PERGAMON

International Journal of Heat and Mass Transfer 43 (2000) 333–339

International Journal of HEAT and MASS TRANSFER

www.elsevier.com/locate/ijhmt

Crystal ice formation of solution and its removal phenomena at cooled horizontal solid surface
Part I: ice removal phenomena

Tetsuo Hirata[a,*], Koji Nagasaka[b], Masaaki Ishikawa[a]

[a]*Department of Mechanical Systems Engineering, Shinshu University, 500 Wakasato, Nagano 380-8553, Japan*
[b]*Shinryo Reinetsu Co. Ltd., 2-4 Yotsuya, Shinjuku-ku, Tokyo 160-8510, Japan*

Received 24 July 1998; received in revised form 12 May 1999

Abstract

Experimental studies for the freezing phenomenon of ethylene glycol solution on a cooled plate have been performed. A polyvinyl chloride as well as an acrylic resin is used as a cooled plate. It is found that the crystal ice formed on the cooled plate is removed from the plate surface due to the buoyancy force acting on the crystal ice. This means that ice formation on a cooled plate without a deposited ice layer is possible by the present method. It is shown that the cooled plate surface is supercooled about 1.0°C below the freezing temperature of the solution during the crystal ice formation and its removal phenomena. The degree of supercooled temperature is unaffected by the cooling temperature of the plate. For higher solute concentrations, it is found that the number of the removed crystal ice per unit of time is increased and the volume of each removed ice is decreased. © 1999 Elsevier Science Ltd. All rights reserved.

---

1. Introduction

When ice is accumulated on a cold wall, the ice layer, in general, deposits on the cold wall and the ice formation rate decreases due to the decrease of thermal conductance of the deposited ice layer. This is a well known phenomenon and is a distinguishing characteristic in the freezing process of water. For a cold thermal energy storage system using ice, this characteristic may be a disadvantage for the efficiency of ice formation rate. For example, in a static-type ice storage system, the cold thermal energy storage rate decreases with ice growth. Authors have tackled to find out a method to make ice without a deposited ice layer on a cold wall. In the previous paper [1], the ice formation phenomenon of a solution at the cooled liquid interface was observed as a first step of the present study. It was found that a number of ice crystals in the shape of thin leaf were formed at the interface and were removed due to the buoyancy force acting on the ice. The removed ice, which rose toward the surface of the solution, was ascertained to be liquid-like ice. It was also observed that the ice formation and its removal phenomena occurred continuously, which implied the possibility of application of the present method to the ice storage device. In the advanced experiment by authors [2], it was found that the same phenomenon was also observed at a cooled solid surface.

In the present paper, the freezing phenomenon of ethylene glycol solution on a polyvinyl chloride plate as well as an acrylic resin plate is observed. The ice

---

* Corresponding author.
E-mail address: hirata@skip.shinshu-u.ac.jp (T. Hirata)

PII: S0017-9310(99)00168-4

Nomenclature

a	thermal diffusivity
A	heat transfer area of cooled plate
C	solute concentration
Cp	specific heat
L	latent heat of freezing
m	total ice volume
Ste	Stefan number
t	time
T_f, T_s, T_w	freezing, plate surface and plate rear surface temperatures
$\bar{T}_s$	mean temperature of cooled plate surface
q	heat flux
y_1	cooled plate thickness

Greek symbols

θ_s	dimensionless mean temperature of plate surface
λ	thermal conductivity
ρ	density
τ	dimensionless time

Subscripts

1	cooled plate
i	ice

formation and the ice removal characteristics are examined experimentally.

2. Experimental apparatus and procedure

Experimental apparatus consisted of a test section, the temperature-controlled room, the temperature-controlled bath and the recirculation system of the cold brine. The test section made of 15 mm-thick acrylic resin plate had the dimensions 110 × 80 × 100 mm inner volume. At the bottom of the test section, the test plate was installed horizontally. As a test plate, a polyvinyl chloride and an acrylic resin were used. The test plate was cooled with the copper plate having the dimensions of 50 × 80 mm in heat transfer area and 3 mm in thickness. The copper plate was installed on the rear surface of the test plate with an adhesive agent, as shown in Fig. 1. The ethylene glycol solution was filled in the upper region of the test plate and the ice formation was made with cooling the rear side of the copper plate with the cold brine. To avoid the supercooling of the solution at the beginning of ice formation experiment, the surface of the test plate was touched with the thermocouple inserted from the pipet installed at the top of the test section. To minimize the heat flow from the surroundings to the test section, the test section was placed in the temperature-controlled room, where the temperature was controlled around the freezing temperature of the solution. The ice for-

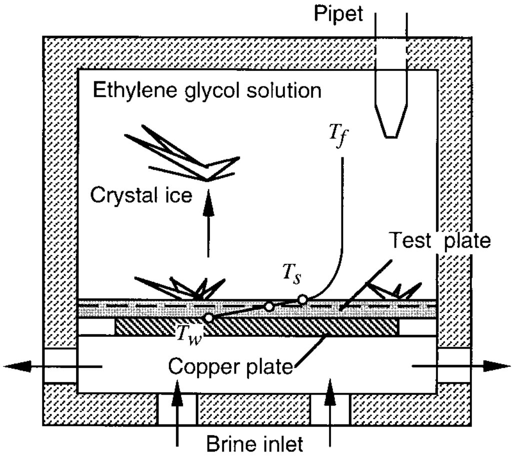

Fig. 1. Experimental apparatus and nomenclature.

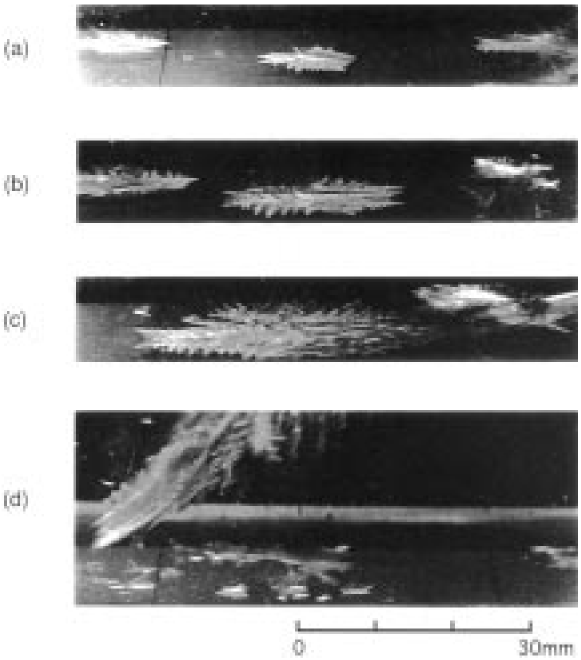

Fig. 2. Ice formation phenomenon ($C = 10.0$ wt%, $T_w = -13.8$°C, $y_1 = 3$ mm, acrylic resin plate).

mation phenomenon was observed after the temperature of the solution was cooled down at its freezing temperature.

For the measurement of the plate surface temperature, 3 mm-thick test plate was made of two plates having a thickness of 1 and 2 mm, respectively. A pair of 0.1 mm-diameter thermocouples was installed between the two plates and the rear surface of the test plate, respectively. The surface temperature of the test plate, T_s, was estimated from the two temperatures by an extrapolation method. The volume of ice formation was measured using the relation of density difference between the solution and ice, which was detected as a change in the height of the solution surface in the pipet. The experimental ranges covered in the present experiment were $-5°C < T_w < -15°C$ for the rear temperature of the test plate and $C = 2.0$, 4.6, 7.0 and 10.0 wt% for the concentration of the ethylene glycol solution.

3. Results and discussion

3.1. Freezing phenomena

Fig. 2 shows a series of photographs for the ice formation and its removal phenomena, where the concentration of the ethylene glycol solution is 10 wt% and the test plate is the acrylic resin of $y_1 = 3$ mm in thickness. As can be seen in Fig. 2, 3-D ice crystals are formed on the cooled plate at various locations. When the crystal ice has grown in a certain dimension, it begins to be removed from the plate and rises in the solution towards the top of the test section due to the buoyancy force acting on the crystal ice. These ice formation and removal phenomena occur continuously. Fig. 2(a) shows the crystal ice immediately after the nucleation at the plate surface. At the first stage of ice formation, the crystal ice nucleates on the cooled plate as a pinpoint of ice. After that, the ice grows in the shape of a thin leaf leaving a very little clearance on the cooled plate. It was observed that the ice could slide and move on the cooled plate with the very weak force when touched with the thin wire. This implies that the cohesion area between the crystal ice and the cooled plate is very small. In Fig. 2(b) and (c), it is shown that the crystal ice continues to grow with maintaining the shape of flat and thin leaf. When the crystal ice has grown in a certain dimension, the ice removal phenomenon occurs, as shown in Fig. 2(d). In the present experiment, the nucleation of crystal ice occurred randomly at various locations on the cooled plate. It was observed that if the neighboring ice crystals had not been removed until they came in touch with each other, they joined at the ice front by freezing. In Fig. 2(d), it is shown that many of the ice crystals nucleate quickly on the plate surface after ice removal. In the preliminary experiment, the ice was formed on the acrylic resin plate whose surface was scratched with sandpaper. It was observed that the crystal ice was not removed from the scratched surface. This result implies that a smooth surface is a necessary condition for the ice removal phenomenon.

(a) C=2.0wt%, T_w=-9.4℃.

(b) C=10.0wt%, T_w=-13.5℃.

Fig. 3. Effect of solute concentration on ice formation phenomenon (polyvinyl chloride, $y_1 = 2$ mm): (a) $C = 2.0$ wt%, $T_w = -9.4°C$; (b) $C = 10.0$ wt%, $T_w = -13.5°C$.

Fig. 3 shows the effect of concentration of the ethylene glycol solution on the ice formation phenomenon. In this case, 2 mm-thick test plate was used. At the top of the test section, the removed ice is piled up and is ascertained to be liquid-like ice. Fig. 3(a) is the result for the concentration of $C = 2.0$ wt%. In this case, the characteristic of the crystal ice is comparatively thick, hard and transparent. Therefore, the structure of the ice crystal can be clearly observed. On the other hand for the high concentration of $C = 10.0$ wt% shown in Fig. 3(b), the characteristic of the crystal ice is soft,

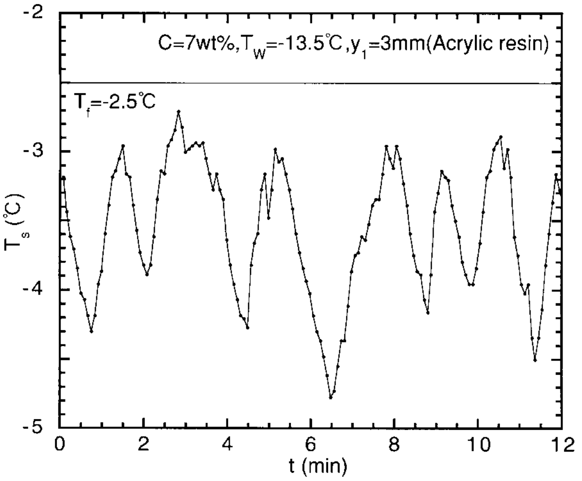

Fig. 4. Temperature variation of cooled plate surface with ice formation.

sparse and cloudy. These differences in the ice characteristic are caused by an increase of solute extraction with increasing concentration during ice formation.

Fig. 4 shows the temperature variation of the cooled plate surface during the ice formation and its removal experiments for the concentration of $C = 7$ wt%. It is shown that the surface temperature, T_s, is supercooled below the freezing temperature of the solution, $T_f = -2.5°C$. It is realized that the temperature variation is related to the ice formation and its removal phenomena. The increase of the surface temperature is due to a discharge of latent heat of freezing with ice growth and the decrease is due to a cooling of the test plate after the ice removal. In Fig. 4, it is seen that the variation of the surface temperature is irregular within a certain temperature range. This is because the ice formation and its removal phenomena occur at random locations on the cooled plate and do not necessarily occur at the location where the thermocouple is installed.

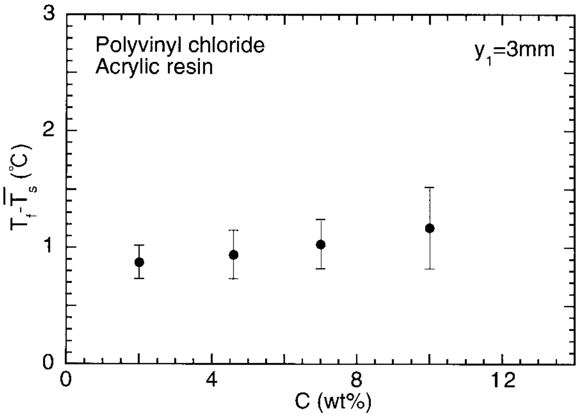

Fig. 6. Mean temperature of cooled plate surface.

Fig. 5 shows the maximum and the minimum values of the temperature variations at the cooled surface of polyvinyl chloride plate for $C = 4.6$ wt%. It is seen that the range of the temperature variation is within 1.0°C below the freezing temperature and is independent of the rear surface temperature of the cooled plate, T_w. This means that the crystal ice nucleates when the surface temperature is supercooled at a certain constant temperature. It should be noted that the independence of the surface temperature from T_w is a distinctive feature of the present ice formation phenomenon. In Fig. 5, it is seen that the maximum values of the plate surface temperature exceed the freezing temperature, $T_f = -1.6°C$. This is caused by an experimental error when the surface temperature of

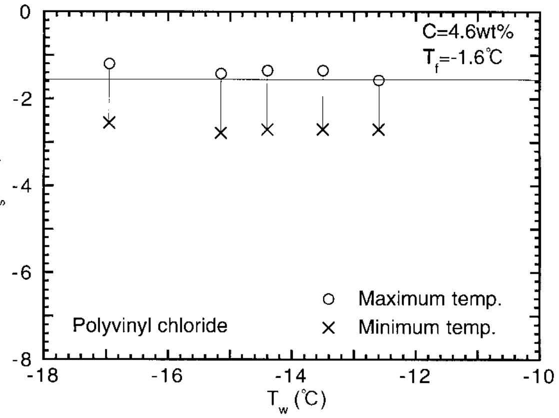

Fig. 5. Range of temperature variation of cooled plate surface.

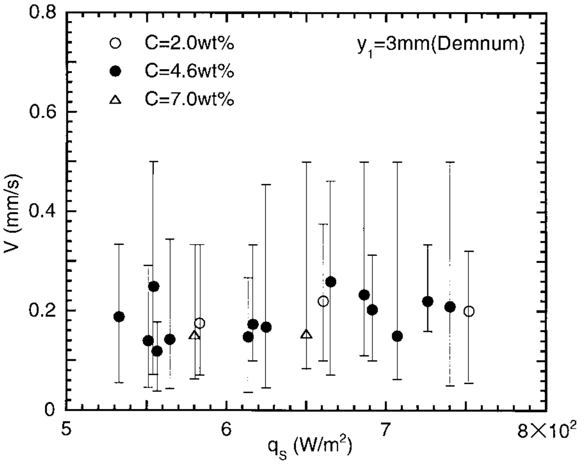

Fig. 7. Ice growth velocity.

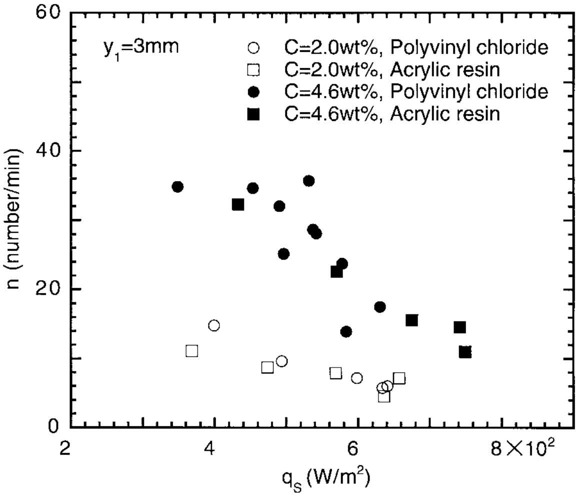

Fig. 8. Number of removed crystal ice per unit time (A = 50 × 80 mm).

the test plate, T_s, was estimated from the inside and the rear surface temperatures of the test plate.

Fig. 6 shows the mean surface temperature of the cooled plate, $\bar{T}_s$, versus the solute concentration. The data are scattered in the ranges shown in Fig. 6 and the mean values of the scattered data are shown with solid circles. In Fig. 6, the mean surface temperatures for the polyvinyl chloride plate as well as for the acrylic resin plate are plotted. In the present experimental conditions, the mean surface temperatures for those plates are not different. This is due to the fact that the thermal diffusivities of those plates are the same as $a_l = 0.12 \times 10^{-6}$ m^2/s. It is seen that the mean surface temperature, $\bar{T}_s$, decreases slightly with increasing concentration. This is caused by the increase of solute extraction on the cooled solid surface with increasing concentration, which results in higher concentration and lower freezing temperature for the solution near the cooled plate surface.

In the preliminary experiment to find out the ice removal conditions, Demnum (fluoric oil, $\rho = 1895$ kg/m^3, $\nu = 8 \times 10^{-5}$ m^2/s, $\lambda = 0.148$ W/m K at 0°C) was used as a cooled liquid instead of the test plate [1], and the ice removal phenomenon was observed. Fig. 7 shows the ice growth velocity at the crystal ice front for Demnum. The mean heat flux in the cooled liquid is given by

$$q_s = \lambda_l \frac{\bar{T}_s - T_w}{y_l} \tag{1}$$

where λ_l and y_l are the thermal conductivity and the thickness ($y_l = 3$ mm) of the cooled liquid, respectively. The ice growth velocity was measured by observation, upto 50 ice crystals under the same heat flux conditions. In Fig. 7, the ranges of scattered data of ice growth velocity and its mean values versus the cooling heat flux are shown. Though the present measurement method is not accurate enough, it is seen that the ice growth velocity is almost constant at 0.2 mm/s. It is known that the growth velocity of dendritic ice in a supercooled solution decreases with concentration and increases with the degree of supercooling [3]. In the present results, however, the observed ice growth velocity is almost constant for the various concentrations. The ice formation phenomenon in the present experiment occurs under the condition that the degree of supercooling increases with increasing concentration, as shown in Fig. 6. Therefore, the effect of both concentration and degree of supercooling on the ice growth velocity may be canceled with each other. It can be noted that the characteristic of the constant ice growth velocity for various concentrations is a distinctive feature in the present ice formation phenomenon.

Fig. 8 shows the number of removed ice per unit of time, n, versus the mean heat flux through the cooled plate. The large ice formed by joining the small ice crystals was counted as one piece. In the experiment, when the large ice was removed from the cooled plate, a number of small ice crystals of dimension: less than 2 mm-diameter nucleated and floated from the plate surface due to the accompanying flow with the removal of large ice. These small ice crystals were neglected because of small volumes as compared to that of the large one. From Fig. 8, it is seen that n increases with decreasing heat flux. This is the result that the crystal ice is removed independently for smaller heat fluxes and the joining of crystal ice occurs for larger heat fluxes. The possibility of the joining of crystal ice becomes higher for larger heat fluxes, because the density of the ice nucleation on the cooled plate increases with heat flux. It is also seen that the values of n are almost the same for the two kinds of cooled plates, the polyvinyl chloride and the acrylic resin, because of the same thermal diffusivities.

The effect of solute concentration on n is also shown in Fig. 8. In the experiment under the same heat fluxes, it was observed that the volume of each removed ice decreased for higher concentrations and, therefore, n increased as compared to that for lower concentrations. This is caused by a fragile nature of the ice formed in higher concentrations. It was also observed that the fragile crystal ice formed on the cooled plate was easily broken with the buoyancy force acting on the ice. These phenomena occurred more frequently for higher concentrations, and the crystal ice less than 2 mm in diameter increased in large quantities. The measurement of n for 7.0 and 10.0 wt% solutions was, therefore, impossible in the present experiment. On the other hand, for lower concentrations, the crystal ice

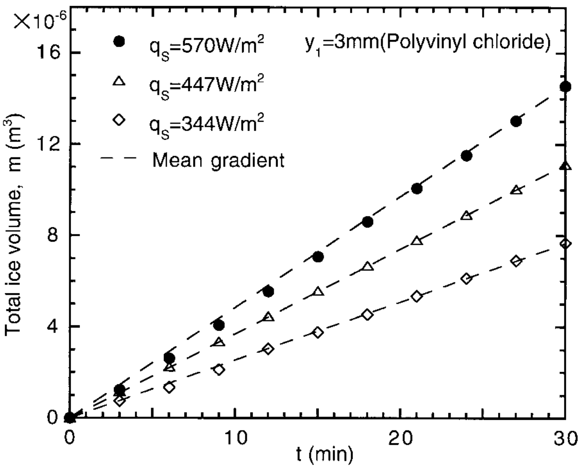

Fig. 9. Ice formation rate with time.

was firm and was removed by the buoyancy force after the growth in comparatively large dimensions.

As can be realized from Fig. 8, the ice removal frequency decreases with increasing heat flux and the ice removal phenomenon does not to occur at a certain value of heat flux. The critical heat flux is about $q_s = 700–800\ \mathrm{W/m^2}$ for the acrylic resin and the polyvinyl chloride plates, in the present experimental conditions. It is considered that the ice removal phenomenon occurs when the buoyancy force acting on the crystal ice becomes larger than the cohesion force between the ice and the cooled plate. The onset condition for the ice removal phenomenon will be discussed in Part II of this paper.

3.2. Measured ice volume

Fig. 9 shows the change of the total ice volume with time for various cooling heat fluxes. As can be seen in Fig. 9, the total ice volume increases linearly with time, that is, the ice formation rate per unit of time is constant. This implies that the present ice formation phenomenon can be considered, in a viewpoint of macro time scale, to be in a steady-state condition. The linear increase of the ice volume is a distinguishing characteristic in the present ice formation phenomenon as compared to the conventional one, in which an ice layer deposits on a cooled plate and the ice formation rate per unit of time decreases with ice growth.

To estimate the formed ice volume analytically, it is assumed that the change of solute concentration with ice formation is negligibly small. Then, the heat balance equation at the cooled surface is given by the relation that the latent heat discharged by the ice formation is equal to the heat rate conducted through the cooled plate.

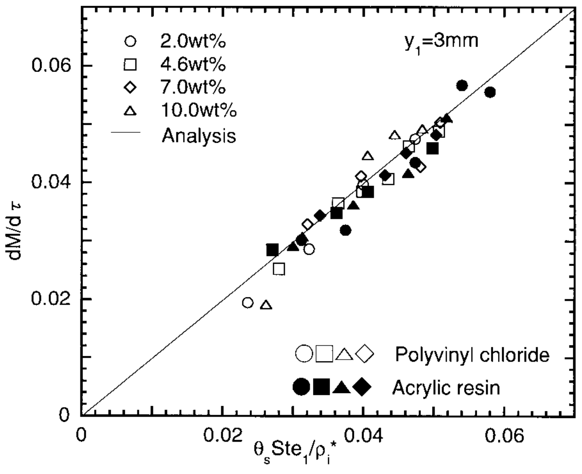

Fig. 10. Dimensionless ice formation rate.

$$\rho_i L \frac{\mathrm{d}m}{\mathrm{d}t} = A\lambda_l \frac{\bar{T}_s - T_w}{y_l} \tag{2}$$

The dimensionless parameters are defined as

$$\theta_s = \frac{\bar{T}_s - T_w}{T_f - T_w}, \quad Ste_l = \frac{Cp_l(T_f - T_w)}{L},$$
$$M = \frac{m}{Ay_l}, \quad \tau = \frac{a_l}{y_l^2}t, \quad \rho_i^* = \frac{\rho_i}{\rho_l} \tag{3}$$

Eq. (2) is rewritten into the dimensionless form as

$$\frac{\mathrm{d}M}{\mathrm{d}\tau} = \frac{\theta_s\, Ste_l}{\rho_i^*} \tag{4}$$

In Fig. 10, the measured ice volumes are plotted on the coordinate system given by the functions in Eq. (4). The experimental data coincide well with the ana-

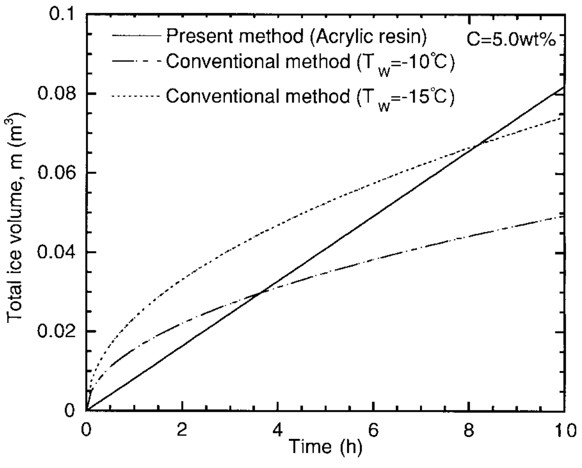

Fig. 11. Comparison between present and conventional ice formation methods.

lytical result for the various conditions of the solute concentration and the plate material. This implies that the present experimental method is reliable enough. From Eq. (4), it is seen that the ice volume can be obtained analytically when the value of θ_s is given.

Fig. 11 shows the comparison of total ice volume between the present and the conventional ice formation methods, where it is assumed that 5.0 wt% ethylene glycol solution at the freezing temperature begins to freeze on the cooled plate of 1 m^2 in heat transfer area. The conventional results shown in Fig. 11 were obtained by a quasi steady-state assumption, where the thermal conductivity of mushy ice was estimated from Fukusako and Yamada [4]. The result for the present ice formation method was obtained for the acrylic resin plate at the heat flux of $q_s = 700$ W/m^2, in which condition, the continuous ice formation and its removal phenomena were observed in the experiment. From Fig. 11, it is seen that the present ice formation method takes advantage when 4 h has elapsed after the beginning of ice formation for $T_w = -10°C$, and 8 h, for $T_w = -15°C$. In the present method, there is a possibility to increase the ice formation rate using a device for the enlargement of heat transfer area.

The important factor that affects the ice formation rate is the thermal resistance of the cooled plate. Integration of Eq. (2) with the initial condition of $m = 0$ at $t = 0$ yields

$$m = \frac{A(\bar{T}_s - T_w)}{\rho_i L} \frac{1}{y_1/\lambda_1} t \tag{5}$$

where y_1/λ_1 represents the thermal resistance of the cooled plate. It is seen from Eq. (5) that the total ice volume, m, is inversely proportional to the thermal resistance. The thermal resistance of the acrylic resin plate used in the present experiment is evaluated as $y_1/\lambda_1 = 0.0143$ (m^2 K/W), where $y_1 = 3$ mm and $\lambda_1 = 0.21$ W/m K. When the thermal conductivity of mushy ice formed by the conventional method is estimated as $\lambda_{mi} = 1.5$ W/m K [4], the thickness of mushy ice, which gives the same thermal resistance as that of the acrylic resin plate, is estimated as $y_{mi} = 7.14 y_1 = 21.4$ mm. This means that the present method takes advantage when the thickness of the mushy ice exceeds 7.14-fold of the cooled plate. Since there is a possibility to reduce the thermal resistance using thinner cooled plates than that of $y_1 = 3$ mm, the present method will be applicable to make liquid-like ice for practical use.

4. Conclusions

Ice formation and its removal phenomena of ethylene glycol solution on the horizontal surface of polyvinyl chloride as well as acrylic resin cooled plates are examined experimentally. The following conclusions can be drawn:

1. The crystal ice formed on the solid surface is removed with the buoyancy force acting on the crystal ice. It is possible to make liquid-like ice continuously, without deposition of the ice layer on the heat transfer surface.
2. For higher solute concentrations, the characteristic of the crystal ice is fragile. This results in that the number of removed crystal ice per unit of time increases and the volume of each crystal ice decreases.
3. The surface temperature of the cooled plate is supercooled and varies in the range of 0–1.0°C during the ice formation and its removal phenomena. The mean surface temperature is independent of the rear surface temperature of the cooled plate and slightly decreases with solute concentration.

Acknowledgements

This work was supported by the ‘Research for the future’ Program of the Japan Society for the Promotion of Science, JSPS-RFTF97P01003.

References

[1] T. Hirata, M. Ishikawa, T. Man'o, Y. Fujii, Crystal ice formation and removal phenomena at cooled liquid interface (in Japanese), Trans. JSME 63 (1997) 3669–3674.

[2] T. Hirata, M. Ishikawa, M. Kato, Crystal ice formation and removal phenomena of solution at cooled solid surface (in Japanese), in: Proceedings of the JSME National Meeting at Hokuriku Sin'etsu branch, Kanazawa, No. 977-1, 1997, pp. 213–214.

[3] Y. Hayashi, K. Kunimine, A. Takamori, M. Yoshida, Micro-heat transfer of solidification of mixtures with supercooling (in Japanese), Trans. JSME 62 (1996) 1517–1522.

[4] S. Fukusako, M. Yamada, Thermal conductivity of a frozen layer of ethylene glycol solution (in Japanese), Bulletin of the Faculty of Engineering, Hokkaido University 146 (1989) 33–41.