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Evaluation of generation amount of superoxide anion radicals generated by ultrasonic cavitation in TiO_2 suspension

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1. Introduction

A phenomenon in which a high concentration of radicals is generated has been found when high-intensity ultrasonic waves are irradiated to a suspension of titanium dioxide (TiO_2), and many studies have been reported on this[1)]. These radicals act on bacteria or viruses to sterilize and inactivate them. Using this phenomenon, a sterilization treatment technology that is harmless to the human body can be realized without the use of chemicals[2)]. As one of the problems to be solved in order to realize the sterilization treatment technology using ultrasonic waves, a quantitative evaluation method of the amount of radical generation is required. In this study, we propose a method to evaluate the amount of superoxide anion radicals generated by ultrasonic waves radiated to a TiO_2 suspension in which luminol is dissolved, using the phenomenon of sonoluminescence from the decay energy of ultrasonic cavitation.

2. Radical generation process

As TiO_2 with a band gap of 3.2 eV is excited by the cavitation decay energy, conduction band electrons (e^-) and valence band holes (h^+) are generated[3)].

$$TiO_2 \rightarrow e^- + h^+ \quad (1)$$

Conductor electrons reduce or supplement the oxygen molecules of TiO_2 to the surface of TiO_2 to become supplemental electrons (e^-) to reduce oxygen molecules to generate superoxide anion radicals (·OH).

$$e^- + O_2 \rightarrow \cdot O_2^- \quad (2)$$

The valence band holes oxidize water molecules to form hydroxyl radicals (·OH).

$$h^+ + H_2O \rightarrow \cdot OH + H^+ \quad (3)$$

From the above process, the amount of hydroxyl radical and superoxide anion radical generated strongly depends on the ultrasonic cavitation.

3. Experimental method

The luminol has been successfully applied to monitor the superoxide anion radical($\cdot O_2^-$) produced on catalysis of TiO_2 in aqueous suspension[4)]. In this study, we generate the superoxide anion radical using ultrasonic cavitation and propose a method to evaluate the concentration of the superoxide anion radical by the luminescence intensity. Figure 1 shows the experimental setup for evaluating the amount of superoxide anion radical. The PM Tube (Photo Multiplier Tube) was used to measure the luminescence intensity of luminol. In a cylindrical metal container with an inner diameter of 90 mm, a suspension prepared by mixing TiO_2 powder and 50 mg of luminol in 500 ml of distilled water is filled. Ultrasonic waves radiated from a Langevin-type ultrasonic transducer with a radiation surface diameter of 44.5 mm mounted on the bottom of the metal container generate the cavitation, and the decay energy of the cavitation excites TiO_2, resulting in hydroxyl radical and superoxide anion radical. At that time, it causes also a luminescence reaction by luminol. The luminous intensity is received from a PM Tube (Hamamatsu, H10722-110) fixed at a distance of 24 mm from the liquid level of the suspension, and the output voltage is measured with an oscilloscope. The admittance characteristics of the Langevin-type ultrasonic transducer used in the experiment were measured, as shown in Fig. 2.

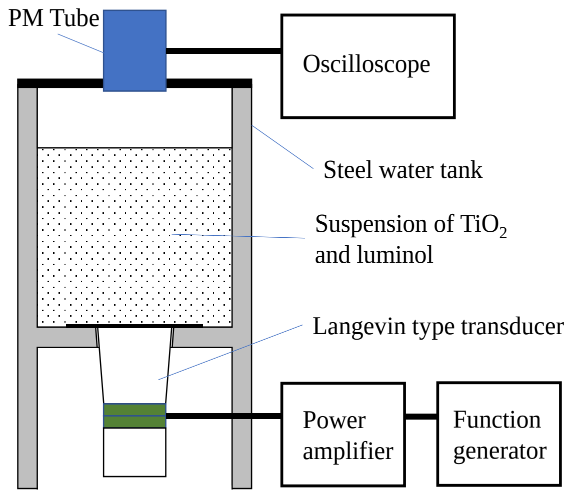

Fig. 1 Experimental setup for evaluating the amount of superoxide anion radical.

Figure 2(a) shows the results of the input admittance when 500 ml of distilled water is filled in a metal container as an acoustic load. It can be seen that the resonant frequency is about 40.64 kHz. Figure 2(b) shows the admittance locus of air load as well as of

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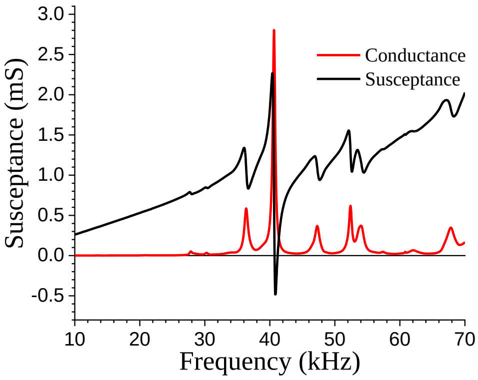
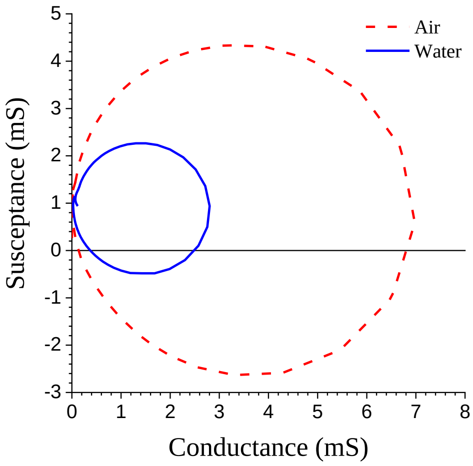
water load in complex coordinates to obtain the electroacoustic efficiency. From this result, it could be estimated that the electroacoustic conversion efficiency was about 59%[5]. The continuous sine wave from the signal generator is amplified by the power amplifier(1020 L, E&I, USA) and input to the Langevin-type ultrasonic transducer. The condition for maximum power transmission was when the driving frequency was 40.3 kHz, and the input current and voltage were 0.22 A_{rms} and 157 V_{rms}, respectively.



(a)



(b)

Fig. 2 Input admittance of Langevin-type ultrasonic transducer. Admittance for the water as acoustic load (a) and admittance loci for air and water as acoustic load (b).

4. Results and discussion

Figure 3 shows the luminous flux received by PM Tube when ultrasonic waves were irradiated for 40 seconds while varying the amount of TiO_2 powder added to distilled water. From this result, even in the absence of TiO_2 (0%), the luminescence phenomenon occurs due to the ultrasonic cavitation. As the TiO_2 concentration in the suspension increases, the luminescence intensity is significantly increased due to the catalytic reaction by TiO_2. However, in the given range of the TiO_2 concentration, as the concentration increases, the increasing rate of the luminescence intensity is reduced. It is thought that the turbidity of the suspension increases as the concentration of TiO_2 increases, and the turbidity disturb the reaching the light energy generated by the ultrasonic energy to the PM Tube.

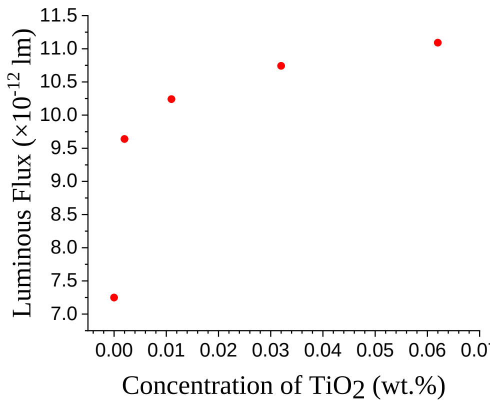


Fig. 3 The luminescence intensity change with TiO_2 concentration.

5. Summary

A method for evaluating the amount of superoxide anion radicals generated by the catalytic reaction of titanium dioxide during ultrasonic cavitation decay was proposed. The cavitation generated by ultrasonic waves radiated to the luminol solution to which the titanium dioxide powder is added causes the generation of superoxide anion radicals and the luminescent reaction of luminol. The luminescent intensity generated at this time was measured by PMT to evaluate the change in the amount of superoxide anion radicals generated depending on the concentration of titanium dioxide.

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