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# Influences of initial concentration and super-cooling degree on the permeability of a porous medium saturated with partially solidified aqueous solution

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**Abstract**—The permeability of a porous medium in a state of solidification saturated with NaCl-aqueous solution was measured by a transient method, with a varying volume fraction of liquid phase  $\chi$ , an initial concentration of solution  $C_i$ , and a super-cooling degree of the solution just before start of solidification  $T_{sc}$ . From the results of measurement, it was shown that the permeability was proportional to the  $n$ th power function of  $\chi$ . The influence of  $C_i$  and  $T_{sc}$  on the permeability was clarified. An empirical equation of the permeability was obtained as a function of  $\chi$ ,  $C_i$  and  $T_{sc}$ . Copyright © 1996 Elsevier Science Ltd.

## 1. INTRODUCTION

The solidification phenomenon of a porous medium saturated with a solution appears in the processes of solidification of a soil containing seawater, solidification of binary phase change material containing a high thermal conductivity material to enhance the heat transfer, and solidification to process or store food. The solidification process of such binary material as a composite alloy in which particles are scattered is also similar to the above solidification phenomenon.

It is known that the solidification process of a solution is mainly governed by a double-diffusive convection caused by temperature and concentration gradients [1, 2]. In this solidification process, the solute is discharged into the solution with a growth of the mushy region containing both the liquid and solidified phases; the discharged solute brings the concentration gradient. The solidification process of a porous medium saturated with an aqueous solution is also sufficiently affected by the double-diffusive convection

[3–7], in addition, a characteristic of the solidification of the porous medium is that a permeability governing the convection of the mushy region changes rapidly with a growth of ice formed in the mushy region [5–8]. Therefore, clarification of the characteristics of the permeability is essential to an analysis of the solidification process of the porous medium saturated with an aqueous solution.

Many measurements of permeability in a state without solidification have been carried out and various equations to arrange and express the measured permeability are shown [9]. But there is no report of a permeability in a mushy region except for the authors' reports in refs. [5–7].

By doing experiments and analyses, the authors clarified the process. In the process, each rectangular cell packed with several kinds of beads was used as porous media. The porous media saturated with a NaCl-aqueous solution of 10 or 3 wt% initial concentration were solidified from one of the vertical walls of the cell [5–7]. Based on a comparison of the experimental results with the analytical ones, it was shown that the local permeability in the mushy region  $k$  was able to be expressed as a function of the local

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## NOMENCLATURE

$C_i$	initial concentration [wt%]	$v$	velocity [ $\text{m s}^{-1}$ ].
$D_b$	mean diameter of beads [m]	Greek symbols	
$g$	gravitational acceleration [ $\text{m s}^{-2}$ ]	$\mu$	coefficient of viscosity [ $\text{kg (m}^{-1} \text{s}^{-1})$ ]
$h_1(t), h_2(t)$	potential head [m]	$\nu$	kinematic viscosity [ $\text{m}^2 \text{s}^{-1}$ ]
$k$	permeability [ $\text{m}^2$ ]	$\rho$	density [ $\text{kg m}^{-3}$ ]
$k_0$	permeability before solidification [ $\text{m}^2$ ]	$\chi$	volume fraction of liquid phase
$L$	length of porous medium [m]	$\phi$	porosity
$p$	pressure [Pa]	$\chi/\phi$	ratio of the volume fraction of a liquid phase in the mushy region to porosity (when a porous medium is not solidified, $\chi/\phi = 1$ ).
$Re$	Reynolds number [ $= (D_b v)/\nu$ ]		
$T_{sc}$	super-cooling degree of solution just before start of solidification [K]		
$t$	time [s]		

volume fraction of the liquid phase  $\chi$ . Namely, the following equation was obtained.

$$(k/k_0) = (\chi/\phi)^n \quad (1)$$

where  $\chi$  means volume of liquid phase per unit volume of a porous medium and satisfies  $0 \leq \chi \leq \phi$ .  $\chi/\phi$  is called a ratio of volume fraction of the liquid phase and satisfies  $0 \leq \chi/\phi \leq 1$ . The value of  $k/k_0$  changes from 0 to 1 according to the change of the value of  $\chi/\phi$  from 0 to 1. The exponent  $n$  of equation (1) is considered to be a constant value which is mainly determined by mean diameter of the beads composing a porous medium and an initial concentration of solution. Besides, the authors offered a new simple transient measurement method of the permeability [8]. By this method, the permeabilities in the mushy region were measured. In these measurements, the initial concentration of the solution, the mean diameter and the material of the beads were varied. It was shown that in the case of the solidification process of a porous medium packed with glass beads with a 2.56 mm mean diameter and saturated with a NaCl-solution of 10 wt%, the experimental result of the permeability was able to be expressed as equation (1) and the exponent  $n$  was 16 [8]. The value of  $n$  ( $n = 16$ ) determined by the above measurements is approximately equal to that of  $n$  ( $n = 14$ ) which was used in the analysis [6] of the solidification process of the porous medium saturated with NaCl-solution. The solidification started by cooling the vertical wall of a rectangular cell. The approximate agreement between both the values of  $n$  means that equation (1) is valid. Permeability in mushy region, of course, is determined by the shape of ice formed in the porous medium. The shape of ice is affected by the diameter of particle composing a porous medium, the thermal properties of a particle, an initial concentration of solution, a super-cooling degree of solution just before the start of solidification and so on. The permeabilities of porous media composed of steel, glass and vinyl chloride beads were measured and from the results it was found that the influence of the thermal properties of beads on per-

meability was small [8]. But the other factors governing the permeability have never been discussed yet. In the previous report [8], a porous medium saturated with a NaCl-aqueous solution was cooled at the predetermined temperature to get a desired volume fraction of liquid phase, and then the porous medium was solidified. After that, the permeability was measured without considering a super-cooling degree of solution just before the start of solidification. So the range of the super-cooling degree was restricted to the difference between the solidification temperature of a solution and the predetermined temperature.

In the present report, the authors measured the permeabilities in mushy region by the transient measurement method described in the previous report [8], varying the volume fraction of the liquid phase, the initial concentration of the solution and the super-cooling degree of the solution just before start of solidification. The influences of the above factors on the permeability are clarified.

## 2. EXPERIMENT

## 2.1. Principle of the transient measurement method and basic equations

The principle of the transient measurement method is shown in Fig. 1. The U-tube manometer with a constant cross-section is applied to the measurement of the permeability. In this method, a porous medium with a length  $L$  is set in a horizontal tube between two

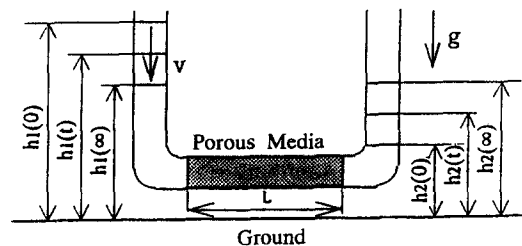


Fig. 1. Principle of the transient measurement method of permeability.

vertical tubes of the U-tube. At first, a fluid is saturated in the U-tube. One of the vertical tubes is closed by a cap and the fluid is poured into the other vertical tube and then an initial potential head difference between two vertical tubes of the U-tube is made. Secondly, the fluid is allowed to flow in the U-tube by removing the cap, and the transient displacements of the potential heads are measured. If Darcy's law is valid for this model ( $Re \leq 1$ ), the permeability can be determined by measuring transient displacements of the potential heads, because the pressure drop is mainly caused by the flow through the porous medium. The influences of the friction of the tube and the pressure loss due to the bend of the tube on permeability were estimated and these were small enough to be neglected. By expressing transient displacement of the potential head from a horizontal plane as  $h(t)$ , equations (2) and (3) can be derived. By solving equations (2) and (3), equations (4) and (5) are obtained, where the assumptions for the principle of the measurement are as follows:

- (1) Darcy's law is valid;
- (2) the inertia force of solution is negligible;
- (3) the heat generation due to viscosity of the solution is negligible;
- (4) the pressure drop occurs only in a porous medium.

$$v = -\frac{k}{\mu} \frac{\partial p}{\partial x} = \frac{k \rho g}{\mu L} [h_1(t) - h_2(t)] \quad (2)$$

$$v = -\frac{dh_1(t)}{dt} = \frac{dh_2(t)}{dt} \quad (3)$$

$$\ln \frac{h_1(t) - h_1(\infty)}{h_1(0) - h_1(\infty)} = -2Kt \quad (4)$$

$$K = \frac{k \rho g}{\mu L}, \quad (5)$$

where  $v (= \mu/\rho)$  was expressed as a function of temperature and concentration. The transient displacement of potential head  $h_1(t)$  is measured as the

function of time. The left hand side of equation (4) is proportional to time  $t$  and  $K$  is obtained as a gradient of a straight line by the least square method on the basis of equation (4). Then, from equation (5), the permeability  $k$  is determined.

2.2. Experimental apparatus and measurement method

Figure 2 shows the schematic diagram of the experimental apparatus for measuring the permeability in a state of solidification of a porous medium saturated with an aqueous solution. In Fig. 2, the beads are packed in 'Section A' and a solution is stored up in 'Section B'. The sections indicate the inner tubes of double-tube construction. The inner tubes with 14 mm inner diameter are transparent and are made of acrylic or polycarbonate resin. The lengths of Section A and B are 500 and 2000 mm, respectively. The temperatures of both sections were controlled by letting a brine flow through the annulus of the double-tube construction. The beads packed between the stainless meshes at both ends of the Section A are treated as a porous medium. The porous medium was saturated with a solution of a certain predetermined concentration. The concentration is called the initial concentration  $C_i$ . The glass beads with mean diameter  $D_b = 2.56$  mm and NaCl-solution were used, and the measured value of porosity  $\phi$  was 0.36. Temperatures at the center of the tube packing beads were measured by probes made with type T thermocouples inserted into stainless pipes. Those probes were set at two points located at 100 mm inward from both ends of Section A. Additionally, the temperature of a connection between Section A and Section B was also measured.

When the solution in Section A is solidified, pure ice is formed in the porous medium, and then the concentration of the solution increases due to the discharged solute. For a certain initial concentration of solution, the temperature to obtain a desired volume fraction of liquid phase in a mushy region is determined by a phase equilibrium diagram [10], and at the

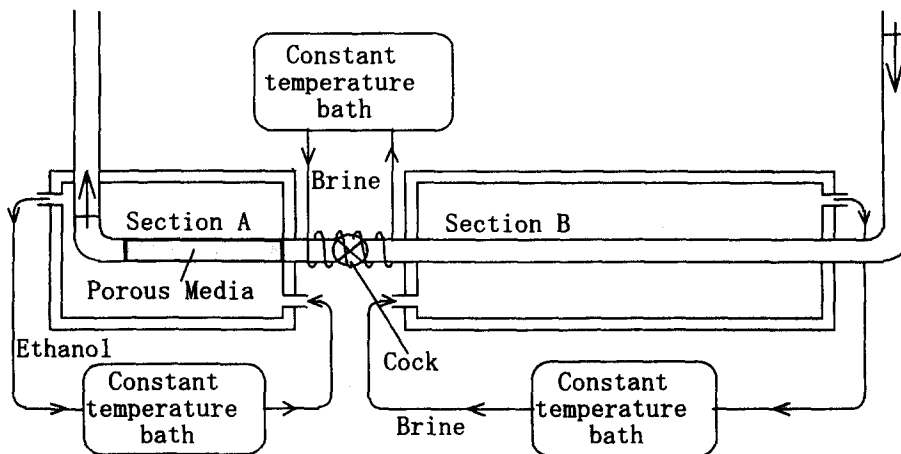


Fig. 2. Schematic diagram of measurement apparatus.

same time the concentration of solution in the porous medium is obtained by the determined temperature, where volume expansion due to solidification is negligible.

The experiments were carried out according to the following procedure. At first, after the solution in Section A was cooled at a desired super-cooling degree by letting a constant temperature ethanol flow outside Section A, the solidification of the porous medium was initiated by dissolution of the super-cooling state. Then, the temperature of the ethanol outside Section A was changed so that a desired volume fraction of liquid phase could be obtained in Section A. The adjusted temperature was kept constant. The solution of the same temperature and concentration as those of the solution in Section A, adjusted by the above procedure, was stored in Section B, and the solution in Section B was let to flow into Section A by opening a cock. So the permeability in the mushy region was able to be measured without melting or growing of the ice formed in the porous medium. The connection between Section A and Section B was cooled by letting the brine flow into the tube which was wound around the connection. Sections A and B and the cooled connection were covered with insulation. Dissolution of the super-cooling state of the solution was carried out by a direct impact on one of the stainless meshes holding both sides of the porous medium. The transient potential heads were taken by CCD camera and were recorded by VTR.

For each of initial concentrations of a solution,  $C_i = 5, 10, 15$  wt%, experiments were carried out under conditions of super-cooling degree of solution just before the start of solidification,  $T_{sc} = 1, 3$  and  $5$  K. Also, under the same condition, the measurements were repeated after the beads and solution were set in Section A again and the desired experimental conditions were reset.

### 2.3. Discussion on the effectiveness of the present transient measurement method

Though in the previous report [8] the effectiveness of the present method was discussed fully, we think that it had better be mentioned again in order to understand the present report. So it is described briefly in the following.

For each desired volume fraction of the liquid phase in a mushy region, each example of the transient displacement of potential head is shown in Fig. 3. The figure is a semi-logarithms graph, where glass beads with  $D_b = 2.56$  mm and a NaCl-solution with  $C_i = 10$  wt% were used. From Fig. 3, it is found that the potential head decreases with time along a straight line. So the permeability can be determined by the gradient of the straight line as shown in equations 4 and 5. The values measured by the present transient method were compared with the values by the steady method and the values in literature under the conditions of room temperature and pure water. The values measured by the present method agree with the

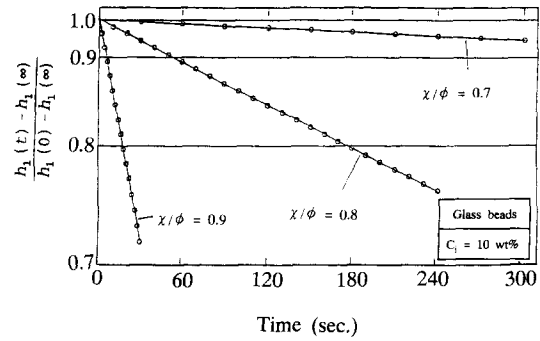


Fig. 3. Time-wise displacement of potential head.

ones by the steady method to within 4%, and the difference between the values measured by the present method and the values shown in ref. [11] which were measured by Krumbein and Monk was within 3%. By calculation, it was also shown that the total influence of the friction of the tube and the pressure loss due to the bend of the tube on permeability was about 1%.

In order to measure the permeability of a porous medium with a desired volume fraction of liquid phase in the mushy region, ice in the porous medium must not grow or melt in the period of measurement. An example of the measured temperatures is shown in Fig. 4. The symbols  $\Delta$  and  $\circ$  in Fig. 4 indicate the temperature near the inlet of the porous medium and that near the outlet, respectively. From Fig. 4, it is found that both temperatures in the porous medium were kept approximately constant and the difference (about  $\pm 0.1^\circ\text{C}$ ) between the temperature at both points was small enough to be neglected. Therefore, it can be considered that the temperature in the porous medium was kept at the solidification temperature corresponding to the desired volume fraction of the liquid phase during the period of measurement. A solution of about  $2\text{ mm}^3$  was collected and the concentration of the solution was measured by a concentration-meter (salt analyzer based on coulometric titration). The measured concentrations of the solution near the inlet and outlet of the porous medium

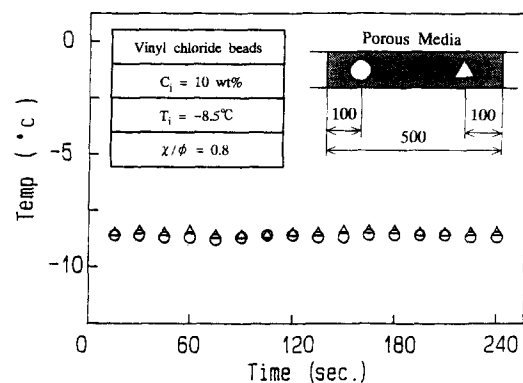


Fig. 4. Temperatures near the inlet and outlet of a porous medium.

Table 1. Comparison of the concentration of solution near the inlet of a porous medium with that near the outlet

$\chi/\phi$	Glass		Steel		Vinyl chloride		Glass		Glass	
	$D_b = 2.56 \text{ mm}$		$D_b = 2.76 \text{ mm}$		$D_b = 3.76 \text{ mm}$		$D_b = 0.92 \text{ mm}$		$D_b = 2.56 \text{ mm}$	
	$C_i = 10 \text{ wt\%}$		$C_i = 10 \text{ wt\%}$		$C_i = 10 \text{ wt\%}$		$C_i = 10 \text{ wt\%}$		$C_i = 15 \text{ wt\%}$	
	A	B	A	B	A	B	A	B	A	B
	g 100 ml <sup>-1</sup> g 100 ml <sup>-1</sup>		g 100 ml <sup>-1</sup> g 100 ml <sup>-1</sup>		g 100 ml <sup>-1</sup> g 100 ml <sup>-1</sup>		g 100 ml <sup>-1</sup> g 100 ml <sup>-1</sup>		g 100 ml <sup>-1</sup> g 100 ml <sup>-1</sup>	
0.95	11.27	11.31								
0.90	11.84	11.84	11.84	11.84	11.95	11.95	11.97	11.96	17.90	17.89
0.80	12.70	12.48	12.83	12.83	12.95	12.71	13.06	13.04	20.40	20.30
0.70	14.63	14.16	14.75	14.68	15.21	15.19	15.23	14.98	23.58	23.43

A = concentration near inlet of porous medium.

B = concentration near outlet of porous medium.

with  $C_i = 10 \text{ wt\%}$  for each value of  $(\chi/\phi)$  are shown in Table 1. We carried out the measurements, taking care not to disorder the concentration field in the porous medium. As shown in Table 1, the concentration difference between the above points was very small (the ratio of the difference to  $C_i$  was within  $\pm 3\%$ ) and was also negligible during the period of the measurement. From the above results, the growing or melting of ice in the porous medium during the period of measurement was hardly caused by the flow of solution through the porous medium, so the permeability for the desired volume fraction of liquid phase in the mushy region was able to be measured by the present experimental apparatus.

### 3. EXPERIMENTAL RESULTS AND DISCUSSION

Examples of the measured results of temperature in a porous medium in a cooling process are shown in Figs. 5 and 6. The temperatures were measured at 100 mm inward from both ends of the porous medium. In both the figures, an initial concentration of solution  $C_i$  is 15 wt% and a super-cooling degree of solution just before start of solidification  $T_{sc}$  is 5 K. The values of  $\chi/\phi$  in Figs. 5 and 6 are 0.8 and 0.7, respectively. The horizontal lines shown in Figs. 5 and 6 represent

the solidification temperature (freezing point) of the solution corresponding to the initial concentration  $C_i$ . In Fig. 5, the initial temperature of the porous medium was kept at  $-16^\circ\text{C}$  (super-cooling degree = 5 K) and after the super-cooling of the solution was dissolved, the temperature of the porous medium was kept at  $-14.5^\circ\text{C}$  so that  $\chi/\phi = 0.8$  could be obtained. The solution was let to flow into the porous medium, and then, the measurement of permeability was carried out. In Fig. 5 a steep rise of temperature shown by symbol  $\diamond$  at  $t \approx 70 \text{ min}$  means that a super-cooling degree was dissolved at that time. Since the super-cooling state was made to be dissolved from the outlet of solution which was located at the left side of the porous medium in Fig. 5, the solidification at the point  $\bullet$  started later than that at the point  $\diamond$  by about 10 min. In Fig. 5, ice formed by dissolving the super-cooling was melted by rising temperature in order to obtain a desired volume fraction of liquid phase. On the other hand in Fig. 6, ice was further grown by reducing temperature.

Under various initial concentrations and super-cooling degrees the relationships between  $(k/k_0)$  and  $(\chi/\phi)$  are obtained and shown as log-log graphs in Figs. 7-9. The values of  $C_i$  in Figs. 7-9 are 5, 10, 15 wt%, respectively. Symbols  $\circ$ ,  $\triangle$  and  $\square$  in those

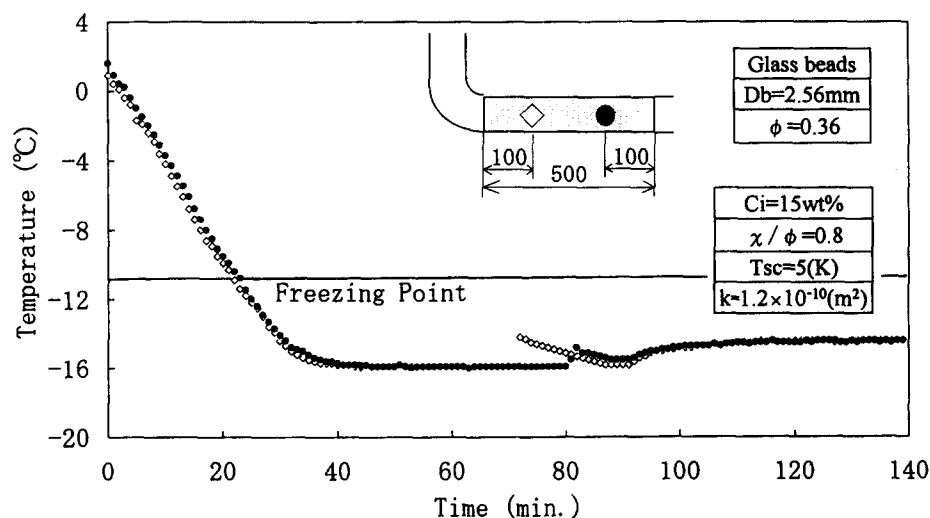


Fig. 5. Relationship between the temperature in a porous medium and time in a cooling process ( $\chi/\phi = 0.8$ ).

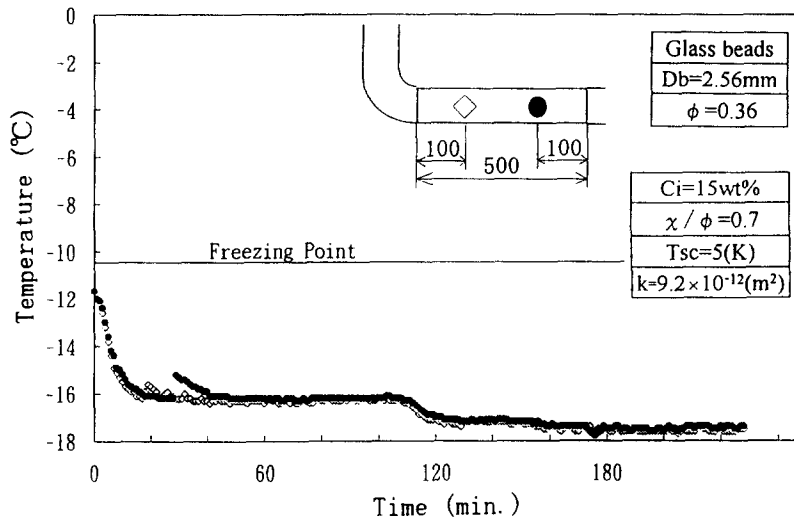


Fig. 6. Relationship between the temperature in a porous medium and time in a cooling process ( $\chi/\phi = 0.7$ ).

figures show  $T_{sc} = 1, 3$  and  $5$  K, respectively. In those figures, the straight lines calculated by  $(k/k_0) = (\chi/\phi)^{14}$  are shown to compare with the experimental results. The exponent of the above equation, 14, just corresponds to the value used in the analysis for the case that a porous medium saturated with NaCl-aqueous solution with  $C_i = 10$  wt% in a rectangular cell was solidified. Large scatters of experimental results of  $(k/k_0)$  appear for each value of  $\chi/\phi$  in Figs. 7-9. The scatters were caused not by a bad reproducibility of the measurement method, but by a difference of structure of micro-path formed after dissolution of super-cooling state for each experiment. As is evident from the result that the value of permeability is decreased rapidly to  $1/500$  when the value of  $(\chi/\phi)$  changes from 1 to 0.7, permeability is a kind of property affected sensitively by micro-structure of ice in a porous medium. Namely, it is considered that the above large

differences were caused by the governing factors in the processes of ice formation, such as a dissolution rate from a super-cooling state and a solidification rate or melting rate after the dissolution. From Figs. 7-9, it is clarified that permeability decreases rapidly with the decrease of volume fraction of liquid phase  $\chi$  in proportion to the  $n$ th power of  $\chi$ . For the experimental results in the case of  $C_i = 10$  wt% shown in Fig. 8, the measured values of  $k/k_0$  for  $\chi/\phi = 0.9$  can be approximated by a straight line in the case of  $n = 14$ , and the measured values for  $\chi/\phi = 0.7$  and  $0.8$  can be approximated by a straight line of  $n = 16$ . The straight line with  $n = 16$  corresponds to the result of the previous report [8] in which the super-cooling was not considered. Permeability expressed by equation (1) with  $n = 14$  was suitable for simulation of a solidification process of the porous medium as shown in the foregone reports [5, 6]. Since the natural con-

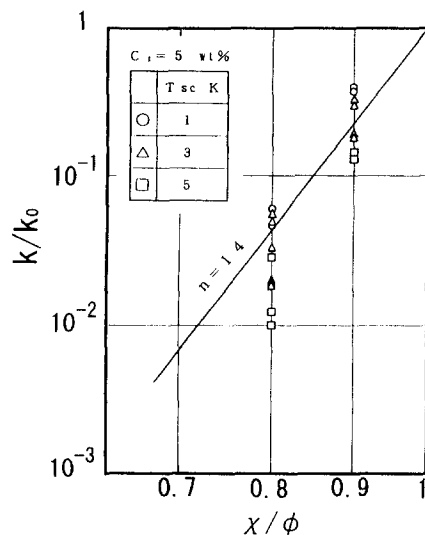


Fig. 7. Relationship between  $(k/k_0)$  and  $(\chi/\phi)$  ( $C_i = 5$  wt%).

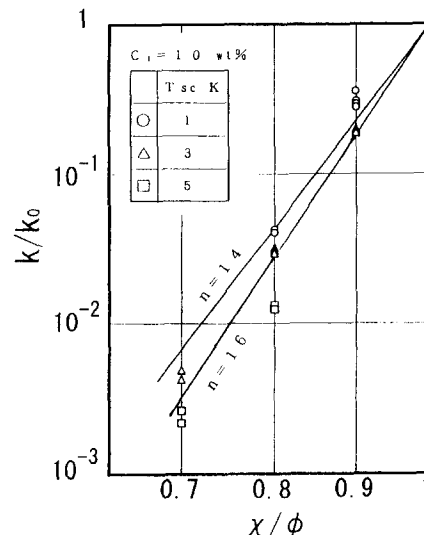


Fig. 8. Relationship between  $(k/k_0)$  and  $(\chi/\phi)$  ( $C_i = 10$  wt%).

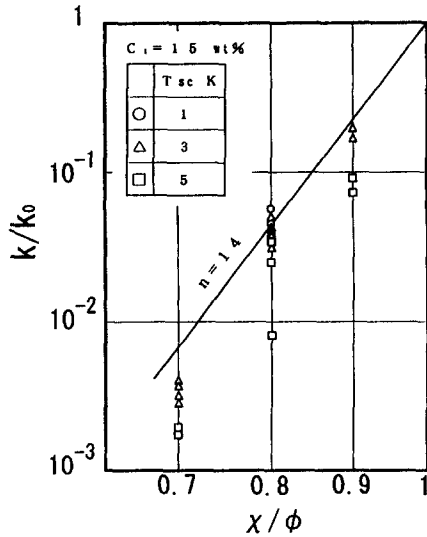


Fig. 9. Relationship between  $(k/k_0)$  and  $(\chi/\phi)$  ( $C_i = 15$  wt%).

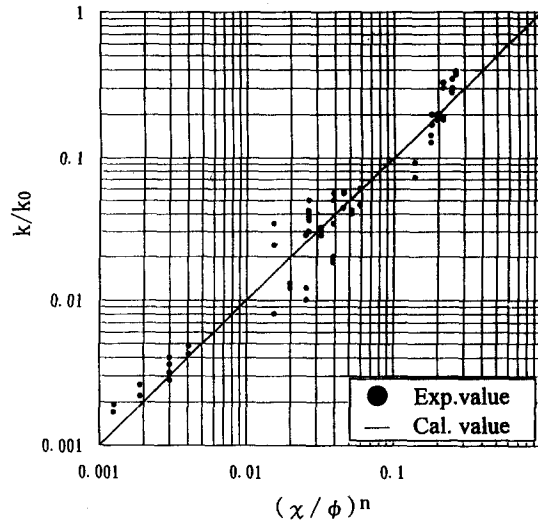


Fig. 10. Correlation between the measured value and calculated value of permeability.

vection caused by temperature and concentration gradients dominates the solidification process, it is required to obtain a suitable value of permeability in order to simulate the solidification process with accuracy. Since permeability increases with the increase of the value of  $\chi/\phi$ , a natural convection in the mushy region is active in a region where the value of  $\chi/\phi$  is large. But the concentration in this region is almost the same as the concentration in the liquid region, which is not solidified yet. On the other hand, a mushy region where  $\chi/\phi$  is small has a low temperature and high concentration, though it has small permeability. Therefore, small velocity in the mushy region with small  $\chi/\phi$  may transport a considerable amount of solute. Consequently, the value of  $n = 14$  was suitable to simulate the solidification process reported in refs. [5, 6]. In general, a suitable value of  $n$  which can be applied to an adequate range of  $\chi/\phi$  should be chosen.

It is found that for each  $C_i$  and  $\chi/\phi$ , shown in Figs. 7-9, the permeability decreases with the increase of the super-cooling degree just before the start of solidification  $T_{sc}$ . Experimental results corresponding to symbol  $\square$  in the case of  $C_i = 5$  wt% and  $\chi/\phi = 0.9$  in Fig. 7 and symbol  $\square$  in the case of  $C_i = 15$  wt% and  $\chi/\phi = 0.9$  in Fig. 9 show that the permeability under the same conditions for  $T_{sc}$  and  $\chi/\phi$  decreases with the increase of  $C_i$ , though this tendency of permeability is not clear. The results obtained by Figs. 7-9 were approximated by equation (1) and the exponent  $n$  of equation (1) was determined for each  $C_i$  and  $T_{sc}$  by a least square method. The empirical equation (6) was also obtained by the least square method.

$$(k/k_0) = (\chi/\phi)^n \quad (1)$$

$$n = 2.8 \times 10^{-2} C_i T_{sc} + 0.8 T_{sc} + 8.8 \times 10^{-2} C_i + 11.3 \quad (6)$$

where,  $5 \leq C_i \leq 15$  wt%,  $1 \leq T_{sc} \leq 5$  K.

In order to check the validity of equation (6), a correlation between the experimental values and calculated ones is shown by Fig. 10, where the experimental values of permeability are plotted in ordinate, and the values calculated by equation (6) are plotted in abscissa. Although large scatters of measured values appear, those values are distributed around the straight line with a gradient of  $45^\circ$ , so it is found that the experimental results can almost be arranged by this equation (6). The coefficients of  $C_i$  in equation (6) are small values, but these are positive, that is, it is suggested that the exponent  $n$  increases with the increase of  $C_i$ . The above suggestion corresponds to the result that in the analysis of the solidification process of the porous medium saturated with NaCl-solution in the rectangular cell, the applicable value of  $n$  increased with the increase of  $C_i$ .

In order to discuss the influence of a super-cooling degree just before the start of solidification on permeability, the relationship between permeability  $k$  and super-cooling degree just before the start of solidification  $T_{sc}$  in the case of  $C_i = 10$  wt% and  $\chi/\phi = 0.8$  is shown in Fig. 11. Figure 11 shows the

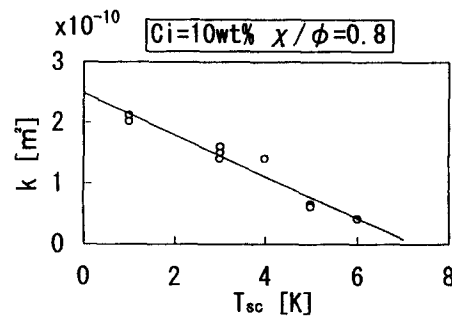


Fig. 11. Relationship between permeability  $k$  and the super-cooling degree of the solution just before the start of solidification  $T_{sc}$ .

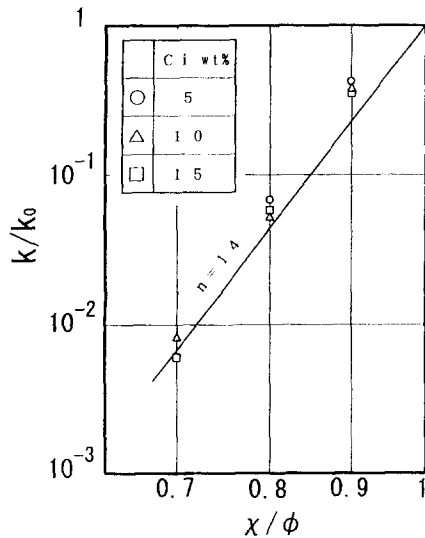


Fig. 12. Relationship between  $(k/k_0)$  and  $(\chi/\phi)$  in the case of  $T_{sc} = 0$  K.

results measured with various values of  $T_{sc}$  in addition to the results shown in Fig. 8. From Fig. 11, it is found that the value of  $k$  decreases with the increase of  $T_{sc}$ . The straight line in Fig. 11 shows the regression line drawn by the least square method. The value of permeability at  $T_{sc} = 0$  can be extrapolated by using this straight line. In the same way as above, the extrapolated values of permeabilities at  $T_{sc} = 0$  for each  $C_i$  and  $\chi/\phi$  were determined, as shown in Fig. 12.

The permeability decreases with the increase of  $C_i$  or  $T_{sc}$ , and the probability that the nucleus of ice is generated increases with the increase of  $T_{sc}$ . A growth rate of a dendritic ice crystal also increases with the increase of  $T_{sc}$ . So, in the process of dissolution of the super-cooling state, the pores among the beads are further filled with fine dendritic ice crystals with the increase of  $T_{sc}$ . Namely, since it can be considered that the state where the pores are filled with the dendritic ice crystal means the state where smaller particles (beads) are distributed in the porous medium and permeability decreases with the increase of  $T_{sc}$ . The increase of  $C_i$  increases the amount of solute discharged to solidify a predetermined proportion of solution and decreases the solidification temperature. Therefore, in the process of the dissolution of the constitutional super-cooling state, it becomes easy to form and grow the second and third arms of the dendritic ice. The decrease of permeability was caused by filling the pores with the dendritic ice crystals, including second and third arms.

The relationship between the extrapolated values of  $k/k_0$  at  $T_{sc} = 0$  K and  $\chi/\phi$  is shown in Fig. 12. The straight line in Fig. 12 shows equation (1) with  $n = 1.4$ . In Fig. 12, the results of  $k/k_0$  for  $\chi/\phi = 0.7, 0.8$  and  $0.9$  can be joined by another straight line, but this straight line does not pass through  $k/k_0 = 1$  when  $\chi/\phi = 1$ . Namely, if a straight line is drawn in the range of  $0.9 \leq \chi/\phi \leq 1$ , where the volume fraction of

solid phase is small, the gradient of the straight line becomes small. The above tendency has already been discussed in Fig. 8. It is found that the influence of  $C_i$  on the extrapolated values of  $(k/k_0)$  at  $T_{sc} = 0$  K is small in the range from  $C_i = 5$  to 15 wt%. Figure 12 shows the upper limit of the permeability in the case that a super-cooling degree is zero.

As mentioned above, the influence of the initial concentration and the super-cooling degree of the solution just before the start of solidification on the relationship between the permeability and volume fraction of a liquid phase was clarified by the experiments. In the solidification problem accompanying the flow of the solution, the solidification progresses with the concentration change of the solution owing to the transportation of the solute. In the present measurement of permeability, the solute after solidification remains uniform in the porous medium. From now on it is necessary to clarify whether permeability  $k/k_0$  can also be expressed as a function of only  $\chi/\phi$  in the case that the solute flows. Besides, the clarification of the relationship between the micro-structure of ice and permeability by observing the micro-structure of ice growing among the porous medium is also essential.

#### 4. CONCLUSIONS

The measurement of permeability in a state of solidification of a packed bed of glass beads saturated with NaCl-aqueous solution was carried out. The influences of the initial concentration and the super-cooling degree of the solution on the permeability were discussed. The following conclusions were obtained:

- (1) The permeability decreases approximately as a power function with the decrease of the volume fraction of liquid phase in the mushy region. The value of the exponent  $n$  of the equation obtained by the above relationship decreases with the increase of the volume fraction of liquid phase in the mushy region.
- (2) The permeability decreases with the increase of the super-cooling degree of the solution just before the start of solidification.
- (3) The influence of the initial concentration on permeability is small, but the permeability decreases with the increase of the initial concentration.
- (4) An empirical equation of the permeability is obtained as a function of the volume fraction of the liquid phase, the super-cooling degree and the initial concentration of the solution.

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