

## Effect of Frequency on Sonolysis Using Several Kinds of Reactors

周波数の異なる超音波発振器を利用した効果的ソノケミカル反応の研究

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### 1. Introduction

Ultrasonic waves provide a variety of physical and chemical effects to a chemical reaction system. Stirring, vibration, and shock waves are examples of the former. The latter means a radical reaction and/or a thermal one. The source of sonochemical effects in the solution is a phenomenon known as acoustic cavitation. In the micro spot area of the cavitation field, temperature and pressure drastically increase and active chemical species such as radicals, oxidants, etc. are produced. Because of the appearance of the special field for chemical reactions, sonochemistry has attracted the attention of a great deal of scientists and engineers over the last few decades. However, it is difficult to apply practical use because many parameters influence the yield.

Ultrasonic frequency, the shape of the reactor, and volume of reactant solution are considered important factors. Dependence of chemical power has reported on frequency below 1 MHz<sup>1), 2)</sup> and on vessel diameter<sup>3)</sup> or shape<sup>4)</sup>. In this presentation, we examine the influences of high frequency (megahertz region) and of glass thickness on chemical power.

### 2. Experimental procedure

Five types of oscillators were prepared: 2.4 MHz (2400 kHz) atomizer type (Honda Electric, HM-303N, 24 W, 7.64 W/cm<sup>2</sup>), 1.6 MHz (1600 kHz) atomizer type (TDK, TU-20A, 30 W, 6.63 W/cm<sup>2</sup>), 500 kHz bath type (Honda Electric, 50 W, 1.30 W/cm<sup>2</sup>), 200 kHz submersible type (Shinka Industry, SR-200K, 100W, 2.60 W/cm<sup>2</sup>), 19 kHz horn type (CHO-ONPAPA KOGYO, H-20, 10W, 20 W/cm<sup>2</sup>).

As reactors, Round bottom glass cylinders like test tubes, flat bottom glass cylinders, thin glass bottom cylinders, and glass sample tubes were used. Chemical effects were estimated by KI dosimetry

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(A Standard Method to Calibrate Sonochemical Efficiency of an Individual Reaction System recommended by Japan Society of Sonochemistry).<sup>1)</sup> 0.1 M KI solution was prepared and coloring of it was monitored at 351 nm ( $\lambda_{\max}$ ) by a UV-Vis spectrophotometer (JASCO, V-650).

### 3. Results and discussion

According to previous reports,<sup>1), 2)</sup> the most effective frequency region for chemical reaction is several hundreds of kilohertz. On the other hand, effective chemical power and sonoluminescence were also obtained at 2400 kHz.<sup>5), 6)</sup> To consider the limits of frequency for getting effective chemical power, sonolysis of KI solution (dosimetry) was carried out at 2400 kHz and at 19 kHz.

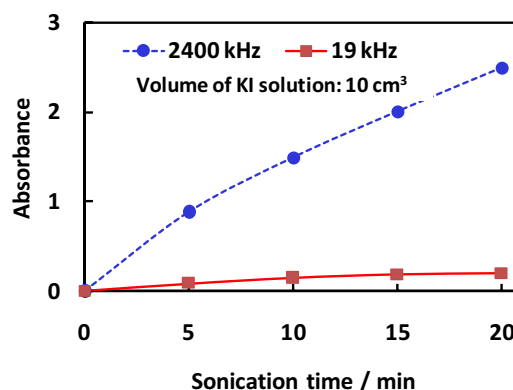


Fig. 1 Comparison of chemical power between 2400 kHz and 19 kHz; 0.1 M KI solution was contact with transducer;  $\lambda_{\max} = 351$  nm; Temperature: 25°C.

**Figure 1** shows the increase in absorbance with sonication time. Strong absorbance was obtained at 2400 kHz although output power density of this transducer was smaller than that at 19 kHz. Capillary waves were observed on the liquid

surface and a large amount of mist and/or very small droplets were formed from a solution, resulting in a foggy state in the area over a gas (air)-liquid interface at 2400 kHz.

Horn type and low frequency oscillator, on the other hand, was unfavorable for sonolysis, as only little absorbance was observed at 19 kHz.

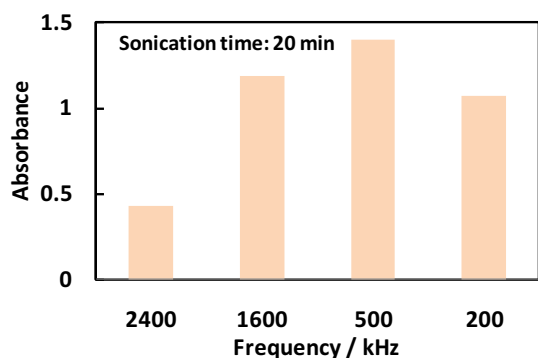


Fig. 2 Effect of ultrasonic frequency on chemical power; Reactor: Flat bottom (thick).

**Figure 2** shows oxidizing power as it appeared in the solution at a variety of frequencies. As different acoustic power was applied at each examination as indicated in the experimental section, comparison among them might be difficult. However, the most effective frequency is similar to reports.<sup>1), 2)</sup>

Figure 2 also indicates that lower absorbance at 2400 kHz was observed compared with Fig. 1. The difference in absorbance was six times (2.57 / 0.427) for 20 min irradiation. In the case of Fig. 1, KI solution was contact with the transducer. In Fig. 2, on the other hand, ultrasonic waves had to pass through apparatus the glass and chemical power decreased.

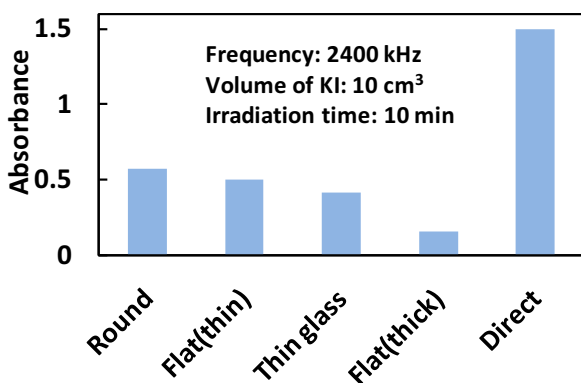


Fig. 3 Effect of glass thickness on chemical power at 2400 kHz; Thin glass: 1.5 μm cover glass for microscope.

According to our previous report,<sup>6)</sup> decrease in chemical power was related to the glass thickness of the apparatus.

To reduce the loss of chemical power, the relationship between chemical power and glass thickness of the apparatus was examined. The influence of apparatus shape was also considered.

**Figure 3** shows how the glass thickness affected the rate of sonolysis. Cylinder tubes made of thin glass with a closed bottom (like a glass test tube) were better for sonolysis.

Round bottom type cylinders made of thin glass were better for sonolysis than flat bottomed ones.

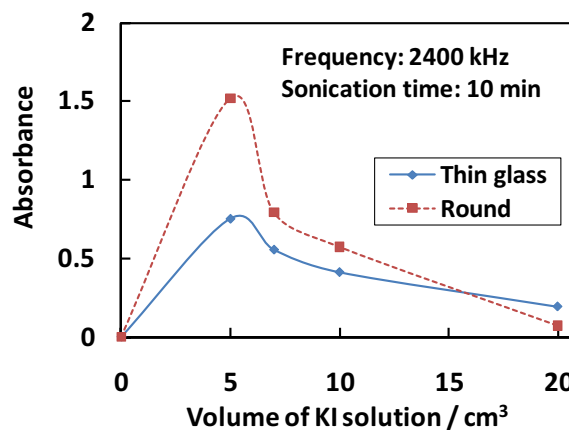


Fig. 4 Effect of volume of solution on chemical power in two types of apparatus; Frequency: 2400 MHz.

**Figure 4** indicates the effect of volume of reactant on the absorbance. The less volume was applied, the more rapid that colouring was obtained. Further discussion is needed because the volume of the solution surface was related to the height of the solution.

#### 4. References

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