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Decrease in the Rate of Sonochemical Oxidation with Introducing CO_2

二酸化炭素導入によるソノケミカル反応の抑制

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1. Introduction

Ultrasonic irradiation system has attracted the attention of a great deal of engineers in food industries from the viewpoint of non-contact mixing, namely a physical effect. This system, however, provides not only physical effect but also chemical one. In some cases, chemical action has a negative effect for the main aim. In the food industries, for example, sauce or soup changes its nature or quality by sonolysis, namely chemical effect. In that case, chemical action should be suppressed with keeping the physical effect. To cope with the problem, addition effect of CO_2 in the reaction system on the rate of oxidation was examined. Cavitation is the key for the sonochemical process during ultrasonic irradiation. The cavitation is dominated by dissolved gas. In previous papers[1) 2)], it was reported that CO_2 prevented sonochemical action in solution because of low temperature for cavitation. In this presentation, we introduced CO_2 in the system for blocking chemical action and determined the boundary concentration of CO_2 in matrix. In addition, a promising approach for the application of ultrasound assisted system will be discussed.

2. Experimental

Ultrasonic irradiation was performed using an ultrasonic atomizer (Honda Electric HM-303N, 2.4 MHz, 24 W) for 10 minutes or an ultrasonic bath (Honda Electric 200 kHz, 15 W) for 30 minutes from the bottom surface of the reactor as shown in Fig. 1. In the former examination, similar atomization behavior was observed in all cases.

The rate of sonochemical oxidation was evaluated by potassium iodide (KI) dosimetry[3)] using UV-Vis spectrophotometer (JASCO V-650).

For the practical use, we try to a sensory test using 20 % ethanol as a model reactant. Sonolysis of this material was carried out and smell change was confirmed before and after sonolysis.

The reactor was filled with atmospheric gases. As atmospheric gases, a variety of concentration of CO_2 was used. After gas introducing, the reactor was sealed and ultrasonic irradiation was started at 25°C.

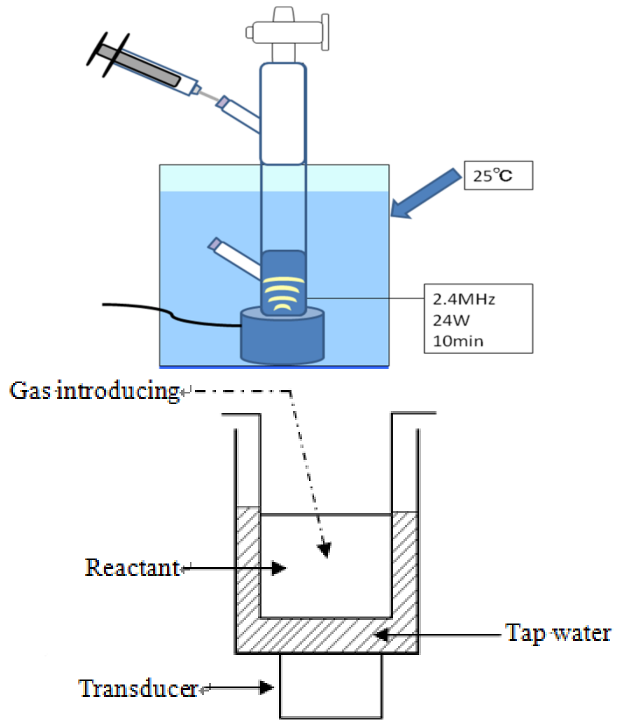

Fig. 1 Experimental setup for examinations
Upper: Ultrsonic atomizer (2.4 MHz, 24 W)
Lower: Ultrasonic bath (200 kHz, 15 W)

3. Results and discussion

In the pure CO_2 atmosphere, ultrasonic irradiation had hardly any chemical effects because CO_2 is consisted to three atoms.

Fig. 2 shows change in absorbance as function of the concentration of CO_2 in the air.

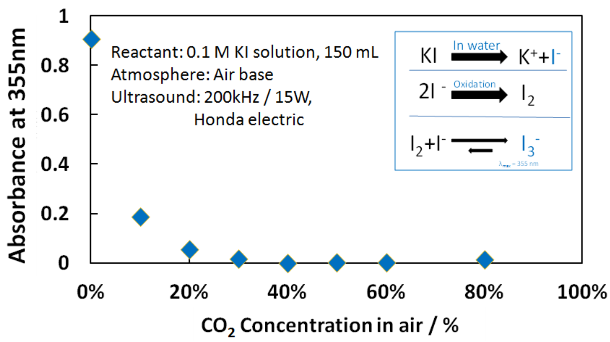

Fig. 2 Decrease in oxidation power with introducing CO_2 gas.

The absorbance decreased with increasing CO_2 concentration. Sonochemical oxidation power was lost over 30 % of CO_2 in the matrix. Thus, this concentration is enough for blocking sonolysis of a target material.

It is known that dissolved gas in the solution plays an important role for sonolysis and argon (Ar) has well capability for sonolysis. In the case of an Ar atmosphere, sonolysis of KI solution was performed with introducing CO_2.

Fig. 3 shows the addition effect of CO_2 on the rate of oxidation at 2.4 MHz. As the result, concentration dependence of Ar was similar to the air. Thus, at high frequency (2400 kHz), introducing over 30 % CO_2 was also effective for blocking sonochemical oxidation.

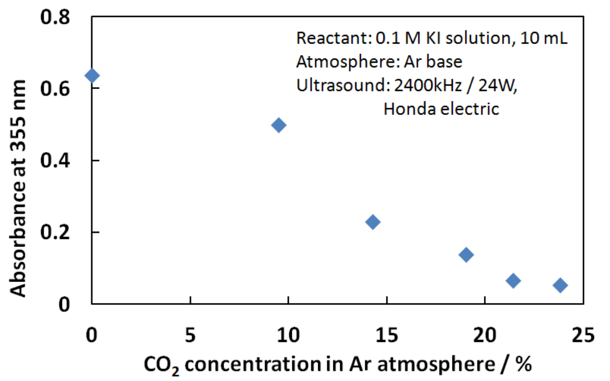

Fig. 3 Effects of atmosphere or frequency on the oxidation power

Incidentally, cavitation occurs not in the atmosphere but in the solution. The amount of CO_2 should be estimated in the solution. However, now we use concentration in the atmosphere. In near future, we try to estimate CO_2 dissolved and discuss clearly the relationship between oxidation behavior and concentration of CO_2.

In addition, we tried not only gaseous CO_2 but also solid CO_2 (dry ice), sodium hydrogen carbonate, which provides CO_2 from an aqueous solution under ultrasonic irradiation, and ascorbic acid were used as adding agents.

Table 1 indicated the results of a sensory test in an Ar and Air atmosphere. This table also indicated gaseous products after sonolysis. In the case of Ar atmosphere sonolysis proceeded. In air atmosphere, on the other hand, products were not identified. It was thought that almost all gaseous products were oxidized to CO_2 by oxygen in the air. However, nasty smell was spread out obviously both pure Ar and Air atmosphere.

By introducing CO_2, on the contrary, the smell was blocked.

Table 1 Sonochemical products and sensory test after sonication.

Atmosphere	Product /μmol 3h^{-1}			Smell test
	H_2	CH_4	C_2H_6	
Ar	18.7	18.3	1.43	Strong
Air	0	0	0	Weak
20%CO_2-Air	0	0	0	Non

Ultrasound: 200 kHz, 15 W; Reactant: 20% ethanol; Irradiation time: 3 hours

4. Conclusion

To prevent an unwanted sonochemical reaction CO_2 was introduced to the ultrasonic irradiation system. The rate of sonochemical oxidation decreased with increasing concentration of CO_2 in atmosphere. To block the unwanted sonolysis 30 % CO_2 was needed in the matrix.

References

1. H. Harada: Ultrason. Sonochem. 5 (1998) 73.
2. H. Harada, N. Iwata, K. Shiratori: Jap. J. Appl. Phys. (2009), 48 07GH01-1-4.
3. S. Koda, T. Kimura, T. Kondo, and H. Mitome: Ultrason. Sonochem. **10** (2003) 149.