

On the Mist Separation and the Sonochemiluminescence under Pulsed Ultrasound

パルス超音波による霧生成と音響化学発光

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1. Introduction

When an ultrasound propagating in a liquid is incident into the interface between air and the liquid, a capillary wave propagating along the liquid surface is induced¹⁻³. Under the condition where the amplitude of sound pressure near the liquid surface is over a certain threshold, the top of the capillary wave breaks to provide tiny liquid drops (mist) that disperse. An attachment of the mist created through the above ultrasonic atomization to an object enables us to make the cooling of the object by the action of the evaporation heat. There have been a number of fundamental studies on atomization using ultrasound waves. Eisenmenger observed capillary waves on a liquid surface that led to atomization over a broad range of ultrasound frequencies from 10 kHz to 1.5 MHz¹. Lang measured the average size of mist droplets in the frequency range 10 kHz to 800 kHz⁴, and obtained the relationship; $D = 0.34 (8\pi T/\rho F^2)^{1/2}$ (D the droplet size, T the surface tension, ρ the density of the liquid, and F the ultrasound frequency). Sato et al. applied the ultrasonic atomization technique to a mixed solution of water and ethanol and successfully accomplished the condensation of ethanol⁵. However, to the best of the author's knowledge, there has been no report on the simultaneous production of both an ultrasonic physical effect (ultrasonic atomization in this paper) and a chemical effect due to ultrasonic cavitation bubbles.

In the present study, the author investigated the power dependence of both the extent of mist separation based on the cooling rate and the sonochemiluminescence (SCL) intensity for pulsed ultrasound, in order to clarify whether the effective power range for the physical and chemical effects are the same. A mechanism that explains the difference in the power dependence for pulsed and CW ultrasound is suggested⁶. The author attempted to maximize the amount of water mist by varying the duty cycle of the pulsed ultrasound.

2. Experiment

The author measured the temperature of a copper plate towards which mist due to ultrasonic atomization was directed. Evaporation of the mist from the plate

absorbs heat and decreases the temperature of the plate. The plate temperature was measured using a thermocouple placed on its surface. A CW or pulsed sinusoidal signal of 135 kHz generated by a function generator was amplified using a 55 dB power amplifier. The signal amplitude was the same for both CW and pulsed operation. To produce ultrasound, a Langevin-type transducer with a diameter of 45 mm was used together with a transducer plate. The transducer plate was surrounded by a circular flange to form a pool, into which distilled water was placed. This was then covered by a transparent plastic hood. The input power to the transducer was measured using a power meter. The mist was transported through a transfer tube to the copper plate using a carrier gas (Ar, 0.05 MPa). Water droplets with large volume splashed from water surface occasionally appeared, but was too heavy to be transported with the carrier gas. The initial volume of water was 10 mL and in some cases this was topped up at a rate of 5.5 mL per minute. The initial water temperature was 24 ± 1 °C. The cooling rate was estimated from the initial slope of the plate temperature vs. time curve, measured just after sonication began. The size of the mist droplets of water, calculated using the above relationship was 11 μ m.

The dependence of the SCL intensity on the input power was investigated using luminol. 10 mL of the luminol solution was placed in the pool on the transducer. Light emitted from the solution was detected using a photomultiplier tube and the output voltage from the PMT was measured using a digital oscilloscope.

3. Results and Discussion

Figure 1 shows the decrease in the copper plate temperature with time due to exposure to mist produced by ultrasonic atomization for both pulsed and CW operation. It can be seen that a larger and more rapid temperature drop occurs for pulsed ultrasound. Figure 2 shows the dependence of the input power to the transducer and the cooling rate on the number of OFF cycles when the number of ON cycles is 300. The origin of the horizontal axis represents the CW condition. Whereas the input power decreased monotonically with increasing OFF cycles, the cooling rate initially increased, showed a maximum, and subsequently

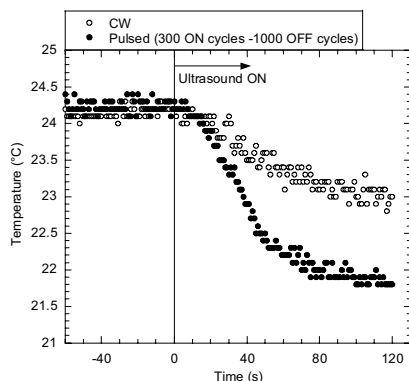


Figure 1. Change in plate temperature with time for CW operation and pulsed operation for a repetition of 300 ON cycles and 1000 OFF cycles (from Ref. 6 © 2012 American Chemical Society)

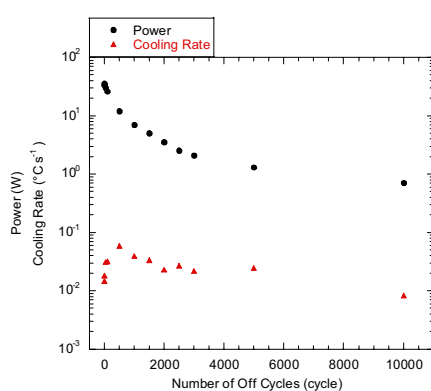


Figure 2. Dependence of transducer input power and cooling rate on the number of OFF cycles (from Ref. 6 ©2012 American Chemical Society)

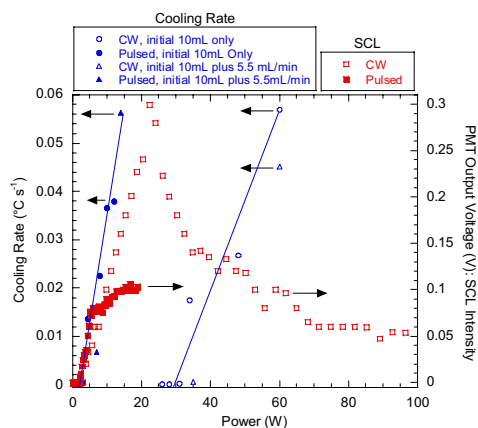


Figure 3. Input power dependence of cooling rate and SCL intensity for CW operation and pulsed operation with 300 ON cycles and 1000 OFF cycles (from Ref. 6 ©2012 American Chemical Society)

decreased. The highest cooling rate occurred for 300 ON cycles and 500 OFF cycles. Under these conditions, the cooling rate and the cooling rate per unit power were 4 and 12 times higher than for CW ultrasound, respectively. Thus, for appropriate pulse conditions, a higher cooling rate could be achieved than for CW ultrasound. During pulsed operation, it is believed that the generation of large bubbles due to coalescence is suppressed, so that

high sound pressure amplitude is maintained near the surface of the liquid. This may be responsible for the larger amount of mist generated during pulsed operation. Figure 3 shows the dependence of the cooling rate and the SCL intensity on the input power. The pulse conditions were a repetition of 300 ON cycles and 1000 OFF cycles. For CW ultrasound, SCL begins to occur at a low power threshold of less than 8 W. The SCL intensity initially increases with power, goes through a maximum, and subsequently decreases. In contrast, no appreciable cooling occurs until a much higher power threshold is reached, and the cooling rate then increases with power. However, at these power levels, the SCL intensity is already decreasing. In the case of pulsed ultrasound, cooling begins to occur at the same low power threshold at which SCL begins. Thus, at power levels of less than 8 W, pulsed operation gives rise to a higher mist production rate and SCL intensity than CW operation. Above the threshold, the cooling seems to increase continuously with power for both CW and pulsed operation, although the data are scattered. The cooling rate for relatively low CW or pulsed power levels seems lower for the case where the water was topped up at 5.5 mL/min. It is thought that the additional supply of liquid caused an increase in the load on the transducer, and the damping effect led to a reduction in the sound pressure amplitude at the liquid surface, and therefore in the mist generation rate.

The power dependence of the SCL intensity and the cooling rate is different for CW and pulsed operation. Measurements of the sound pressure amplitude near the liquid surface suggested that the formation of large sized bubbles during CW operation reduces the amplitude until the primary Bjerknes force expels these bubbles, allowing high amplitude sufficient for capillary wave excitation and mist separation⁶. The fact that the creation of such large bubbles is suppressed during pulsed operation allows the inception and promotion of sonochemical reaction and mist separation for the same range of ultrasonic power.

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