

## **Thermal Conductivity of a Dendritic Ice Layer<sup>1</sup>**

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Measurements of the thermal conductivities of the frozen layer of aqueous binary solutions have been performed using the transient hot-wire method. Solutions of ethylene glycol and sodium chloride were utilized as the testing fluids, and they were frozen up in the test section in which the platinum wires 40  $\mu\text{m}$  in diameter and 170 mm in length were strung. Measurements were carried out under equilibrium at a variety of both the initial concentration of the solution and the temperature of the frozen layer. The expressions of the thermal conductivity of the frozen layer were determined. It was found that the thermal conductivity of the dendritic ice layer was favorably assessed with the Lichteneker's model by introducing the solid fraction under an assumption of the equilibrium within the range of parameters examined.

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**KEY WORDS:** dendrite; ice; solutions; thermal conductivity; transient hot-wire method.

### **1. INTRODUCTION**

A peak maximum demand for electric power which can be of the order of about twice the base load demand usually takes place on a hot summer afternoon. To satisfy the peaks, utilities have to construct and maintain a considerable generating capacity which is used only a few hundred hours each year. Thus, the utilities have an incentive to persuade the customer to shift electrical load from on-peak, daytime periods to off-peak periods at night and on weekends. As a result of these incentives, there has been a growing interest in the use of thermal storage systems. Considerable attention has been focused on the liquid-ice storage system, which consists

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essentially of the mixture of fine-particle ice and liquid. In general, the liquid ice can be efficiently obtained by freezing a variety of solutions.

Consequently, it is important to determine the freezing characteristics of solutions and to predict both the property and the quantity of the frozen layer under various conditions. However, because of the close dependency of the property of frozen layer upon the solute type, the concentration, and the cooling conditions, most of the past studies have tended to be experimental, but a few analytical studies of both sodium chloride solutions [1–5] and ethylene-glycol solutions [6] have been reported under conditions of equilibrium.

The objective of the present study is to determine the thermal conductivity of the dendritic ice layer formed on the cooled surface under *in situ* conditions. Solutions of ethylene glycol and sodium chloride were utilized as testing liquids. The transient hot-wire method, whose wires were naturally embedded within the growing dendritic ice layer, was employed as a measurement technique. Measurements were carried out under a variety of conditions of the initial liquid concentrations and temperature levels at a quasi-steady state so as to assume equilibrium.

## 2. EXPERIMENTAL APPARATUS AND PROCEDURES

### 2.1. Experimental Apparatus

The system consists essentially of a test section, a Wheatstone bridge circuit, a DC power supply, and a data recorder connected to both the controller and a data processor. Seven platinum wires, each of which forms a part of the bridge circuit, are strung within the test section, and each of them corresponds to the different temperature level in the measurement. For the present study, the fixed resistors were set at  $20\text{ k}\Omega$  and the variable resistor could be set within an accuracy of  $0.001\ \Omega$ . The data recorder was controlled using a 16-bit microcomputer via the GP-IB interface.

Figure 1 shows details of the transparent Lucite box test section. The test section has dimensions 250 mm in width, 50 mm in depth, and 80 mm in longitudinal depth. A copper plate 3 mm in thickness served as a cooled plate, and cooling brine was sprayed from below to attain a uniform temperature. Seven polyester-coated platinum wires, whose diameter and length were  $40\ \mu\text{m}$  and 170 mm, respectively, were strung horizontally at 5 mm vertical distance each other. Just near the each platinum wire, the chromel-alumel thermocouples of 0.32 mm in diameter were set at five locations in the same horizontal plane as the platinum wire.

Both ends of the platinum wire were carefully insulated in order that the conducting portion was not in contact with the solution. One end of

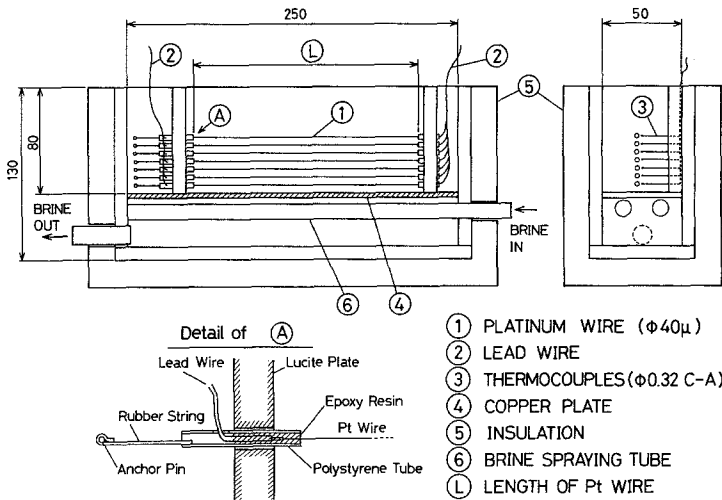


Fig. 1. Details of the test section.

the platinum wire, which was let in a styrene tube, was fixed, and the other end was connected to a rubber band to provide a suitable tension for preventing the platinum wire from slacking caused by the temperature change. The test section was covered with insulation to minimize heat losses.

**2.2. Procedures**

By circulating cooling brine, the dendritic ice layer formed slowly in the test section during a period of 3 to 4 h. The measurement was carried out after the quasi-steady state of the temperature distribution was achieved in both the ice layer and the liquid region. The quasi-steady state was defined as the state in which changes of the frozen-layer thickness and consequently the temperature distribution within both the frozen layer and the liquid layer were not observed over an hour. In the measurement, first, after balance of the bridge circuit was completely attained using a galvanometer, the recorder was connected. DC power was then supplied to the circuit, and the variation of nonbalanced voltage, which was produced by the change of the electric resistance of the platinum wire, was measured. At the same time, both the voltage of the power supply and the temperature distribution within the testing material were measured. The procedures of measurement described above were repeated successively for all seven platinum wires. Each data set was sampled at 0.5-s intervals for 10 s, recor-

ded on the floppy disk, and processed after finishing the measurement. The thermal conductivity was obtained from the following basic equation for the transient hot-wire method:

$$\lambda = (q/4\pi)/(d\theta/d \ln t) \quad (1)$$

where  $q$  and  $t$  denote the heat generation in the hot wire per unit length and time, respectively, and the temperature variation  $d\theta$  is determined based on the change of the nonbalanced voltage of the bridge circuit caused by the change of the electric resistance of the platinum wire. The thermal conductivity of frozen layer in the present study is regarded as the averaged value of all the portion of the frozen layer with which the platinum wire contacts, and effects of the variation of thermal conductivity in the upward direction and that in the downward direction cancel each other. Then, Eq. (1) can be adapted to the frozen layer in which the thermal conductivity varies. Nagasaka and Nagashima [7] have shown that Eq. (1) is adequately valid for coated platinum wire.

### 3. RESULTS AND DISCUSSION

#### 3.1. Estimation of Measurement Precision

The relative errors in the observed values of  $\lambda$  are assessed by use of the following equation, which is derived from Eq. (1):

$$\Delta\lambda/\lambda = \Delta q/q + \Delta(dV/d\theta)/(dV/d\theta) + \Delta(dV/d \ln t)/(dV/d \ln t) \quad (2)$$

For the first term on the right-hand side of the above equation, the error was estimated to be about 0.2% of the measurement precision of the electric resistance, the voltage, and the dimensions of the platinum wire. For the second term, involving the thermal coefficient of the resistance of the platinum heater wire, the error was estimated to be about 0.5%. In addition, the third term, which is concerned with the nonbalanced voltage, had an error of about 0.8%. The influence that the property and the dimensions of the platinum wire provide was estimated to be about 0.1%. Consequently, the precision of the measurement of thermal conductivity can be estimated to be about 1.6% in the present study. The precision of the temperature measurement was estimated to be about 2%. This error is caused mainly by the recorder.

#### 3.2. Measurement Results

The experimental data on the thermal conductivity as a function of temperature are presented in Figs. 2 and 3. Although the main subject of

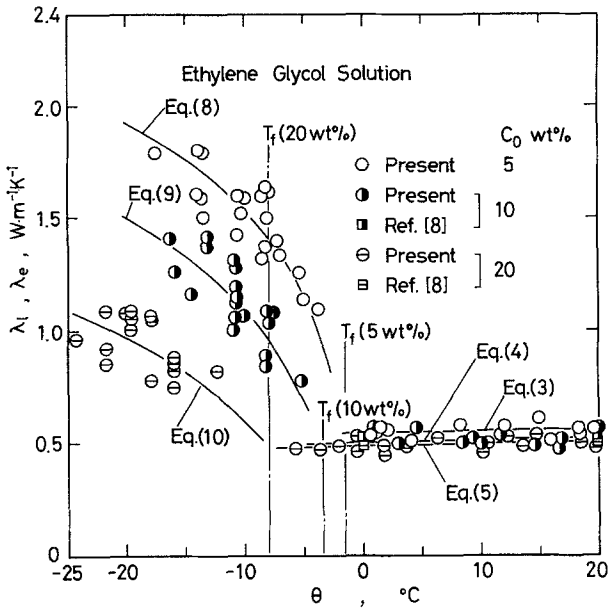


Fig. 2. Thermal conductivity of the ethylene glycol solution and its frozen layer.

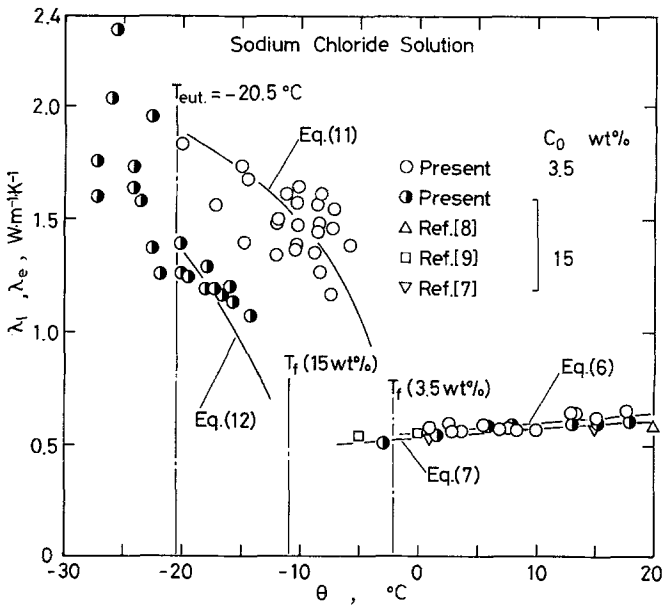


Fig. 3. Thermal conductivity of the sodium chloride solution and its frozen layer.

the measurement was the thermal conductivity of the frozen layer, the measurements have also been conducted extensively for the liquid phase within the temperatures ranging from  $T_f$  to  $20^\circ\text{C}$ , where  $T_f$  is the solidification temperature at equilibrium.

For the temperature range investigated, the results for the liquid-phase region could be expressed by the following equations within the deviation range of about 2.3 to 3.3%, respectively:

$$\lambda_l = 0.544 + 5.63 \times 10^{-4}\theta \quad (\pm 2.3\%) \quad (3)$$

$$\lambda_l = 0.497 + 1.98 \times 10^{-3}\theta \quad (\pm 2.6\%) \quad (4)$$

$$\lambda_l = 0.484 + 1.14 \times 10^{-3}\theta \quad (\pm 2.7\%) \quad (5)$$

$$\lambda_l = 0.549 + 5.14 \times 10^{-3}\theta \quad (\pm 3.2\%) \quad (6)$$

$$\lambda_l = 0.538 + 4.43 \times 10^{-3}\theta \quad (\pm 3.3\%) \quad (7)$$

where  $\lambda_l$  ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ ) is the thermal conductivity of the liquid-phase region,  $\theta$  is the temperature of the solution, and the values in parentheses are the maximum deviations of the measured values, respectively. In Fig. 2, the data for ethylene glycol of 10 and 20% concentration lie close to those of Riedel [8], respectively. On the other hand, as shown in Fig. 3, the present experimental data for the sodium chloride solution of 15 wt% in concentration are seen to coincide with those of Sugawara et al. [9], Nagasaka and Nagashima [7], and Riedel [8] within the deviation range of about 3%. At the temperature close to the equilibrium solidification temperature  $T_f$ , this favorable result was not available for every substance. This is attributed to the fact that the assumption of equilibrium is not achieved near the interface between the frozen layer and the liquid-phase region.

The thermal conductivity of the frozen layer  $\lambda_e$  can be expressed by the following equations within the deviation range of  $\pm 15\%$  except for the temperatures near  $T_f$ . It is, however, to be noted that for the sodium chloride solution the application of the equations is restricted for temperatures higher than the eutectic temperature  $T_{\text{eut}}$  ( $\theta = -20.5^\circ\text{C}$ ).

$$\lambda_e = 0.543 + 0.542 \ln(\theta/T_f) \quad (8)$$

$$\lambda_e = 0.489 + 0.588 \ln(\theta/T_f) \quad (9)$$

$$\lambda_e = 0.475 + 0.531 \ln(\theta/T_f) \quad (10)$$

$$\lambda_e = 0.557 + 0.588 \ln(\theta/T_f) \quad (11)$$

$$\lambda_e = 0.490 + 1.41 \ln(\theta/T_f) \quad (12)$$

In the frozen layer,  $\lambda_e$  increases as the temperature decreases, and at a specified temperature,  $\lambda_e$  decreases as the concentration increases. This may be explained by the fact that at the lower temperature, the concentration of the solution is also lower and the solid fraction, namely, the volume ratio of the ice at equilibrium is higher.

The deviation of the measured value in the liquid region is about 3%, which is very close to the estimated precision of measurement, while that for the frozen layer is about 15% and much greater than that for the liquid region. This can be explained as follows. The measured value of  $\lambda_e$  is taken as the averaged value of all the portion of the frozen layer with which the platinum wire makes contact (solidus region and enriched liquid region). In addition, the factor related to the variability of the constitution of the frozen layer must be added to the measurement errors. For the sodium chloride solution of 15 wt% concentration, the great scatter band of the value of  $\lambda_e$  was observed for the temperatures less than the eutectic one. This may be due to the fact that the condition of the eutectic region of the layer varies according to the freezing conditions (e.g., the freezing rate).

### 3.3. Data Arrangement by Solid Fraction

The relation among the initial concentration of the solution  $C_0$ , the equilibrium concentration  $C_e$ , the concentration of solute in the solid phase  $C_s$ , and the solid fraction  $f_s$  can, in general, be expressed from the lever rule as follows:

$$f_s = (C_e - C_0)/(C_e - C_s) \quad (13)$$

Assuming that the solute is not captured within the ice ( $C_s = 0$ ), the equilibrium concentration  $C_e$  at a specified temperature obtained from the table results in the solid fraction due to Eq. (14).

$$f_s = (C_e - C_0)/C_e \quad (14)$$

In addition, rearranging the data (setting  $f_s$  as the ordinate and  $\lambda_e$  as the abscissa), one obtains Figs. 4 and 5 for the substances covered. In the figures, the dashed line denotes the calculated value based on the series model shown in Fig. 6b and the dashed-dotted line denotes that based on the parallel model shown in Fig. 6c, respectively. The expressions for each model are as follows:

$$\text{for the series model,} \quad \lambda_e = [f_s/\lambda_s + (1 - f_s)/\lambda_1]^{-1} \quad (15)$$

$$\text{for the parallel model,} \quad \lambda_e = f_s\lambda_s + (1 - f_s)\lambda_1 \quad (16)$$

where  $\lambda_s$  ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ ) is the thermal conductivity of ice.

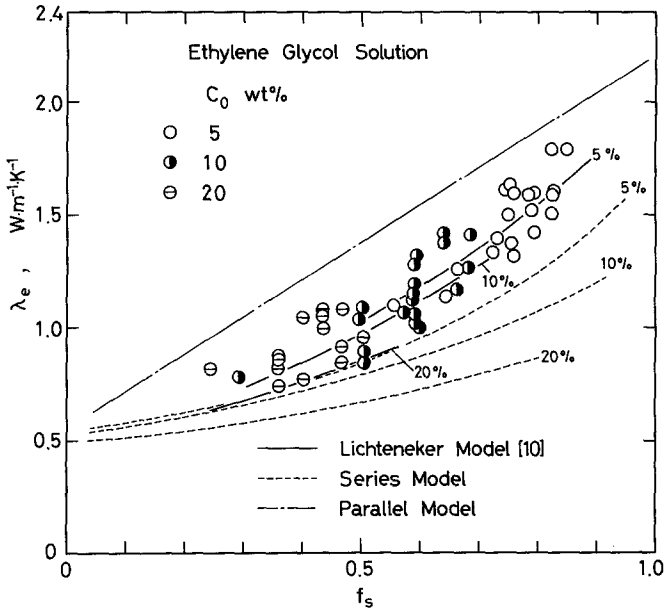


Fig. 4. Thermal conductivity of the frozen layer of the ethylene glycol solution.

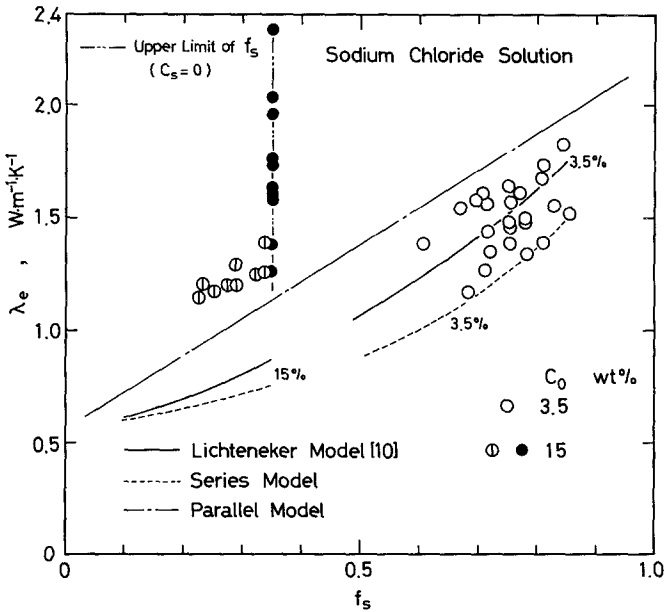


Fig. 5. Thermal conductivity of the frozen layer of the sodium chloride solution.



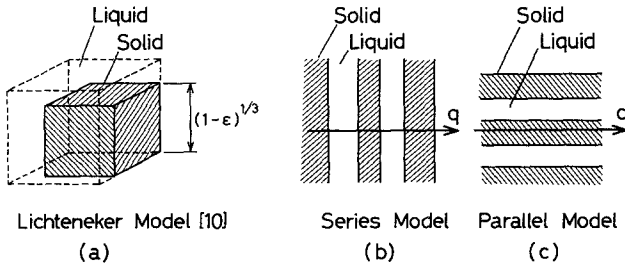


Fig. 6. Models for the effective thermal conductivity.

The frozen layer in the present study is the dendritic ice layer and actually the condition of the frozen layer appears to be located between the series model and the parallel model from the three-dimensional character of the dendritic ice. Therefore, according to the Lichteneker model [10] shown in Fig. 6a, the solid lines in Figs. 4 and 5 are obtained. The thermal conductivity of the frozen layer by the Lichteneker model is obtained from the following equation:

$$\lambda_e = \frac{2 - \varepsilon - (1 - \varepsilon)^{1/3} + (\lambda_l/\lambda_s)[(1 - \varepsilon)^{1/3} - (1 - \varepsilon)]}{1 - (1 - \varepsilon)^{1/3} + (\lambda_l/\lambda_s)(1 - \varepsilon)^{1/3}} \quad (17)$$

where  $\varepsilon$  is the void fraction ( $1 - f_s$ ). The hatched part in Figs. 6a–c denotes the solid region and the open part denotes the liquid region, respectively; the arrows in the figure show the direction of heat flow. For the model in Fig. 6a, the direction of heat flow is not provided. Inspection of Figs. 4 and 5 reveals that the present data for the ethylene glycol solutions of 5 and 10 wt% and for the sodium chloride solution of 3.5 wt% may be well correlated with the solid fraction by using the Lichteneker model within the same precision as the data arrangement to the temperature. However, in Fig. 4, the data for 20 wt% tend to be closer to the parallel model compared to those for 5 and 10 wt%. However, for the sodium chloride solution of 15 wt%, the data result in greater values than that of the parallel model, as shown in Fig. 5. The following are possible reasons: (1) great error is caused by the approximation of the concentration–temperature curve, and (2) the solute is trapped in the ice ( $C_s \neq 0$ ).

As for the first reason, the precision of approximation is estimated to be about 1% within the temperature range covered, and thus this may not be the main reason for the tendency described above. With regard to the second reason, for example, in the case of the sodium chloride solution of 15 wt% and  $\theta = -20.5^\circ\text{C}$ , if  $C_s = 0$ , then one obtains  $f_s = 0.34$ , and if  $C_s = 5\%$ , then  $f_s = 0.44$ , and furthermore, if  $C_s = 10\%$ , then  $f_s = 0.62$ .

Thus, it seems that the value of  $C_s$  may exert a quite considerable influence on  $f_s$ . By assuming that  $C_s = 10\%$  for the ethylene glycol solution of 20 wt% and for the sodium chloride solution of 15 wt%, the results in the present study could be reasonably expressed using the Lichteneker model in a similar way for  $C_0 = 3.5, 5,$  and  $10\text{ wt}\%$ . Therefore, it appears that the assumption of  $C_s = 0$  may no longer be achieved for the high concentration of the solutions. An additional investigation is necessary on this problem.

#### 4. CONCLUSIONS

Measurements of thermal conductivities of the solutions of ethylene glycol and sodium chloride and also of the frozen layer of these solutions have been carried out by the use of the transient hot-wire method. The following conclusions may be drawn within the range of the parameters covered in the present work.

(1) The thermal conductivity of the frozen layer  $\lambda_e$  is favorably expressed by the following form within the deviation range of  $\pm 15\%$ :

$$\lambda_e = \lambda_0 + A \ln(\theta/T_f)$$

The deviation range of  $\pm 15\%$  is much greater than the estimated overall precision of measurement. This could be due mainly to the variability of the constitution of frozen layer.

(2) By introducing the solid fraction  $f_s$  at equilibrium, the thermal conductivity of the frozen layer can be expressed based on the Lichteneker model for initial concentrations less than about 10%.

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