

Water and Solute Distributions near an Ice Lens in a Glass-Powder Medium Saturated with Sodium Chloride Solution under Unidirectional Freezing

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ABSTRACT: Water and solute distributions near ice lenses, which were formed in uniformly sized glass powder saturated with sodium chloride solution at various concentrations under unidirectional freezing, were measured. At higher initial solute concentrations, ice lenses were thinner and developed at lower temperatures. In the warmer adjacent region, extending about 30 mm from the growing ice lens into the unfrozen area, the water content and solute concentration of pore water decreased from their initial values, while in other unfrozen areas the initial values were maintained. The water content and solute concentration in the warmer adjacent region increased with increasing initial solute concentration.

Introduction

Freezing sometimes causes the water in unfrozen soil to flow toward the freezing front, where regions of ice form that are almost devoid of soil particles; these are called ice lenses. Ice lenses raise the ground surface and cause severe frost heave. A feature of ice lensing is the formation of intermittent ice layers, which result from the repeated generation, growth, and spacing of ice lenses near the freezing front. The formation of ice lenses has been observed in various porous media; the phenomenon is not limited to soil.^{1,2}

When a porous medium containing solution is frozen unidirectionally, water and solute are transported in varying degrees toward the freezing front.^{3–5} This redistribution of soil water has profound effects on the drainage, erosion, and trafficability of surface soils and on the stability of earthen dams, hillsides, building foundations, highways, and airport runways. The redistribution of solute toward the soil surface by the water flow also affects plant growth and agricultural work.⁶ Changes in soil–water salinity can drastically alter the mechanical properties of frozen soils and are of major concern when working with frozen soils.^{7,8} The formation of ice lenses plays an important role in redistributing water and solute. It has been reported that when ice lenses appear, a low water content region is formed near the freezing front.⁹ Chamberlain¹⁰ identified unbonded brine-rich soil zones between ice lenses as layers of potentially low shear strength. It is generally agreed that increasing concentrations of solution cause a significant reduction in the rate of frost heave in fine-grained soils.^{10,11}

The freezing of water containing solute is based on the same mechanism as that when crystal growth excludes solutes.⁵ The first research into the dual processes of solute rejection and redistribution was by metallurgical engineers, and their work provides foun-

dation material. For instance, when ice grows in a solution, solute is rejected from the growing ice surface, establishing a solute diffusion field near the ice surface that depends on the rate of ice growth.¹² This diffusion field influences both the growth rate and shape of ice crystals.^{13,14} When a solution freezes in a porous medium, the solute may be bound to nonmiscible particles, such as clayey colloids or complexes, and these kinds of particles are also excluded from the surface of the ice as it grows.¹⁵ The behavior of solute and particles near ice growth surfaces has recently been studied from an environmental perspective.¹⁶

Conte¹⁷ and Taber¹⁸ carried out the classic studies on frost heave associated with the formation of ice lenses. Later, study of frost heave became a major topic in civil engineering, and numerous theoretical and experimental studies were performed.^{19–24} Jackson et al.²⁵ described ice lens growth, based on experimental data for particle rejection from the surface of ice as it freezes. Miller²⁶ proposed a theory of secondary frost heave, which considered the stress at each phase. This theory is widely used, since it can explain the formation of intermittent layers of ice lenses. However, it cannot explain ice lens formation in some unconfined fine porous powder media.²⁷ Recently, approaches to understanding the formation of ice lenses have appeared that are based on intermolecular forces.²⁸ Nevertheless, neither the redistribution of water and solute in the vicinity of ice lenses nor the impact of solute concentration on ice lens growth have been sufficiently verified.

Thus, the formation of ice lenses in saline porous media is a complex natural phenomenon that remains poorly understood. Knowledge of the effect of different porous media, water content, and concentration and type of solute on the growth of ice lenses is important to gain a better understanding of the redistribution of the water and solute. Here, we present the results of in situ observations of ice lenses in a unidirectionally freezing porous medium that consisted of glass micro-particles saturated with different concentrations of sodium chloride solution. The influence of solute concentration on the formation of ice lenses, and on the

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Table 1. Characteristics of the Powder

mean diameter of particle (μm)	2.2
mean diameter of nanopore (nm)	3.3
specific nanopore volume (cm^3/g)	0.3
specific surface area (m^2/g)	128.6
specific gravity of particle (g/cm^3)	2.12
dry powder density (g/cm^3)	0.85
hydraulic conductivity (m/s)	1.92×10^{-8}
mineral components	SiO ₂ (99.4%) Na ₂ O (little) etc. (very little)

redistribution of water and solute due to ice lensing, is then determined.

Materials and Methods

Sample. The porous medium that we used consisted of Vycor glass particles, which form a fine powder. Table 1 lists some characteristics of the powder. The mean diameter of the particles was estimated from electron micrographs; they were spherical, of roughly equal diameter, and their surfaces were covered with nanopores of uniform diameter. The mean diameter of the nanopores and specific surface area were determined by nitrogen adsorption. The saturated hydraulic conductivity represents an average over a length of 10 cm. Using an NMR technique, we showed that the liquid-water/total-water ratio in powder saturated with pure water at 80% by mass was more than 40% at $-1\text{ }^\circ\text{C}$.²⁷

Distilled, deionized, and degassed water and sodium chloride of 99.9% purity were used. The saturated porous medium was prepared by placing the glass particles in a sealed chamber with the sodium chloride solution, evacuating the chamber, allowing a few days for solid–vapor equilibrium to be established, and then adding solution and allowing the system to equilibrate for 1 day. The prepared sample consisted of unconfined particles, water, and solute and did not contain any gases. The initial water content of each sample was $78 \pm 2\%$ by mass. The solute concentrations of the pore water in the samples were 0, 0.005, 0.01, 0.015, 0.03, 0.05, and 0.1 mol dm^{-3} (samples G000, G005, G010, G015, G030, G050, and G100, respectively). The prepared sample was poured into a sample cell that consisted of two $26 \times 76 \times 1\text{ mm}^3$ glass slides and acrylic spacers. The cell was sealed with silicon, except for a small hole on one side to relieve pressure; its internal volume was $20 \times 70 \times 3\text{ mm}^3$. Some sample cells included two copper–constantan thermocouples, accurate to $0.03\text{ }^\circ\text{C}$, to measure internal temperatures. An identical cell was prepared containing only sodium chloride solution for comparison. The concentrations of the sodium chloride solutions were 0, 0.03, 0.05, and 0.1 mol dm^{-3} (samples S000, S030, S050, and S100, respectively). Each cell was cooled to $2\text{ }^\circ\text{C}$ before the unidirectional freezing experiment commenced.

Unidirectional Freezing Experiment. Each sample was frozen using a unidirectional freezing apparatus that was basically as designed by Nagashima and Furukawa¹³. Figure 1 shows a schematic illustration of the experiment. The sample cell was sandwiched between a pair of copper blocks, each held at a different constant temperature, $T_H = 2.1\text{ }^\circ\text{C}$ and $T_L = -4.0\text{ }^\circ\text{C}$, respectively, by Peltier thermoelements to establish a temperature gradient, $G (=0.21\text{ }^\circ\text{C mm}^{-1})$. The effective length of the temperature gradient was 32 mm. Once the temperature gradient was established, the sample cell, connected to a translational stage with a computer-controlled pulse motor, was pulled at constant rate $V_c (=0.4\text{ }\mu\text{m s}^{-1})$ through the temperature gradient; this continued for 10 000 s. After a short initial transition, the isotherms in the sample advanced at a constant rate, $V_f = -V_c$, under a fixed temperature gradient for a total distance of 4 mm. The temperature gradient, G , was both monitored by the thermocouples in the samples and deduced from the temperatures of the blocks during the experiment.

A microscope equipped with a charge-coupled device camera and videotape recorder system was placed above the cell and

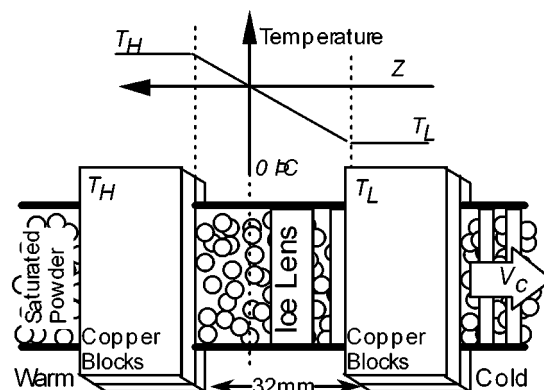


Figure 1. Schematic illustration of the unidirectional freezing experiment. A temperature gradient was established along the Z axis. The solution-saturated powder was placed between two glass slides and pulled at constant speed, V_c , through the temperature gradient. The growth surface of the ice lens lagged behind the $0\text{ }^\circ\text{C}$ isotherm.

used to observe the formation of ice lenses. Images were captured at 1 min intervals; each was divided into a $12.5\text{ }\mu\text{m}$ grid by computer, and the location of the ice lens was recorded. In addition, the temperature of the ice surface was determined by tracing the temperature gradient onto the image.

Measurement of Water and Solute Distributions. First, we checked the water and solute migration in a sample kept at $25\text{ }^\circ\text{C}$ for 300 min. During this time, no significant redistribution of water or solute was observed.

After the sample was unidirectionally frozen with $V_c = 0.4\text{ }\mu\text{m s}^{-1}$ for 4 mm, it was removed from the apparatus and cut into slices a few millimeters thick, parallel to the growth surface of the warmest ice lens. The sample was kept at an ambient temperature of $2\text{ }^\circ\text{C}$ during cutting. Any melting and water migration in the sample during cutting was ignored, since only 1 min elapsed during this procedure. Each slice was placed into a dry oven for 24 h to measure the water content: the ratio of the measured mass of the pure water divided by that of the dry sample. Then, the slice was mixed with a quantity of deionized, distilled water equal in mass to the powder in the sample, and the electric conductivity was measured. The amount of solute in the slice was then estimated from the electrical conductivity, using a previously determined calibration curve. The molar concentration of the pore water in the slice was then calculated.

Results and Discussion

Observation of Ice Lenses in Samples Frozen at a Constant Rate. Once a stable temperature gradient was established, $V_c = 0.4\text{ }\mu\text{m s}^{-1}$ was applied to the sample. The freezing front remained virtually still in the view field of the microscope throughout. Repeated generation and growth of three to five layers of ice lenses at the freezing front were observed. Figure 2 shows the ice lenses observed in sample G000, while V_c was applied.³² Each ice lens appeared as a planar layer in this porous medium, so we could clearly identify the boundary and location of the growing surface of the ice lens.^{2,27} During the experiment, no air bubbles were seen in the view field, suggesting that the sample remained saturated during the experiment, since air bubbles appear at the ice–water interface when water is frozen at $V_c = 0.4\text{ }\mu\text{m s}^{-1}$.²⁹

Under V_c , the layers of ice lenses had almost the same thickness and spacing. Figure 3 shows the mean thickness of the ice lenses and the mean spacing between them as a function of the initial solute concentration.

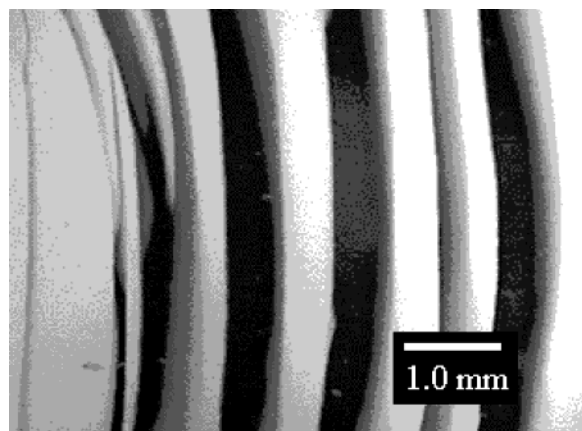


Figure 2. Ice lenses observed in sample G000 while $V_c = 0.4 \mu\text{m s}^{-1}$ was applied. The right-hand side is colder, and the left-hand side is warmer. The ice lenses appear black, since the background can be seen through the ice lenses, and the porous medium appears white.

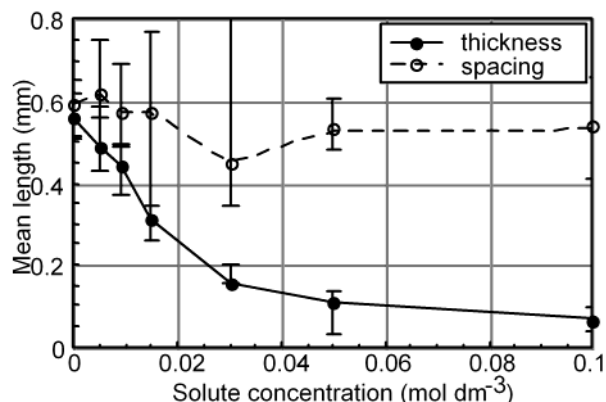


Figure 3. Mean thickness of ice lenses and mean spacing between ice lenses as a function of the initial solute concentration of the pore water. Open circles show the mean thickness of ice lenses, and solid circles show the mean spacing between ice lenses.

The ice lenses were thinner when solute concentration was high. Spacing was not uniform and not concentration-related. If we assume that the sum of the thicknesses of the ice lenses in a sample corresponds to the amount of frost heave, the results suggest that the amount of heave decreases with increasing solute concentration in the pore water. This agrees with the results reported by Chamberlain.¹⁰

During freezing at V_c , the ice lenses that formed and grew remained in essentially the same place in the view field of the microscope, implying that they formed at a constant temperature. The location in the view field varied with the initial solute concentration. On completion of each experiment, we located the growth surface of the warmest ice lens in each sample (Figure 4). The higher the concentration, the colder the location at which the ice lens formed. If we assume a linear temperature gradient, G , in the sample, the temperature at the growth surface in the sample can be determined from the right axis in Figure 4 by using the location of the surface and $G = 0.21 \text{ }^\circ\text{C mm}^{-1}$. The dashed line in the figure indicates the molar freezing depression of bulk sodium chloride solution. The temperature at the growth surface of the warmest lens is lower than that at the ice–liquid water interface in a

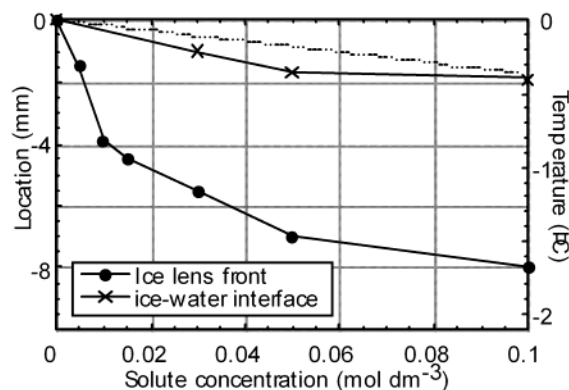


Figure 4. Location of the growth surface of the warmest ice lens, when the sample was moved at $0.4 \mu\text{m s}^{-1}$ for 4 mm. The left axis indicates the location of the growth surface of the warmest ice lens (solid circle) and ice–liquid interface (cross). Zero on the axis equals the ice–liquid interface observed in sample S000 (pure water), and negative values refer to location on the cold side. The right axis indicates the temperature at the growth surface, calculated from the linear temperature gradient, $G = 0.21 \text{ }^\circ\text{C mm}^{-1}$. The dashed line indicates the molar freezing depression of bulk sodium chloride solution T_f , calculated using $T_f = iK_fC$, where C is the molar concentration of the solution, i is the dissociation constant of sodium chloride ($=2$), and K_f is the molar freezing point constant ($=1.86 \text{ }^\circ\text{C mol}^{-1}$).

solution at the same concentration. The slope of the temperature at the growth surface approaches the slope of the molar freezing depression curve when the solute concentration is high. These results suggest that solute concentration near the growth surface increases considerably above its initial value, because of solute rejection from the ice lens surface, and that this effect is particularly apparent with a lower initial solute concentration. The results also suggest that the solute concentration near the ice surface may increase if there are particles.

Any particle on the growth surface of an ice lens is subject to two counteracting forces: one attractive and one repulsive.^{2,28} The attractive force comes from viscous drag due to fluid flow around the particle, and the repulsive force originates from intermolecular forces between the ice lens, the particle, and the liquid water film between them. An ice lens will keep growing if the repulsive force is greater and will stop if the attractive force becomes greater. It is thought that the repulsive force depends on the solute concentration of the water film between the ice lens and the particle. The dependence of the thickness of the water film on the solute has been investigated.^{30,31} In this context, increasing the solute concentration of the pore water causes ice lenses to develop at a colder location and be thinner.

Water Redistribution near the Growth Surface of the Warmest Ice Lens. Figure 5 shows the distribution of the water content near the growth surface of the warmest ice lens in a sample, just after the sample was moved 4 mm. The region $Z < -15 \text{ mm}$ corresponds to the region where the sample was first frozen, immediately after T_H and T_L were applied. In this region, the water content in each sample rose slightly above its initial value ($\sim 0.78 \text{ g/g}$), while no ice lens was observed. In the region from $Z = -15$ to -4 mm , where the freezing front gradually advanced, nonuniform

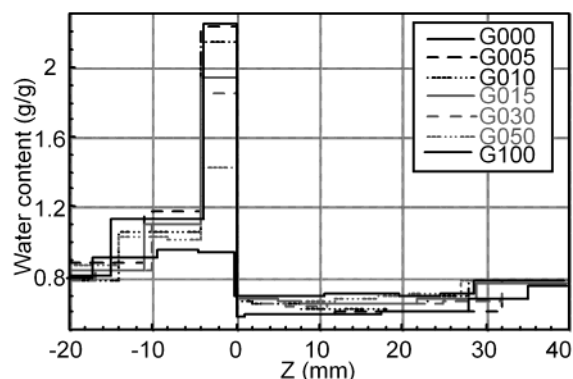


Figure 5. Distribution of the water content near a growth surface of the warmest ice lens, when $V_c = 0.4 \mu\text{m s}^{-1}$ was applied to the sample for 4 mm. The growth surface is located at $Z = 0$. The left-hand side of the figure is colder, and the right-hand side is warmer.

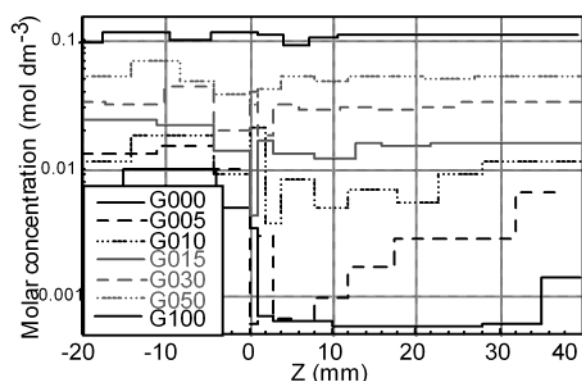


Figure 6. Distribution of the solute concentration of pore water near the growth surface of the warmest ice lens.

layers of ice lenses were observed, and the water content increased. In the region from $Z = -4$ to 0 mm, where V_c was applied to the sample, a series of ice lenses of virtually identical thickness and spacing was observed (see Figure 2). The water content decreased with increasing initial solute concentration. This change in water content is derived from the dependence of ice lens thickness on initial solute concentration, as shown in Figure 3. There was a region of lower water content that extended about 30 mm from the growth surface into the unfrozen area. Increasing the initial solute concentration increased the water content of this region and decreased its length. These results indicate that water migration from the unfrozen area to the freezing front depends on initial solute concentration.

The saturated hydraulic conductivity of the powder, measured macroscopically, was $1.92 \times 10^{-8} \text{ m s}^{-1}$, which is less than the growth rate of the ice lenses ($0.4 \times 10^{-6} \text{ m s}^{-1}$). This implies that the supply of water to the growth surface of the ice lens lags behind its growth, causing a region of lower water content in the sample as the ice lens grows.

Solute Redistribution near the Growth Surface of the Warmest Ice Lens. The solute concentration distribution of pore water near the growth surface of the warmest ice lens was calculated from the water content and the electric conductivity. Figure 6 shows the distribution of solute concentration, using the same Z axis as Figure 5. In the region where no ice lenses were observed ($Z < -15$), the pore water retained its

initial solute concentration. In the region where ice lenses of uniform thickness were observed under V_c ($-4 \text{ mm} < Z < 0 \text{ mm}$), no significant change in solute concentration was observed, while the water content and electric conductivity values increased. The solute concentration of the pore water between ice lenses became several times higher than the initial value, presumably because the ice lenses almost totally rejected the solute. In the region $Z = 0-3 \text{ mm}$, the solute concentration of the pore water varied from sample to sample. For those samples in which the warmest ice lens had grown sufficiently before freezing was stopped (samples G000, G010, G030, and G100), the solute concentration in this region was high, decreasing with distance from the growth surface. Conversely, for samples in which the warmest ice lens had only just started to grow when freezing was stopped (samples G005, G015, and G050), the solute concentration in this region was low. These results suggest that the increased solute concentration near an ice lens is because of the growth of the ice lens. In the region from $Z = 3$ to $Z \sim 30 \text{ mm}$, the solute concentration of the pore water fell below the initial value. The solute concentration of this region increased, and its length decreased, with increasing initial solute concentration, while in the warmer part of the sample the pore water retained the initial solute concentration.

When an ice grows in a solution, the diffusion length, l , of the solute in front of the ice–water interface is estimated to be $l = D/V$ by two-dimensional theory, where D is the diffusion coefficient of the solute in the solution and V is the movement velocity of the interface. Since D for sodium chloride is about $1.5 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ with solute concentrations of $0-5 \text{ mol dm}^{-3}$, and the growth rate of the ice lens was $= 0.4 \mu\text{m s}^{-1}$, in this case $l = 3.8 \text{ mm}$. The solute that is rejected at the growth surface of the ice lens should therefore diffuse within 4 mm of its surface, and outside 4 mm the solute concentration should reach equilibrium. In our experiment, when the ice lens had grown sufficiently (samples G000, G010, G030, and G100), the solute concentration in the region extending a few millimeters from the growth surface of the warmest ice lens into the unfrozen part of the sample decreased with distance from the ice surface. The length of the solute diffusion field caused by the growth of an ice lens was within this distance, although we did not measure below 1 mm. On the other hand, the solute concentration was lower than the initial value in a region extending about 30 mm into the unfrozen area from the ice surface. This redistribution of solute may arise not only from solute diffusion but also from water migration due to ice lensing and interactions between the solute and the particles.

Conclusions

Unidirectional freezing experiments were carried out on an unconfined, uniformly sized, porous medium consisting of glass microparticles saturated with sodium chloride solution at different concentrations. The formation of ice lenses in the porous medium was observed to depend on the initial solute concentration of the pore water. The distributions of water content and solute concentration near the growth surface of the warmest ice lens in the porous medium were measured. When

the initial concentration was high, thinner ice lenses grew, and at lower temperatures. In a warmer, adjacent region that extended about 30 mm into the unfrozen area from the growing ice lens, the water content and solute concentration fell below the initial value, while in other unfrozen areas initial values were maintained. The water content of this warmer region increased with high initial solute concentration, which also increased the solute concentration of this region and decreased its length.

The grain size of the samples that we used corresponds to that of silt. When dealing with water and solutes in actual soil, it is important to consider migration due to ice lensing. Water and solute redistribution in samples saturated with other solutions will be measured in the future.

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- (32) Video images of Figure 2 are available free of charge via the Internet at <http://soil.en.a.u-tokyo/~mizo/openpro/icelens/v04.mpg>.

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