$mol \cdot dm^{-3} \cdot s^{-1}$], was calculated using the following equation:

$$k_e = C/\tau \ ,$$

Y. Noiri: noiri.yousuke@a.mbox.nagoya-u.ac.jp where C [mol·dm^{-3}] and τ [s] denote the concentration of I_3^- at the outlet of a reactor and space time, respectively.

3. Results and Discussion

Fig. 2 shows the pressure dependence of k_e/P_{US} in the temperature range of 293 to 373 K at the frequency of ca. 500 kHz. At the constant temperature of 293, 313 or 333 K, the k_e/P_{US} decreases with an increase in the pressure and no KI oxidation reaction proceeds above 0.1 MPa, probably due to partially suppression of the formation/growth of cavitation bubbles which are seeds required to form hotspots. At 353 and 373 K, on the other hand, the k_e/P_{US} increases with the pressure until reaching weakly peaks at 0.05 and 0.2 MPa, respectively. It appears that, since vapor pressure of water solvent is higher at the higher temperature, cavitation bubbles are moderately formed even under higher gauge pressure conditions and chemical effect is induced via explosive collapse of the bubbles.

Fig. 3 shows the pressure dependence of k_e/P_{US} in the temperature range of 293 to 373 K at the frequency of ca. 1 MHz. At the constant temperature of 313, 333, 353 or 373 K, the k_e/P_{US} increases with the pressure until reaching its maximum value. In the case of 1 MHz as compared with 500 kHz under the same temperature condition, the k_e/P_{US} reaches its maximum value at the higher pressure. In the present work, the most effective sonochemical reaction was achieved under the operating condition of 0.1 MPa, 333 K and 1 MHz.

4. Conclusions

In the present study, the effects of pressure, temperature and frequency on sonochemical reaction rate on were investigated. The results indicate that it is important to optimize the relationships among operating pressure, temperature and frequency conditions in order to achieve effective sonochemical reaction.

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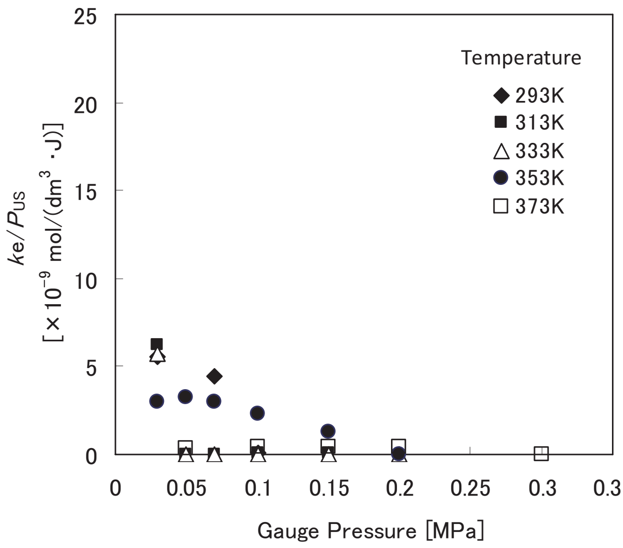

Fig. 2 Pressure dependence of k_e/P_{US} in the temperature range of 293 to 373 K at the frequency of ca. 500 kHz.

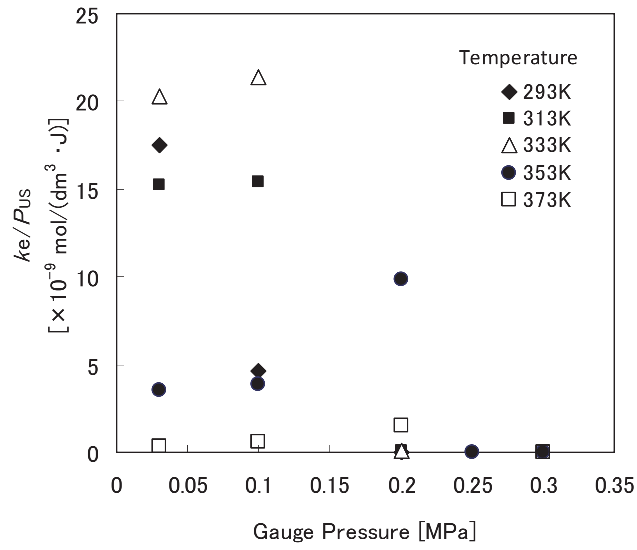

Fig. 3 Pressure dependence of k_e/P_{US} in the temperature range of 293 to 373 K at the frequency of ca. 1MHz.