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Effect of Initial Concentration on Ultrasonic Degradation of Methylene Blue

メチレンブルーの超音波分解速度に初期濃度がおよぼす影響

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1. Introduction

Recently, ultrasound has been used as an advanced oxidation method for wastewater treatment[1,2]. Sonochemical degradation kinetics of hazardous organic compounds has also been investigated, and some kinetics models have been proposed[3,4]. In these models, the effect of ultrasonic frequency on degradation rate has not been investigated. On the other hand, a simple pseudo-first-order reaction model is typically suggested[5]. And, it is reported that the effect of ultrasonic frequency on degradation of phenol correlates with the degree of hydrogen peroxide formation. However, there have been few quantitative analyses of the effect of ultrasonic frequency on degradation rate.

In our previous study, ultrasonic degradation of methylene blue as a model hazardous organic compound was performed at frequencies of 22.8, 127, and 490 kHz. Apparent degradation rate constants were evaluated using a pseudo-first-order reaction model, and we have proposed a simple model for estimating the apparent degradation rate constant of methylene blue on the basis of SE_{KI}[6]. In this study, degradation of methylene blue was performed at various frequencies, especially in the high frequency region around 1 MHz. In addition, the effect of initial concentration of methylene blue on degradation rate was also investigated. The purpose of this study is to determine whether the operable frequency and intial concentration regions of our proposed model can be extended to increase versatility.

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.

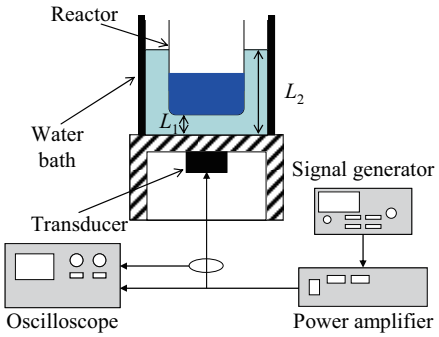

Fig. 1 Experimental apparatus

Table 1 shows experimental conditions for methylene blue degradation. Process variables are defined as follows: the ultrasonic frequency (f), ultrasound output power (P), initial methylene blue concentration (C_0), distance between the ultrasonic transducer and bottom of the reactor (L_1), distance between the ultrasonic transducer and level of the water bath (L_2), and irradiation time (t). The volume of the sample solution (V) and temperature of the water bath (T) were maintained constant. Before ultrasonic irradiation, the sample solution and remaining space in the reactor were deoxygenated with a nitrogen gas flow for 20 min at 298 K. After deoxygenation, the sample was irradiated with ultrasound under a continuous nitrogen gas flow

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(0.1 L/min).

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]
22.8	0 - 20	45	80	298	0.005 - 0.07	0 - 60	0.1×10^{-3}
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. The ultrasonic power in the reactor was measured by calorimetry[7]. The effects of frequency on sonochemical efficiency were also evaluated using SE_{KI} value[8].

3. Results and Discussions

Fig. 2 shows the effect of ultrasonic frequency on the relationship between apparent degradation rate constant and ultrasonic power. The rate of degradation of methylene blue was influenced by ultrasonic frequency, and the degradation rate at 490 kHz was much higher than those at 22.8 and 1640 kHz. It has been reported that sonochemical efficiency in the frequency range of 200–500 kHz is 10 times greater than those in low and high frequency regions[8]. For degradation of methylene blue, the apparent degradation rate constant at 490 kHz was approximately 15 times greater than that at 22.8 kHz under the same ultrasonic power. Our results suggested that there is a relationship between degradation rate constant and sonochemical efficiency, and the apparent degradation rate constant may be quantitatively estimated using SE_{KI}.

Fig. 3 shows the effect of initial concentration of methylene blue on the degradation rate constant at the frequency of 490 kHz, and at the ultrasonic power of 5 W. The degradation rate decreases with increasing initial concentration of methylene blue. In addition, the initial concentration and apparent degradation rate constant are inversely proportional, which agrees with our proposed model.

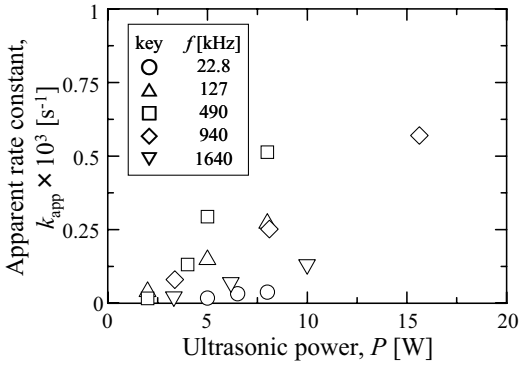

Fig. 2 Effect of ultrasonic frequency on the relationship between the apparent degradation rate constant and ultrasonic power

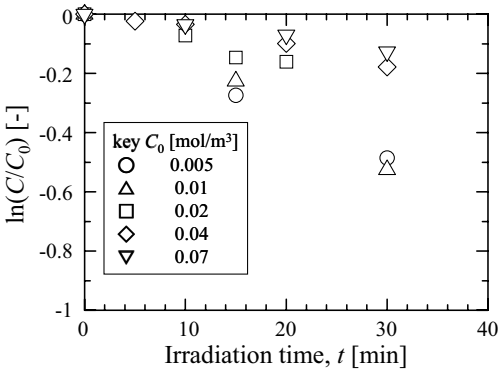

Fig. 3 Effect of initial concentration of methylene blue on the time dependence of methylene blue concentration (f = 490 kHz, P = 5 W)

4. Conclusions

Our proposed model for estimating the apparent degradation rate constant using power and SE_{KI}, which extended the frequency and initial concentration range, is applicable to this study.

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