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Effect of Particle Addition on Ultrasonic Degradation Reaction Rate

超音波分解反応速度に固体粒子添加がおよぼす影響

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1. Introduction

Recently, ultrasound has been used as an advanced oxidation method for wastewater treatment[1,2]. In addition, the sonochemical reaction is enhanced by particle addition. Especially, the combination of photocatalytic and ultrasonic irradiation is considered to enhance the degradation rate due to the increase in the generation of OH radical. The degradation of phenol by ultrasonic irradiation in the presence of particles has been investigated in complete darkness[3]. The degradation rate of phenol and the concentration of the low molecular weight compounds increase with the amount of TiO_2 particle. However, enhancement mechanism of particle addition has not been investigated well, because ultrasound enhances not only chemical reaction but also mass transfer. Therefore, it is important to classify ultrasonic effect into chemical and physical effects.

In this study, the degradation process of methylene blue as the model hazardous organic compound by ultrasonic irradiation was investigated. The effects of ultrasonic irradiation condition on degradation rate were investigated. The effect of ultrasonic frequency on improvement of degradation reaction by particle addition was also investigated.

2. Experimental

Fig. 1 shows the experimental apparatus. A stainless steel plate attached with PZT transducer (Honda Electronics Co., Ltd.) was installed in the center of the water bath at the bottom. The ultrasonic frequency was operated at 22.8 kHz, 127 kHz, 490 kHz, 940 kHz and 1640 kHz. The transducers were driven by a power amplifier (1040L, E&J), which in turn was driven by a continuous sinusoidal wave produced using a signal generator (WF1974, NF Corp.). The effective electric power input to the transducer was calculated from the voltage at both ends of the transducer, the current measured using an oscilloscope (TDS3012C, Tektronix Inc.), and a current probe (TCP202, Tektronix Inc.). The diameter and the approximate volume of the glass reactor were 85 mm and 1 L, respectively. The temperature of the water bath was kept constant by a thermostat.

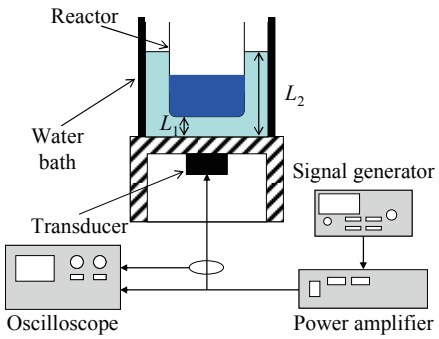

Fig. 1 Experimental apparatus

Table 1 shows the experimental conditions for methylene blue degradation. Process variables were defined as follows: ultrasonic frequency (f), ultrasonic output power (P), distance between the ultrasonic transducer and the bottom of the reactor (L_1), distance between the ultrasonic transducer and the level of the water bath (L_2), irradiation time, and amount of particle (TiO_2 or Al_2O_3) addition (w). The volume of the sample solution (V), temperature of the water bath (T), and initial methylene blue concentration (C_0) were kept constant. The diameters of additive particles of TiO_2 and Al_2O_3 were 300 nm and 50 μm, respectively.

Table 1 Experimental conditions

f [kHz]	P [W]	L_1 [mm]	L_2 [mm]	T [K]	C_0 [mol/m^3]	t [min]	V [m^3]	w [g]
22.8	0 - 20	45	80	298	0.01	0 - 60	0.1×10^{-3}	0 - 1
127		10	60					
490		10	60					
940		10	60					
1640		10	60					

Before ultrasonic irradiation, the sample solution and the remaining space in the reactor were deoxygenated with a nitrogen gas flow for 20 min. After deoxygenation, the sample was irradiated with ultrasound under a continuous flow of nitrogen gas (0.1 L/min). After ultrasonic irradiation, the methylene blue concentration (C) was determined by measuring the absorbance of the sample at a wavelength of 665 nm using UV-vis spectrometer (Agilent 8453, Agilent Technologies). The determined absorbance was converted to a concentration through the standard curve of methylene blue. Before the analysis, the suspension was centrifuged to remove particles. The ultrasonic power in the reactor was measured by calorimetry[4]. The effects of frequency on sonochemical efficiency were also evaluated using SE_{KI} value[5].

3. Results and Discussion

Fig. 2 shows the effect of amount of particle addition on the apparent degradation rate constant at frequency of 490 kHz and power of 8 W. The apparent degradation rate constant in the presence of TiO_2 particles increased with increasing particle concentration linearly until 13 g/L. The optimal concentration of TiO_2 was 0.25 g/L in the photocatalytic irradiation system, because the UV light was hindered by the excess TiO_2 particles[3]. However, such a phenomenon was not observed in this ultrasonic irradiation system. Therefore, TiO_2 particles are used effectively in the sonocatalytic irradiation system. On the other hand, the apparent degradation rate constant in the presence of Al_2O_3 particles also increased with increasing particle concentration. It is reported that degradation rate of chlorobenzene was enhanced by the addition of Al_2O_3 particles, and the surface area influenced the improvement of degradation rate[6]. The result of this study agrees with previous study.

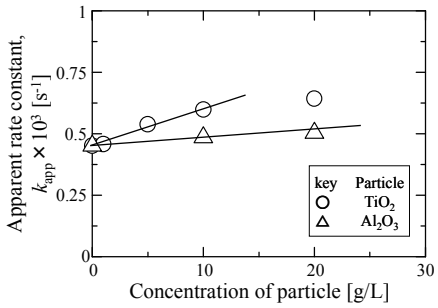

Fig. 2 Effect of particle concentration on apparent degradation rate constant (f = 490 kHz, P = 8 W)

Fig. 3 shows the effect of ultrasonic frequency on the apparent degradation rate constant for various particles at ultrasonic power of 8 W and particle amount of 1 g. The apparent degradation rate constant of methylene blue was increased by particle addition in these 3 frequencies. In the conditions of frequencies of 490 and 1640 kHz, TiO_2 particles were more effective for degradation than Al_2O_3 particles. However, apparent degradation rate with Al_2O_3 particle was more effective than TiO_2 particle at the frequency of 127 kHz. It is considered that the enhancement of apparent rate constant by particle addition was influenced by both ultrasonic frequency and particle type or diameter.

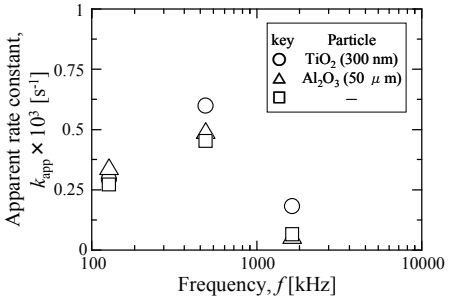

Fig. 3 Effect of frequency on apparent degradation rate constant with particle (P = 8 W, w = 1g)

4. Conclusions

The degradation process of methylene blue was intensified by particle addition, and the degradation rate increased with increasing amount of particle addition. The enhancement of degradation rate by particle addition was influenced by both ultrasonic frequency and type or diameter of particles.

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