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Effect of Particle Addition on Degradation Rate of Methylene Blue in an Ultrasonic Field

超音波を用いたメチレンブルー分解速度に固体粒子がおよぼす影響

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1. Introduction

Many organic compounds have been released and appeared as environmental pollutants. Techniques for the degradation of hazardous organic compounds have been investigated. UV light irradiation to TiO_2 photocatalysis is one of the favorite techniques[1)]. The positive holes generated by UV light irradiation in the vicinity of the surface of TiO_2 particles react with water to produce OH radicals. However, UV light is screened by TiO_2 particles, so that there is a limit amount of particle addition in which degradation rate increases.

Ultrasound has been found to be an attractive advanced technology for the degradation in water[2)]. In addition, the sonochemical reaction is enhanced by particle addition. Especially, the combination of photocatalysis and ultrasound is considered to enhance the degradation rate. The degradation of phenol by ultrasonic irradiation in the presence of TiO_2 particles has been investigated in complete darkness[3)]. Sekiguchi and Saita have been investigated the effect of Al_2O_3 particles on the degradation of chlorobenzene in an ultrasonic field[4)]. However, the enhancement mechanism of particle addition has not been investigated well, because ultrasound enhances not only chemical reaction but also mass transfer.

In this study, the ultrasonic degradation of methylene blue was carried out, and the effects of ultrasonic irradiation condition on the degradation rate were investigated. The effects of ultrasonic frequency on improvement of degradation 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.

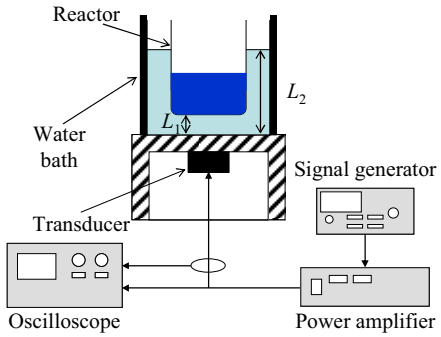

Fig. 1 Experimental apparatus

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.

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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		45	80					
127		10	60					
490	0 - 20	10	60	298	0.01	0 - 60	0.1×10^{-3}	0 - 1
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[5]. The effects of frequency on sonochemical efficiency were also evaluated using SE_{KI} value[6].

3. Results and Discussions

Fig. 2 shows the effect of TiO_2 concentration on apparent degradation rate constant at f = 490 kHz and P = 8 W. The apparent degradation rate constant increases with increasing TiO_2 concentration. 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[7]. However, such a phenomenon was not observed in this ultrasonic irradiation system.

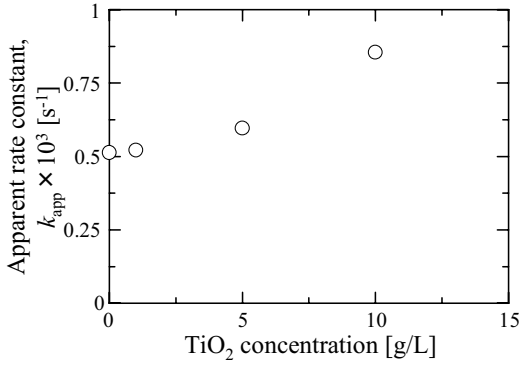

Fig. 2 Effect of TiO_2 concentration on apparent degradation rate constant at f = 490 kHz and P = 8W

Fig. 3 shows the effect of particle addition on the time dependence of methylene blue concentration at P = 8W and w = 1.0 g. The degradation rate of methylene blue was increased by particle addition. The degradation rate was improved by TiO_2 particle addition at f = 490 kHz. On the other hand, the degradation rate was improved by Al_2O_3 particle addition at f = 127 kHz. The relationship between particle size and resonance diameter is important to enhance the ultrasonic reaction in the presence of particle.

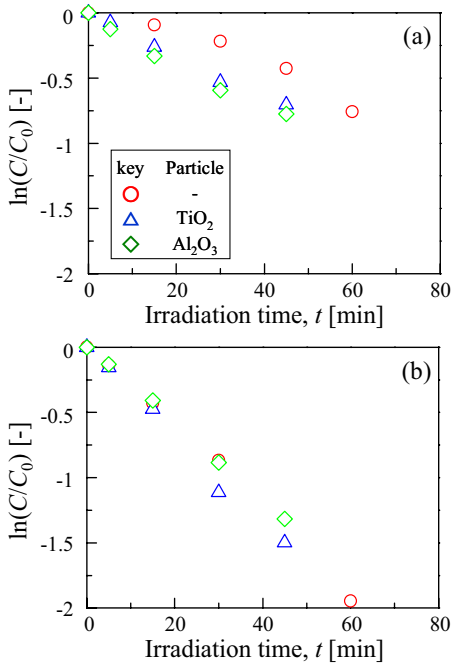

Fig. 3 Effect of particle addition on time dependence of methylene blue concentration at P = 8 W and w = 1.0 g: (a) f = 127 kHz, (b) f = 490 kHz

4. Conclusion

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 of particle.

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