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Proceedings of Symposium on Ultrasonic Electronics, Vol. 30 (2009) pp. 135-136
18-20 November, 2009

Effects of Pressure and Temperature on the Sonochemical Reaction in a Flow-Type Sonochemical Reactor
流通式反応器のソノケミカル反応に及ぼす圧力，温度の影響

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1. Introduction

The growth and collapse processes of cavitation bubbles under ultrasonication lead to generation of enormous pressures and temperatures, so that physical and chemical effects emerge at the local locations in the liquid in a reactor. The chemical reaction processes utilizing these effects are called sonochemical processes. The applicability of the sonochemical process to the decomposition of pollutants and synthesis of chemical compounds in solution has been demonstrated using a batch-type sonochemical reactor[1)]. On designing the sonochemical reactor, it is important to know how a variety of physical or chemical parameters influence acoustic power or cavitation intensity.

The purpose of present study is to investigate the relation between the temperature or pressure and the efficiency of the chemical reaction induced by ultrasonication in a flow-type sonochemical reactor. The frequency-dependence of the transducer impedance was investigated under different pressure and temperature conditions. The effects of the temperature and pressure on the sonochemical effect were evaluated by KI dosimetry.

2. Experimental

Special grade chemicals potassium iodide (KI, Waco Chemicals) was used to prepare the aqueous solution of KI with concentration 0.1 mol·dm^{-3} by dissolving the reagent in the ion-exchange water.

Figure 1 shows a schematic diagram of the ultrasonic apparatus. The diameter of the horn tip of a homogenizer is 13 mm. The cylindrical reactor of a stainless-steel with an effective volumetric capacity of ca. 4× 10^{-2} dm^3 was immersed in a water bath and the sample mixture solution was controlled. The horn tip was located at 18 mm in height from the bottom of the reactor. The solution in a tank left in the atmosphere at 298K until attaining oxygen saturating concentration. The solution pressurized

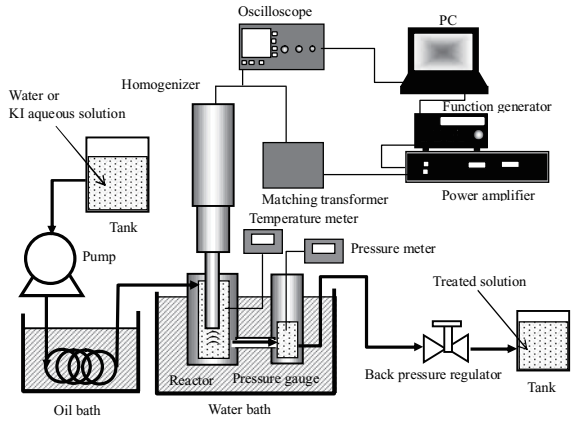

Fig. 1 A schematic diagram of experimental set-up

by a pump (PU-2086, Jasco Corp.) was fed into the sonochemical reactor at the flow rate of 10^{-3} dm^3·min^{-1}. The temperature and pressure of solution were measured by a temperature sensor (Digital Panelmeter, Asahikeiki Co., Ltd.) and a pressure gauge (Diaphragm-seal Pressure gauges for High Temperature, Nagano Keiki Co., Ltd.) equipped with the reactor. The temperature was controlled using a water bath and an oil bath. The pressure was controlled by a back pressure regulator (TESCOM Corp.). The effective electric power input to the transducer was calculated from the voltage at the both ends of the transducer and the current measured by using an oscilloscope (TDS3012B, Tektronix Inc.) and a current probe (TCP202, Tektronix Inc.). The absolute value of impedance of the transducer was calculated in the ratio of effective voltage to effective current. In this work, the electric power applied to the transducer was 50 ± 1W for the sonochemical effect evaluation. For measurement of the impedance of the transducer, the voltage of the function generator was set at 0.4 $V_{p\text{-}p}$.

The oxidation reaction of KI in aqueous solution was used in order to evaluate the sonochemical effect in the reactor[2)]. When ultrasound waves irradiate the aqueous solution of

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KI, I^- ions are oxidized to I_2. When excess I^- ions are present in the solution, I_2 reacts with an excess I^- ion to form an I_3^- ion. After ultrasonication, the solution was stirred and 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.).

3. Results and Discussion

Figure 2 illustrates typical frequency -impedance curves. In the frequency range of 20-22 kHz, there is a local minimum value of the impedance of the transducer. The results in Fig.2 demonstrate that the frequency at which the local minimum value of the impedance is obtained depends on the pressure and temperature. The influence of pressure and temperature on the frequency was investigated. The results are illustrated in **Fig. 3**. The frequency increases with the pressure in the range of 0.13-2.0 MPa, while the frequency decreases with an increase in the temperature in the range of 293-303 K. These may be attributed to a change in the acoustic velocity according to temperature and pressure conditions.

For the operation of a flow-type sonochemical reactor, a typical change of I_3^- absorbance is shown in **Fig. 4**. In the case of the flow rate of 10^{-3} $dm^3 \cdot min^{-1}$, the steady-state is achieved after 180min. For different pressures and temperatures, the data of the steady-state absorbance were taken. In **Fig. 5**, the absorbance values are plotted against pressures at the temperatures of 293, 298 and 303 K. Experiments were carried out at the frequency at which the local minimum value of the impedance is obtained under the different pressure and temperature conditions (Fig.3). The absorbance decreases with an increase in pressure, probably partial suppression of the formation/growth of cavitation bubbles. The results in Fig.5 indicate that the absorbance decreases with an increase in temperature. The rise in temperature leads to an increase of vapor pressure in cavitation bubbles. This may give rise to the partial difficulty in contraction/collapse of the cavitation bubbles, which is probably responsible for the reduction of sonochemical effects.

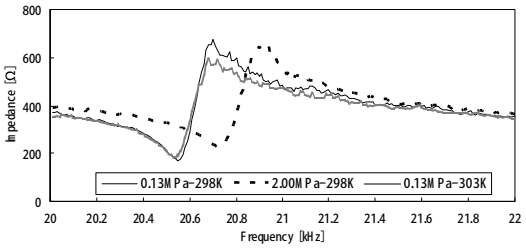

Fig. 2 Typical frequency -impedance curves.

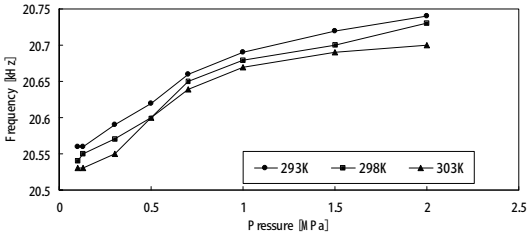

Fig. 3 The influence of pressure and temperature on the frequency at which the local minimum value of the impedance is obtained.

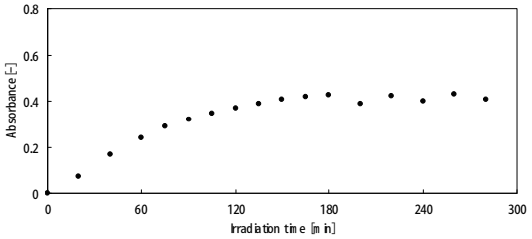

Fig. 4 Typical experimental result of I_3^- absorbance. (298K, 0.13MPa)

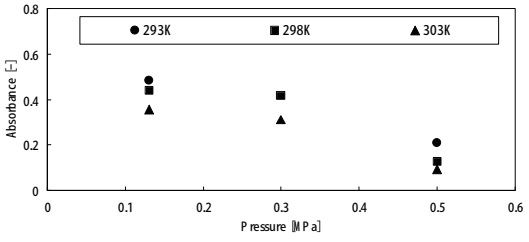

Fig. 5 Pressure dependence of I_3^- absorbance at the temperatures of 293, 298 and 303 K.

Acknowledgement

A part of this work was supported by a Grant-in-Aid from the Japan Society for the Promotion of Science (for Exploratory Research (No. 20656130)).

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