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Relationship between growth rate and supercooling in the formation of ice lenses in a glass powder

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Abstract

When a saturated powder is cooled below the freezing temperature of the interstitial water, the water sometimes forms ice lenses, which are layers of ice from which all particles have been expelled. To clarify the relationship between the rate of ice lens growth and the temperature at the growth surface, ice lens formation in a water-saturated, fine powder consisting of uniform-sized, spherical, glass micro-particles was observed in situ, using a microscope and a directional freezing apparatus. When the powder was frozen rapidly, the ice lenses grew faster, at lower temperatures, and formed thinner layers. The ice lens grew at a rate that was linearly correlated with the degree of supercooling at the growth surface, and was significantly slower than that of ice in water. The results suggest that the particles near the growth surface inhibit ice lens growth. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

When water containing fine particles, such as soil, is frozen directionally, the water sometimes forms regions of ice that are almost devoid of particles; these regions are called ice lens [1,2]. When ice grows in water, its growth rate is expected to be proportional to the degree of supercooling at the growth surface. However, if there are particles in the water, the particles affect the growth of the ice. Vignes and Dijkema [3]

confirmed that the growth rate of ice in a capillary depends only on the degree of supercooling at the growth surface. Ozawa and Kinoshita [4] observed ice growth on a microporous filter, and showed that the growth rate is proportional to the degree of supercooling at the freezing front ($3.4 \mu\text{m s}^{-1} \text{C}^{-1}$), and is independent of pore size. Watanabe et al. [5] experimentally determined the proportionality constant of growth rate of an ice lens in silt ($1.0 \mu\text{m s}^{-1} \text{C}^{-1}$). However, we still need to understand the growth process of ice lenses in fine powder. This paper presents the results of in situ observations of ice lenses in a powder. The relationship between the growth rate of the ice lenses and supercooling at the growth surface, and the influence of surrounding particles on ice lens growth, are then determined.

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2. Experiments

The sample powder that we used consisted of uniform spherical glass particles with a diameter of $2.2\ \mu\text{m}$ [6]. The powder was saturated with distilled, deionized, and degassed water (Sample P). The initial water content was $78 \pm 2\%$ by mass. The sample was poured into sample cells that consisted of two glass slides and acrylic spacers. The internal volume of each cell was $20 \times 70 \times 3\ \text{mm}^3$. Some sample cells included two thermocouples, accurate to 0.03°C , to monitor internal temperatures. An identical cell was prepared containing only distilled water for comparison (Sample W). Each cell was cooled to 2°C before being frozen.

The samples were frozen using the directional freezing apparatus (Fig. 1) that was designed by Nagashima and Furukawa [7]. The sample cell was forced to move transversally at an arbitrary constant rate, R , in a fixed temperature gradient. In this experiment, the temperature gradient $G = 0.19^\circ\text{C}\ \text{mm}^{-1}$ was observed when the sample cell was pulled at a rate of $0.04 \leq R \leq 2\ \mu\text{m}\ \text{s}^{-1}$. A microscope was combined with the directional freezing apparatus. Images were captured at 1-min intervals; each image was divided into a $12.5\text{-}\mu\text{m}$ grid by computer, and the location of ice lenses was recorded. The temperature at the growth surface of the ice lens was determined by tracing the temperature gradient onto the image.

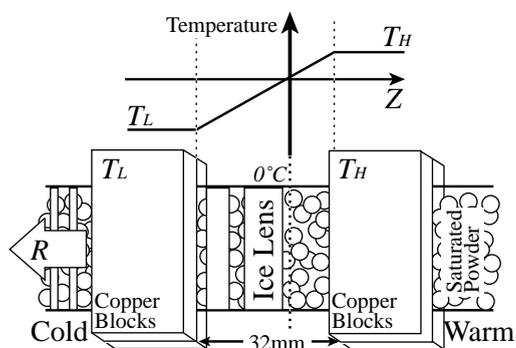


Fig. 1. Schematic illustration of the unidirectional freezing experiment. A temperature gradient was established along the Z -direction. The sample cell was pulled at constant rate, R , through the temperature gradient.

3. Results

3.1. Directional freezing of water-saturated powder

Fig. 2 shows the temperatures measured by the thermocouples in Sample P during the experiment. Fig. 3 shows the ice lenses observed in Sample P, when $R = 0.4\ \mu\text{m}\ \text{s}^{-1}$ was applied for 250 min. At time zero, the sample cell was placed in the directional freezing apparatus and the freezing

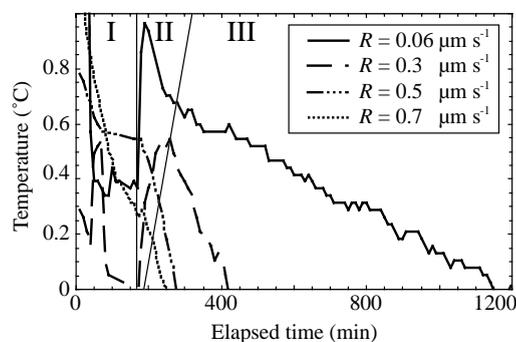


Fig. 2. Change in temperature in powder during directional freezing experiment. The pulling rate R is measured in $\mu\text{m}\ \text{s}^{-1}$. In regions I, II and III, the sample was frozen without sample cell movement, with R before fixed temperature gradient was established, and with R in the fixed temperature gradient, respectively.

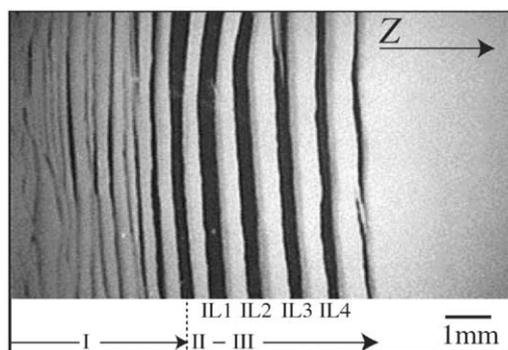


Fig. 3. Ice lenses in water-saturated powder. Right-hand is warmer and left-hand is colder. The temperature gradient established along the Z -direction. IL1-4 are ice lenses observed during $R = 0.4\ \mu\text{m}\ \text{s}^{-1}$ was applied. The ice lenses appear black since background can be seen through ice lenses, and the powder appears white.

front began to advance in the sample. It took about 100 min to establish a fixed temperature gradient (Fig. 2-I). During this stage of freezing, a series of ice lenses formed from the cold side as an intermittent layer (Fig. 3-I). After 150 min, R was applied to the sample. After a short initial transition stage (Fig. 2-II), the temperature decreased linearly with time (Fig. 2-III), since the thermocouples moved at a constant rate. The length of the initial transition stage decreased with increasing R . Under R , the freezing front was virtually fixed in the view of the microscope, and repeated generation and growth of ice lenses was observed at the freezing front (Fig. 3-III, IL1-4). At this stage of freezing, ice lenses grew at a constant rate and formed with virtually the same thickness and spacing.

3.2. Ice lens formation under a constant pulling rate, R

Fig. 4 shows the growth rate of an ice lens and that of ice in water. The ice lens grew more slowly than R , while the ice in water grew at almost the same rate as R . When R was slower than $0.1 \mu\text{m s}^{-1}$, a single ice lens continued to grow until it extended more than 4 mm, and the growth rate was close to R . On the other hand, when R was faster than $0.2 \mu\text{m s}^{-1}$, new ice lenses appeared repeatedly at the freezing front, and formed several layers while the sample was moved above 4 mm. With increasing R , the growth rate of

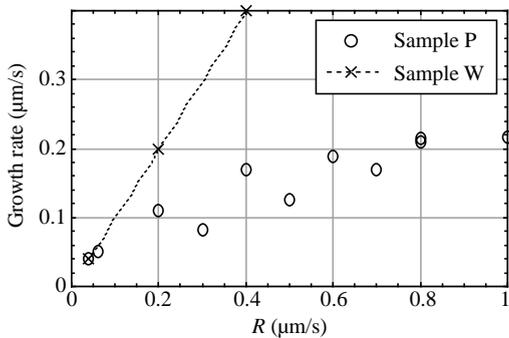


Fig. 4. Relationship between growth rate of ice and R . The Sample P and Sample W are the ice lens in water-saturated powder and ice in water, respectively.

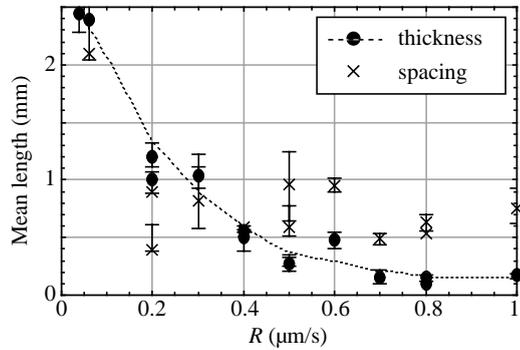


Fig. 5. Mean thickness of the ice lens and mean spacing between them as a function of R .

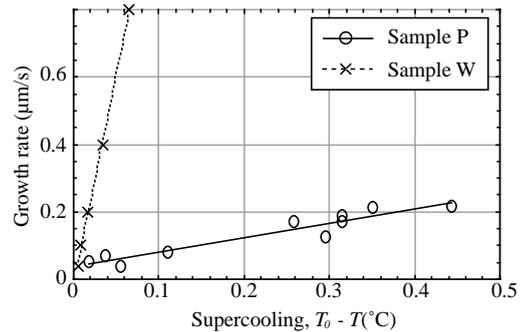


Fig. 6. Relationship between growth rate of ice and degree of supercooling at the growth surface.

individual ice lenses lagged behind R , and approached $0.24 \mu\text{m s}^{-1}$ asymptotically. When R was faster than $2.0 \mu\text{m s}^{-1}$, no ice lenses were observed in the sample.

Fig. 5 shows the mean thickness of ice lenses and the mean spacing between them as a function of R . Ice lenses were thinner when R was high, while no good relationship between spacing and R was found. Fig. 6 shows the relationship between the growth rate and the degree of supercooling $T_0 - T$ at the growth surface. When R was high, ice lenses grew faster, at lower temperatures. The growth rate of the ice was linearly correlated with the degree of supercooling ($0.43 \mu\text{m s}^{-1} \text{ } ^\circ\text{C}^{-1}$), and was about one-thirteenth the growth rate of ice in water ($12.3 \mu\text{m s}^{-1} \text{ } ^\circ\text{C}^{-1}$).

4. Discussion

4.1. Ice lens thickness

Some studies have examined particle rejection from the advancing ice surface, and an inverse relationship between the critical freezing rate for particle rejection and particle radius has been proposed [8–10]. As water containing suspended fine particles freezes directionally, the particles are rejected from the ice growth surface and then accumulate as a layer near the surface; the ice then suddenly engulfs all the particles once the layer reaches a certain thickness [11]. A similar situation occurs during ice lensing. To generate a new ice lens, the ice must engulf the layer of particles that accumulated at the surface of the previous ice lens. The thickness of the particle layer depends on the thickness of the previous ice lens, L_{il} . If we assume that the critical growth rate of an ice lens is determined by L_{il} (as is particle radius for the rejection of an isolated particle), the relationship is expressed as $R_{il} = A/L_{il}$. Substituting the experimental results (Fig. 5) into this relationship, then $A = 0.86 \times 10^{-4} \text{ s}^{-1}$. Although the physical meaning of A has not been investigated, it may depend on the difference in the surface energy between particle–water and water–ice, and on the viscosity of the water and the packing of the particles near the ice lens.

4.2. Ice lens growth rate

Considering the difference between the chemical potential of the pre-melted water film at the growth surface of an ice lens and that of the water in the pores between particles, Kuroda [12] represented the growth rate of an ice lens, R_{il} , as follows:

$$R_{il} = \frac{Q_m(T_0 - T)/T_0 - p\Delta v}{akT/D + d/k}, \quad (1)$$

where Q_m is the latent heat of fusion of ice per molecule, T_0 is the melting temperature of ice, p is the overburden pressure, Δv is the molecular volume difference between ice and water, a is the molecular distance, k is the Boltzmann factor, D is the self-diffusion coefficient of a water molecule in

pre-melted water film, K is the hydraulic permeability of the powder, and d is the length in which K is effective. According to Kuroda's theory, the growth rate of an ice lens is affected by the resistance to the freezing process and the diffusion process, akT/D , and the hydraulic resistance, d/K . In our experiment, the growth rate of an ice lens was $R_{il} = 0.43 (T_0 - T) + \text{const. } \mu\text{m s}^{-1}$ (Fig. 6), and p could be ignored. Using $T_0/Q_m = 2.71 \times 10^{22} \text{ kJ}^{-1}$, $d = 0.02 \text{ m}$ [6], $a = 3.00 \times 10^{-10} \text{ m}$, $k = 1.38 \times 10^{-23} \text{ J K}^{-1}$, and $K = 6.54 \times 10^{16} \text{ m}^2 \text{ s J}^{-1}$ [6], we obtain $D = 1.3 \times 10^{-14} \text{ m}^2 \text{ s}^{-1}$. The value of D is five orders of magnitude smaller than that of bulk water at 0°C [13], and is about one-twentieth that of the pre-melted water film at the ice–air interface at -1.5°C [14]. The results show that surrounding particles, which surface has an attractive interaction with water molecules, inhibit the mobility of water molecules near the ice lens.

5. Conclusions

Directional freezing experiments were carried out on an unconfined, water-saturated powder consisting of uniform-sized, glass micro-particles. The formation of ice lenses in the powder was observed; the thickness and growth rate of the ice lenses, and the temperature at the growth surface, were investigated under a constant pulling rate, R . When R was high, the ice lenses grew faster and at lower temperatures. The growth rate of the ice lenses was linearly correlated with the degree of supercooling at the growth surface, and was significantly slower than that of ice in water. These results suggest that particles near to the growth surface inhibit ice lens growth through the inhibition of the mobility of water molecules.

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