



# Active control of phase change from supercooled water to ice by ultrasonic vibration 1. Control of freezing temperature

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## Abstract

A method to actively control the supercooling of water is a critical issue in cold-energy storage and transport systems that use ice slurries. We experimentally studied the use of ultrasonic vibration to control the phase change from supercooled water to ice, and simulated the phase change assuming that ultrasonic-induced cavitation influences ice nucleation. The experimental results indicate that ultrasonic vibration strongly promotes the phase change from supercooled water to ice and that a reliable method to actively control the phase change by ultrasonic vibration can be realized. The results of the simulation agreed well with the experimental results. © 2001 Elsevier Science Ltd. All rights reserved.

## 1. Introduction

Electric power load in the daytime in summer has been steadily increasing in hot and humid countries such as Japan, primarily because of air-conditioning demands. Cold-energy storage and transport devices can potentially level the electric power load throughout day and night. Ice slurry that is a mixture of fine ice crystals and liquid water is a promising working fluid for such devices due to its good flowability and large latent heat of fusion. Although ice slurry is usually made from supercooled water, we encounter a few problems related to the supercooling of water; undesirable freezing may block the piping for transporting supercooled water, and a large degree of supercooling of water degrades the coefficient of performance (COP) of the refrigerator for cooling and freezing water. Therefore, a method to actively control the phase change from supercooled water to ice is necessary for practical use of ice-slurry systems. Several researchers

have clarified various factors that influence the phase change from supercooled water to ice and have tried to realize methods to control the freezing temperature [1–3]. However, an effective method for practical use has not yet been established.

To develop such a method, we focused on ultrasonic vibration to control the freezing temperature of supercooled water, because ultrasonic vibration is known to cause ice nucleation in supercooled water [4–8]. First, we experimentally investigated the effect of ultrasonic vibration on the phase change from supercooled water to ice. In the experiments, we measured the probability of the phase change, changing the cavitation intensity, for both pure water and tap water. Then, we simulated the phase change from supercooled water to ice induced by ultrasonic vibration and compared the results of the simulation to the experimental results.

## 2. Experiments

### 2.1. Apparatus

Fig. 1 shows a schematic of the experimental apparatus used to generate and observe the phase change from supercooled water to ice. A vessel (inner diameter

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Nomenclature	
$A_e$	rate of surface erosion, $\text{m}^2/\text{s}$
$a$	constant
$c$	constant
$f$	distribution function for number of bubble nuclei per unit volume
$I_u$	electrical current passing through an ultrasonic oscillator, A
$l$	latent heat, $\text{J}/\text{m}^3$
$m$	constant
$N$	number of experiments
$N_i$	number of bubble nuclei per unit volume in a specific range of normalized radius of bubble nuclei ( $i = 1, 2, 3, \dots$ ), $1/\text{m}^3$
$N_0$	total number of bubble nuclei per unit volume, $1/\text{m}^3$
$n$	number of bubble nuclei per unit volume in a unit range of radius of bubble nuclei, $1/\text{m}^4$
$p$	pressure, Pa
$p_A$	acoustic pressure amplitude, Pa
$p_{A \max}$	maximum acoustic pressure amplitude, Pa
$p_{cr}$	critical acoustic pressure amplitude for cavitation, Pa
$p_h$	atmospheric pressure, Pa
$p_v$	vapor pressure, Pa
$Q$	probability of phase change from supercooled water to ice
$Q_a$	probability of phase change from supercooled water to ice when ultrasonic vibration is applied
$Q_{cav}$	probability of cavitation with one bubble nucleus
$Q_s$	probability of phase change from supercooled water to ice without ultrasonic vibration
$Q_u$	probability of phase change from supercooled water to ice due to effect of ultrasonic vibration
$Q_{u0}$	probability of phase change from supercooled water to ice due to effect of ultrasonic vibration with one bubble nucleus
$R$	normalized radius of a bubble nucleus
$R_i$	normalized radius of a bubble nucleus ( $i = 1, 2, 3, \dots$ )
$r_g$	radius of a bubble nucleus, m
$r_{g \max}$	maximum measurable radius of bubble nuclei for an optical particle counter, m
$r_s$	critical radius for solid nucleation, m
$S_{td}$	standard deviation
$T_E$	equilibrium freezing temperature, K
$V$	volume of test water, $\text{m}^3$
$v$	growth rate of a solid nucleus, m/s
$v_0$	growth rate of an ice crystal, m/s
<i>Greek symbols</i>	
$\Delta R$	specific range of normalized radius of bubble nuclei
$\Delta T$	degree of supercooling, K
$\Delta T_m$	average maximum degree of supercooling, K
$\Delta T_{mi}$	average maximum degree of supercooling for an infinite number of experiments, K
$\Delta T_g$	critical degree of supercooling for crystal growth of a solid nucleus, K
$\Delta t$	duration of high pressure during the collapse of a bubble, s
$\lambda$	ultrasonic wavelength, m
$\sigma_{ls}$	surface energy at liquid–solid interface, N/m
$\sigma_{lg}$	surface energy at liquid–gas interface, N/m
<i>Subscripts</i>	
0	at atmospheric pressure
1	at high pressure during the collapse of a bubble

of 140 mm) equipped with an ultrasonic generator was filled with water. The frequency of the ultrasonic generator was 28 kHz and the output power was adjustable from 0 to 100 W (0 to  $6.5 \text{ kW}/\text{m}^2$ ). The frequency and power were chosen so that cavitation could occur in water. The temperature of the water was controlled by coolant from the lower sidewall. The upper part of the sidewall of the vessel was made of acrylic resin so that the phase-change process from supercooled water to ice could be observed by using a video camera. The temperature of the water was measured by thermocouples at four locations (A, B, C, D) as shown in Fig. 1. A heat-transfer plate made of copper was immersed in the water and cooled by coolant from its upper side. The distance between the lower surface of the heat-transfer plate and

the bottom of the vessel was 101 mm, which is twice as long as the ultrasonic wavelength corresponding to a water temperature of  $0^\circ\text{C}$ .

Fig. 2 shows the details of the heat-transfer plate. A circular groove (20 mm inner diameter, 13 mm wide, and 14 mm deep) was cut into the heat-transfer plate. We call the part within this groove the “center part”, and the other part the “surrounding part”. To make the temperature of the center part lower than that of the surrounding part during cooling, we filled the groove with a low thermal-conductivity material. Consequently, ice crystals always appeared from the surface of the center part when no ultrasonic vibration was used, indicating that the geometric influence of the outer rim of the heat-transfer plate on the phase change from

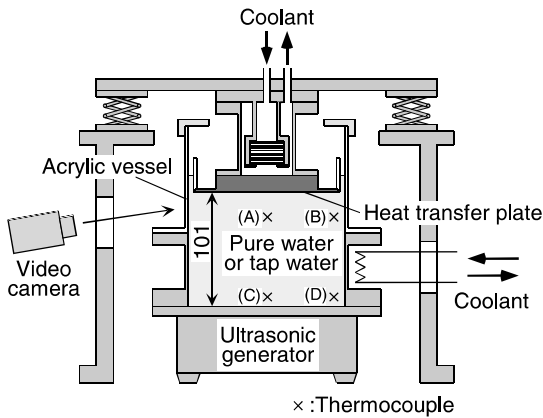


Fig. 1. Schematic of the experimental apparatus used to generate and observe the phase change from supercooled water to ice.

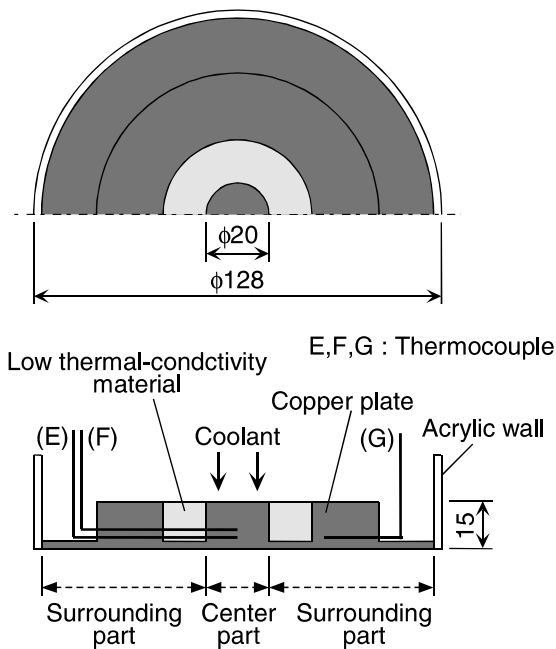


Fig. 2. Details of the heat-transfer plate.

supercooled water to ice was eliminated. Furthermore, we polished the surface of the center part with 60-grit sandpaper so that ice crystals were more apt to appear from the center part; the center part had a surface roughness corresponding to pits more than 10  $\mu\text{m}$  in depth, whereas the surrounding part had pits only about 1  $\mu\text{m}$  in depth. Thermocouples at (E) and (G) were inserted into the heat-transfer plate 2 mm from the surface, whereas thermocouple at (F) was 5.5 mm from the surface (Fig. 2). During the cooling process, tempera-

tures at (E) and (G) differed by more than 1 K. The temperature of the water in contact with the surface of the center part of the heat-transfer plate was assumed to be that indicated by the thermocouple at (E), because no significant difference between temperatures at (E) and (F) in the vertical direction was detected in the heat-transfer plate.

2.2. Procedures

Two types of experiments were done. In Exp. 1, the heat-transfer surface was cooled at a given cooling rate without ultrasonic vibration, until the supercooled water froze on the surface. The degree of supercooling at the moment the ice appeared was measured. In Exp. 2, ultrasonic vibration was applied to water at different degrees of supercooling (1.5, 3.0, 4.5, and 6.0 K) for 5 s. We used different cavitation intensities of ultrasonic vibration as mentioned later. We then determined if the supercooled water had changed to ice.

For both experiments, we used not only pure water but also tap water to determine the effectiveness of ultrasonic vibration in practical applications. The pure water was produced by ion exchange and filtering process (Millipore, Milli-Q Jr.). The specific resistance of the pure water ranged from 0.8 to 2.0  $\text{M}\Omega\text{ cm}$ , and solid particles larger than 0.2  $\mu\text{m}$  were filtered. The specific resistance of the tap water ranged from 2.6 to 2.9  $\text{k}\Omega\text{ cm}$ , and the water was not filtered.

For both experiments, before the cooling process, the temperature of the center part of the heat-transfer plate was maintained at 1.9°C, and the average temperature of the water at (A) and (B) was 2.3°C and that at (C) and (D) was 2.9°C. The heat-transfer plate was cooled by using coolant that was at -30°C. Natural convection did not affect these experiments because the upper portion of the water was cold and the lower part was hot during the cooling process and because the density of water decreases with decreasing temperature when the temperature is below 4°C. When the supercooled water changed to the solid phase on and/or near the surface of the heat-transfer plate, the temperature of the plate increased suddenly because of the release of latent heat from the supercooled water. For example, a cooling curve for Exp. 1 for the pure water is shown in Fig. 3. The phase change from supercooled water to ice was recognized by this temperature change as well as by observations from the video camera.

As shown in Fig. 3, the cooling rate was not constant during the cooling process. Therefore, in both Exp. 1 and Exp. 2, we defined the cooling rate as that for the temperature range from 0.0°C to -5.0°C, namely, 0.044  $\text{K/s} \pm 12\%$ .

Before each experiment, the ice on the heat-transfer surface was completely melted by heating the heat-

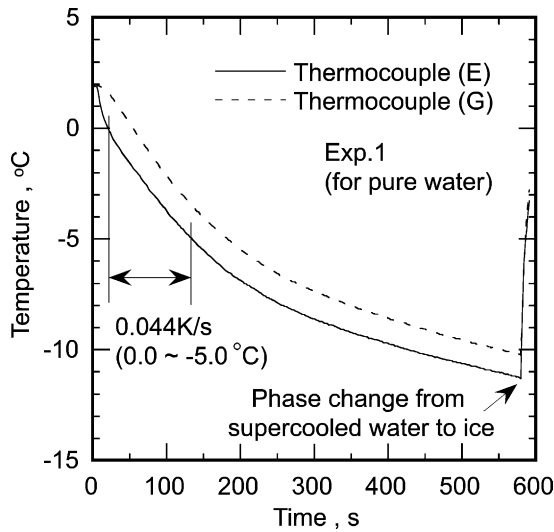


Fig. 3. Temperature change of the heat-transfer plate.

transfer plate, and then the visible bubbles that accompanied this melting process were removed from the surface by removing the heat-transfer plate from the water.

### 2.3. Measurement of cavitation intensity

To investigate the effect of the intensity of ultrasonic vibration on the phase change from supercooled water to ice, we needed to estimate the acoustic pressure amplitude caused by ultrasonic vibration. However, because accurate measurement of the acoustic pressure amplitude was generally difficult, we used the intensity of cavitation as an index of the acoustic pressure amplitude. We defined the intensity of cavitation due to ultrasonic vibration as the erosion loss of a 15- $\mu\text{m}$ -thick aluminum film that was attached to the heat-transfer surface. Measurement of this loss should be made under the same cooling conditions as the experiments. However, this was impossible because ice appeared on the aluminum film when ultrasonic vibration was used. Therefore, we measured the erosion loss at the water temperature at the initial conditions for three different levels of electric current passing through the ultrasonic oscillator,  $I_u$  (0.4, 0.6, and 0.8 A). The erosion area was measured by using image processing.

Fig. 4 shows the erosion area as a function of the duration of the applied ultrasonic vibration. As  $I_u$  was increased, the erosion area also increased. Based on these results, we defined the intensity of cavitation by the rate of surface erosion,  $A_e$ , as shown in the figure. Although no apparent erosion loss was evident at  $I_u = 0.4$  A from the image processing, the aluminum films showed many small dents.

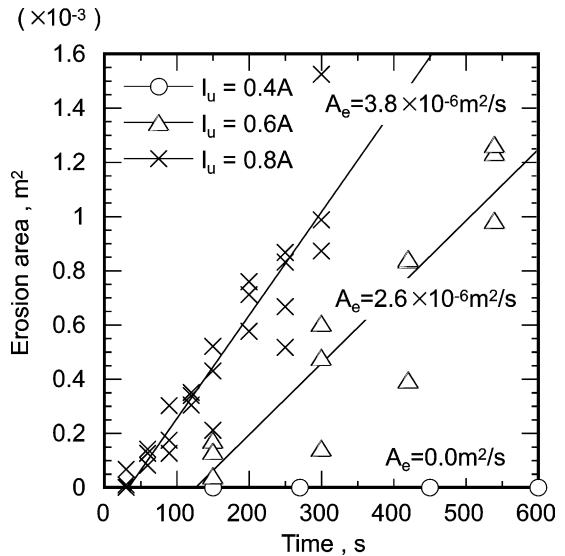


Fig. 4. Erosion area of an aluminum film, caused by ultrasonic vibration.

### 3. Simulation model

We developed a simulation model for the phase change from supercooled water to ice induced by ultrasonic vibration. In the model, we assumed that the phase change is caused by the collapse of a cavitation bubble. This collapse causes a momentary and local pressure increase, followed by an increase in the equilibrium freezing temperature of water, thereby causing nucleation of solids [4]. For simplicity, we assumed that cavitation bubbles always collapse.

According to the dynamic instability condition of a single bubble [9], the necessary condition for the occurrence of cavitation is given by

$$r_g \geq \frac{4\sigma_{lg}}{3(p_v - p)}, \quad (1)$$

where  $r_g$  is the radius of a bubble nucleus,  $\sigma_{lg}$  is the surface energy at the liquid–gas interface,  $p_v$  is the vapor pressure, and  $p$  is the pressure in water. Because ultrasonic vibration at a constant frequency forms a standing wave after repeated reflections within a vessel, the critical acoustic pressure amplitude for occurrence of cavitation,  $p_{cr}$ , is given by

$$p_{cr} = \frac{4\sigma_{lg}}{3r_g} - p_v + p_h, \quad (2)$$

where  $p_h$  is the atmospheric pressure.

The probability of occurrence of cavitation by ultrasonic vibration with an arbitrary bubble nucleus is defined as the probability that acoustic pressure amplitude at an arbitrary position is larger than  $p_{cr}$ . Although

the actual distribution of acoustic pressure amplitude is three-dimensional, a one-dimensional sinusoidal distribution is satisfactory to consider this probability (Fig. 5). We assume that a distribution of acoustic pressure amplitude is

$$p_A = p_{A \max} \left| \sin \frac{2\pi}{\lambda} x \right|, \quad (3)$$

where  $p_{A \max}$  is the maximum acoustic pressure amplitude and  $\lambda$  is the ultrasonic wavelength. Therefore, from Fig. 5, the probability of occurrence of cavitation by ultrasonic vibration with an arbitrary bubble nucleus is given by

$$Q_{\text{cav}} = 0 \left( p_{A \max} < p_{\text{cr}}; r_g < \frac{4\sigma_{\text{lg}}}{3(p_{A \max} + p_v - p_h)} \right), \quad (4a)$$

$$Q_{\text{cav}} = \frac{(\lambda/4) - (\lambda/2\pi) \sin^{-1}(p_{\text{cr}}/p_{A \max})}{\lambda/4} \\ = 1 - \frac{2}{\pi} \sin^{-1} \frac{(4\sigma_{\text{lg}}/3r_g) - p_v + p_h}{p_{A \max}} \\ \left( p_{A \max} \geq p_{\text{cr}}; r_g \geq \frac{4\sigma_{\text{lg}}}{3(p_{A \max} + p_v - p_h)} \right). \quad (4b)$$

The water near the cavitation bubble is subjected to an average pressure of about  $4 \times 10^9$  Pa during the collapse of the cavitation bubble [4,10]. Because the equilibrium freezing temperature,  $T_{E1}$ , is about 190°C (463 K) at this pressure [11] and the temperature increase due to adiabatic compression of the bubble is about 100 K [4], the estimated degree of supercooling,  $\Delta T_1$ , is more than 90 K when the temperature of bulk water is less than 0°C (273 K). The highest degree of supercooling of water determined experimentally is  $0.14T_{E1}$  [12], that is, 65 K. Therefore,  $\Delta T_1$  is larger than  $0.14T_{E1}$ , and thus the collapse of a cavitation bubble leads to instantaneous nucleation of solids.

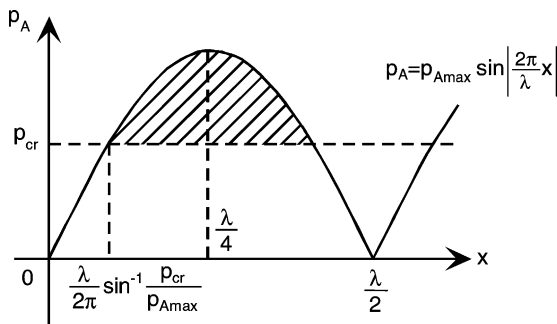


Fig. 5. One-dimensional distribution of pressure amplitude. The probability of occurrence of cavitation by ultrasonic vibration with an arbitrary bubble nucleus corresponds to the probability that pressure amplitude at an arbitrary position,  $p_A$  is larger than  $p_{\text{cr}}$ .

According to the classical theory for homogeneous nucleation [13], the critical radius of the solid nuclei during the collapse of a cavitation bubble is given by

$$r_{s1} = \frac{2\sigma_{\text{ls}}T_{E1}}{L(0.14T_{E1})}, \quad (5)$$

where  $\sigma_{\text{ls}}$  is the surface energy at the liquid–solid interface and  $L$  is the latent heat. However, the critical radius of the solid nuclei will return to its original value at atmospheric pressure,  $r_{s0}$ , after  $10^{-9}$  s, which is the duration of the high pressure [4,10]

$$r_{s0} = \frac{2\sigma_{\text{ls}}T_{E0}}{L\Delta T_0}, \quad (6)$$

where  $T_{E0}$  is the equilibrium freezing temperature at atmospheric pressure and  $\Delta T_0$  is the degree of supercooling with respect to  $T_{E0}$ . For visible phase change from supercooled water to ice, the solid nuclei must grow from  $r_{s1}$  to  $r_{s0}$  in radius within the duration of the high pressure,  $\Delta t$ . Let  $v$  denote the growth rate of the solid nuclei; then, for a visible phase change from supercooled water to ice,  $v$  must satisfy the following condition:

$$v\Delta t \geq r_{s0} - r_{s1}. \quad (7)$$

Let  $\Delta T_g$  denote the degree of supercooling with respect to  $T_{E0}$  that satisfies the equality portion in Eq. (7), then the probability of the phase change from supercooled water to ice due to the effect of ultrasonic vibration with an arbitrary bubble nucleus is given by

$$Q_{u0} = 0 \quad (\Delta T_0 < \Delta T_g), \quad (8a)$$

$$Q_{u0} = Q_{\text{cav}} \quad (\Delta T_0 \geq \Delta T_g). \quad (8b)$$

We now apply Eqs. (8a) and (8b) to bubble nuclei with a specified radius distribution. We normalize the radius of bubble nuclei by the maximum measurable value in the experiment

$$R = \frac{r_g}{r_{g \max}}, \quad (9)$$

where  $R$  is the normalized radius,  $r_g$  is the radius of bubble nuclei, and  $r_{g \max}$  is the maximum measurable value of the radius of bubble nuclei for an optical particle counter (Pacific Scientific, Hiac/Royco PA-720) used here. Let  $f$  denote the distribution function for the number of bubble nuclei per unit volume ( $0 \leq f \leq 1$ )

$$f = \int_0^{r_g} \frac{n}{N_0} dr_g = \frac{r_{g \max}}{N_0} \int_0^R n dR, \quad (10)$$

where  $n$  is the number of bubble nuclei per unit volume in a unit range of radius of bubble nuclei, and  $N_0$  is the total number of bubble nuclei per unit volume. According to the existing data [14],  $\log n$  is approximately proportional to  $\log r_g$  when the radius of bubble nuclei is limited to between  $1 \times 10^{-6}$  and  $1 \times 10^{-4}$  m. Therefore  $f$  can be approximated by

$$\frac{df}{dR} = CR^m, \tag{11}$$

where  $C$  and  $m$  are constants. Fig. 6 shows  $df/dR$  for the pure water that we used, as measured by using the optical particle counter. In this measurement,  $r_{g \max}$  was  $2.08 \times 10^{-5}$  m. Although bubble nuclei and solid particles cannot be distinguished by this particle counter, the number of solid particles was negligible in the pure water that we used here [15]. The constants  $C$  and  $m$  in Eq. (11) were  $6.02 \times 10^{-3}$  and  $-2.93$ , respectively.

Therefore, when we consider the radius distribution of bubble nuclei, the probability of the phase change from supercooled water to ice due to the effect of ultrasonic vibration is given by

$$Q_u = 1 - \{1 - Q_{u0}(r_{g1})\}^{N_1 V} \{1 - Q_{u0}(r_{g2})\}^{N_2 V} \dots \{1 - Q_{u0}(r_{gi})\}^{N_i V} \dots, \tag{12}$$

where

$$N_i = N_0 \Delta R \left( \frac{df}{dR} \right)_{R=R_i}, \tag{13}$$

$$R_i = (i - 1) \Delta R \tag{14}$$

and  $\Delta R$  is a specific range of the normalized radius of bubble nuclei, which is  $5.0 \times 10^{-7}/r_{g \max}$  in our simulation.  $V$  in Eq. (12) represents the volume of the water. For the conditions of our simulation, we consider only a range of bubble nuclei from  $1 \times 10^{-6}$  to  $1 \times 10^{-4}$  m, because  $Q_{cav}$  is negligible for bubble nuclei less than

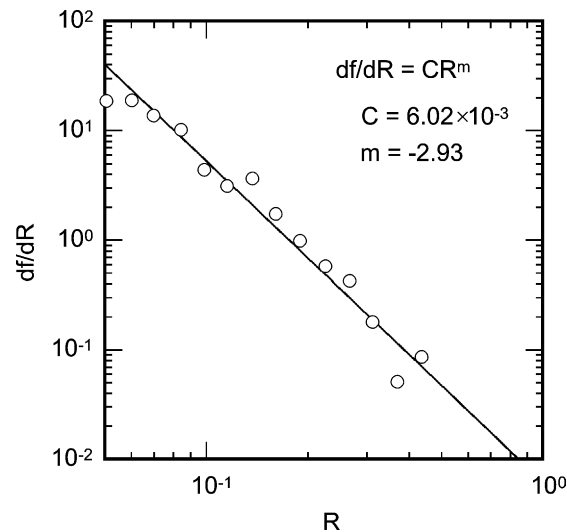


Fig. 6.  $df/dR$  for the pure water used in the experiment.  $f$  is the distribution function for the number of bubble nuclei per unit volume and  $R$  is the normalized radius.

$1 \times 10^{-6}$  m in radius and  $df/dR$  is negligible for bubble nuclei more than  $1 \times 10^{-4}$  m in radius.

In our experiments, the water had a temperature distribution in the vessel. We calculated this distribution from the cooling curve of the heat-transfer surface, neglecting the convection in the water and assuming a one-dimensional temperature distribution perpendicular to the heat-transfer surface. Then, using this temperature distribution, we calculated the probability of the phase change from supercooled water to ice due to the effect of ultrasonic vibration.

If we define the probability of the phase change from supercooled water to ice without ultrasonic vibration as  $Q_s$ , then the probability of the phase change when ultrasonic vibration is applied is given by

$$Q_a = 1 - (1 - Q_u)(1 - Q_s). \tag{15}$$

In the simulation, we used  $Q_s$  obtained from our experimental results.

#### 4. Results and discussion

##### 4.1. Pure water

Fig. 7 shows the frequency distribution of the maximum degree of supercooling in Exp. 1 involving the pure water. A total of 52 measurements was made. The maximum degree of supercooling ranged from 9.5 to 13.5 K, with an average value,  $\Delta T_m$ , of 11.6 K and standard deviation,  $S_{td}$ , of 0.72.  $\Delta T_m$  for infinite measurements,  $\Delta T_{mi}$ , for a 99% confidence level is given by

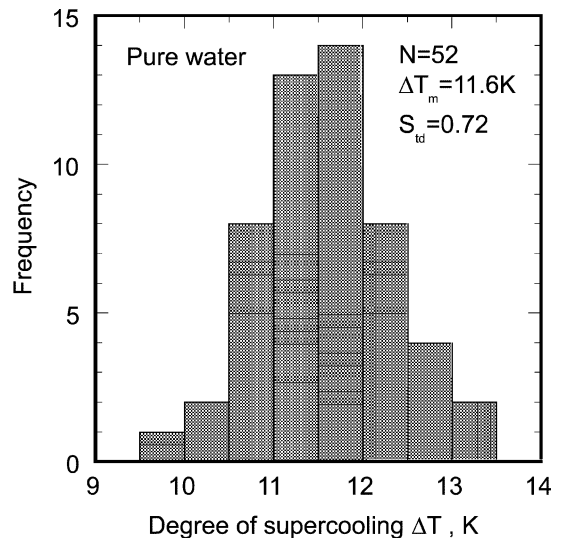


Fig. 7. Frequency distribution of the maximum degree of supercooling of pure water.

$$\Delta T_m - 2.58 \frac{S_{id}}{\sqrt{N}} < \Delta T_{mi} < \Delta T_m + 2.58 \frac{S_{id}}{\sqrt{N}}, \quad (16)$$

where  $N$  represents the number of measurements. Here, we determined  $N$  by using Eq. (16) so that the value of  $\Delta T_{mi}$  was within the range of  $\Delta T_m \pm 0.5$  K at a 99% confidence level.

In Exp. 2 involving the pure water, ultrasonic vibrations of different cavitation intensities were used at  $\Delta T$  of 3.0, 4.5, and 6.0 K. We made 25 measurements for each experimental condition. Fig. 8 shows the probability of the phase change from supercooled water to ice,  $Q$ , as a function of  $\Delta T$  for both Exp. 1 and Exp. 2. The result of Exp. 1 is shown as the cumulative distribution of Fig. 7. Comparison of the results of Exp. 1 and Exp. 2 indicates that ultrasonic vibration was effective for promoting the phase change from supercooled water to ice. The results of Exp. 2 indicate that  $Q$  approached 1 as the cavitation intensity increased. These results suggest the possibility of active control of the freezing temperature of supercooled pure water by using ultrasonic vibration.

We found that the phase change from supercooled water to ice on and/or near the heat-transfer surface started not only from the center part, but also from the surrounding part when  $\Delta T$  was either 4.5 or 6.0 K, because the surrounding part of the heat-transfer plate also

became supercooling state as shown in Fig. 3. On the other hand, when  $\Delta T$  was 3.0 K, the phase change always started from the center part, because the degree of supercooling at the surrounding part was about 1 K and it was too small to cause the phase change from supercooled water to ice. When the phase change from the surrounding part was observed, the phase change from the center part was always observed almost simultaneously. This indicates that the data shown in Fig. 8 were obtained as the results of the phase change from the center part.

The generation of ice on the heat-transfer surface was indistinguishable from that in bulk supercooled water from the observation, because the supercooled layer was just 5-mm thick according to the simulation even when the degree of supercooling at the heat-transfer surface was 6 K.

The simulation results are also shown in Fig. 8. To fit the simulation results to the experimental results, we adjusted two parameters, that is, the growth rate of the solid nuclei,  $v$ , and the maximum acoustic pressure amplitude,  $p_{A \max}$ . Although it is difficult to estimate  $v$  in Eq. (7), the free-growth rate of an ice crystal,  $v_0$ , can be approximated for small degrees of supercooling less than 6 K by the following empirical equation [4,16]:

$$v_0 = 2.8 \times 10^{-4} \Delta T^{2.3}. \quad (17)$$

To fit the simulation results to the experimental results, we set  $v$  equal to  $av_0$  and then determined the coefficient,  $a$ . Although precise measurement of the acoustic pressure amplitude in water was not done, the maximum acoustic pressure amplitude measured by using an ultrasonic pressure sensor was approximately  $1 \times 10^5$  Pa. Therefore, in the simulation, we set  $p_{A \max}$  to be around  $1 \times 10^5$  Pa.

By optimizing the two parameters,  $v$  and  $p_{A \max}$ , the simulation results agree with the experimental results. This agreement indicates that the phase change from supercooled water to ice induced by ultrasonic vibration could be simulated by assuming that the collapse of cavitation bubbles causes nucleation of solids, if the two parameters,  $v$  and  $p_{A \max}$ , are obtained properly.

The simulation results show that the probability of the phase change from supercooled water to ice induced by ultrasonic vibration is greatly influenced by the degree of supercooling, although the cavitation phenomenon is not influenced very much by the degree of supercooling. This is due to the change of the temperature distribution in the water. Because the thickness of the supercooled layer increases as the degree of supercooling at the heat-transfer surface increases, the probability of the phase change from supercooled water to ice also increases as the degree of supercooling.

According to Eq. (15), the simulation results can be divided into two components,  $Q_u$  and  $Q_s$ . Fig. 9 shows

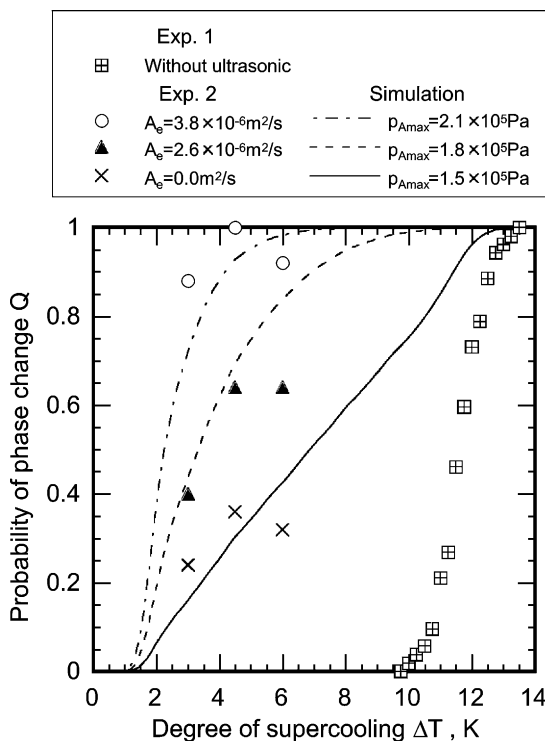


Fig. 8. Probability of the phase change from supercooled pure water to ice as a function of degree of supercooling.

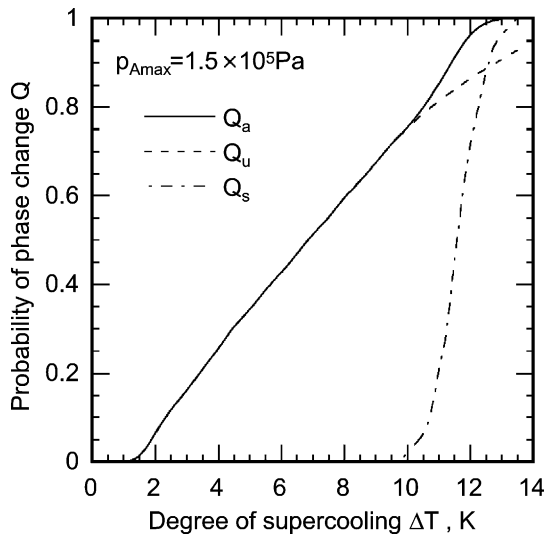


Fig. 9. Simulation probability of the phase change of pure water and its two components,  $Q_u$  and  $Q_s$ .

the simulation  $Q_a$  and these two components when  $p_{A \max} = 1.5 \times 10^5$  Pa and  $Q_s$  is based on the results of Exp. 1 for the pure water. These simulation results show that the influence of  $Q_s$  emerges only when  $\Delta T > 10$  K.

#### 4.2. Tap water

Although we demonstrated the effectiveness of ultrasonic vibration for actively controlling the freezing temperature of supercooled pure water, for practical applications we must also demonstrate its effectiveness for tap water. We therefore repeated the same experiments for tap water.

Based on Eq. (16), we made 30 measurements in Exp. 1 so that  $\Delta T_{mi}$  was within the range of  $\Delta T_m \pm 0.5$  K at a 99% confidence level.  $\Delta T_m$  for the tap water was 9.0 K, which is lower than that for the pure water (11.6 K) due to the solid particles larger than  $0.2 \mu\text{m}$  and the ions present in the tap water.

In Exp. 2, ultrasonic vibrations of different cavitation intensities were applied at  $\Delta T$  of 1.5, 3.0, and 4.5 K. We made 10 measurements for each experimental condition. Fig. 10 shows  $Q$  as a function of  $\Delta T$  for both Exp. 1 and Exp. 2. Comparison of the results of these two experiments reveals that, similar to that for the supercooled pure water, ultrasonic vibration is effective for controlling the freezing temperature of the supercooled tap water.

We found that the phase change from supercooled water to ice on and/or near the heat-transfer surface started not only from the center part, but also from the surrounding part when  $\Delta T$  was 4.5 K, because the surrounding part of the heat-transfer plate also became

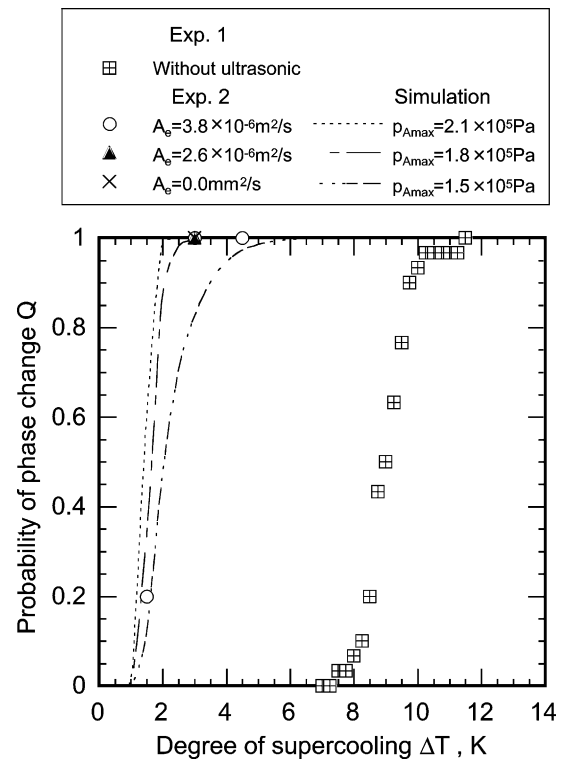


Fig. 10. Probability of the phase change from supercooled tap water to ice as a function of degree of supercooling.

supercooling state as shown in Fig. 3. On the other hand, when  $\Delta T$  was either 3.0 or 1.5 K, the phase change always started from the center part, because the degree of supercooling at the surrounding part was too small to cause the phase change from supercooled water to ice. In the same way as for the pure water, the data shown in Fig. 10 were obtained as the results of the phase change from the center part.

The simulation results for the tap water are also shown in Fig. 10. The number of bubble nuclei contained in the tap water was higher than that in the pure water, because many solid particles remained in the tap water, thus trapping the bubble nuclei on their surfaces. It is difficult to measure the number of bubble nuclei in the tap water by using an optical particle counter, because the particle counter cannot distinguish between the bubble nuclei and solid particles. Therefore, in the simulation of the tap water, we used a value of  $C$  in Eq. (11) that was 10 times higher than that for the pure water so that the simulation results fit the experimental results. The simulation results agree with the experimental results, similar to the result for the pure water.

Figs. 8 and 10 show that  $Q$  for the tap water is higher than that for the pure water under the same experimental conditions in Exp. 2. The difference in the specific resistance between the pure water and tap water



does not influence  $Q$ , because the specific resistance does not influence the cavitation phenomena. Therefore, according to the simulation results, this higher probability for the tap water is due to the large number of bubble nuclei in the tap water sustained by a high concentration of solid particles.

## 5. Conclusions

The effect of ultrasonic vibration on the phase change from supercooled water to ice on and/or near a heat-transfer surface was experimentally investigated both for pure water and for tap water. Then, the phase change from supercooled water to ice induced by ultrasonic vibration was simulated based on the cavitation phenomena. Our findings are summarized as follows:

1. Ultrasonic vibration strongly promotes the phase change from supercooled water to ice, for both pure water and tap water. The experimental results agreed well with the simulations based on a model that considers the cavitation phenomena.
2. A reliable method to actively control the freezing temperature of supercooled water by ultrasonic vibration can be realized when the appropriate cavitation intensity is chosen.

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## References

- [1] T. Shichiri, Y. Araki, Nucleation mechanism of ice crystals under electrical effect, *J. Cryst. Growth* 78 (1986) 502–508.
- [2] A. Saito, Y. Utaka, S. Okawa, K. Matsuzawa, A. Tamaki, Fundamental research on the supercooling phenomenon on heat transfer surfaces – investigation of an effect of characteristics of surface and cooling rate on a freezing temperature of supercooled water, *Int. J. Heat Mass Transfer* 33 (1990) 1697–1709.
- [3] H. Inaba, K. Takeya, S. Nozu, Fundamental study on continuous ice making using flowing supercooled water, *JSME Int. J. B* 37 (1994) 385–393.
- [4] R. Hickling, Nucleation of freezing by cavity collapse and its relation to cavitation damage, *Nature* 206 (1965) 915–917.
- [5] J.D. Hunt, K.A. Jackson, Nucleation of solid in an undercooled liquid by cavitation, *J. Appl. Phys.* 37 (1966) 254–257.
- [6] C.P. Lee, T.G. Wang, The effects of pressure on the nucleation rate of an undercooled liquid, *J. Appl. Phys.* 71 (1992) 5721–5723.
- [7] K. Ohsaka, E.H. Trinh, Dynamic nucleation of ice induced by a single stable cavitation bubble, *Appl. Phys. Lett.* 73 (1998) 129–131.
- [8] T. Hozumi, S. Saito, S. Okawa, Effect of ultrasonic waves on freezing of supercooled water, in: K. Hutter, Y. Wang, H. Beer (Eds.), in: *Advances in Cold-Region Thermal Engineering and Sciences*, Springer, Berlin, 1999, pp. 65–72.
- [9] P. Dergarabedian, P. Calif, The rate of growth of vapor bubbles in superheated water, *Trans. ASME, J. Appl. Mech.* 20 (1953) 537–545.
- [10] R. Hickling, M.S. Plesset, Collapse and rebound of a spherical bubble in water, *Phys. Fluids* 7 (1964) 7–14.
- [11] American Institute of Physics Handbook, second ed., McGraw-Hill, New York, 1957, p. 4.39.
- [12] D. Turnbull, Formation of crystal nuclei in liquid metals, *J. Appl. Phys.* 21 (1950) 1022–1028.
- [13] N.H. Fletcher, *The Chemical Physics of Ice*, first ed., Cambridge University Press, London, 1970, pp. 85–97.
- [14] R.S. Meyer, M.L. Billet, J.W. Holl, Freestream nuclei and traveling bubble cavitation, *Trans. ASME, J. Fluids Eng.* 114 (1992) 672–679.
- [15] X. Zhang, T. Inada, A. Yabe, S. Lu, Y. Kozawa, Active control of phase change from supercooled water to ice by ultrasonic vibration 2. Generation of ice slurries and effect of bubble nuclei, *Int. J. Heat Mass Transfer* 44 (2001) 4533–4539.
- [16] C.S. Lindenmeyer, G.T. Orrok, K.A. Jackson, B. Chalmers, Rate of growth of ice crystals in supercooled water, *J. Chem. Phys.* 27 (1957) 822.