The Influence of Temperature Gradient on Sintering of Ice Particles

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ABSTRACT

The instability of snowpacks and the occurrence of avalanches is the reason for the current interest in studying the effects of temperature gradients on dry snow metamorphism. Snow on the ground undergoes complex changes in the snowpack structure due to macroscopic temperature gradients. To simplify the situation, we began with a two-dimensional array of high-purity laboratory-grown ice spheres laid in contact with each other. The center of mass between the particles and the contact bond growth between two ice spheres were observed using optical microscopy and recorded as a function of time in order to understand the processes occurring. High-resolution images of the bonding between particles were recorded using a cold stage-equipped scanning electron microscope. Three-dimensional images of specimens were obtained using X-ray computed tomography. In addition, a series of experiments was performed with doped ice, in which changes in the sintering processes due to the dissolved impurities were found.

Keywords: dry snow metamorphism; temperature gradient; SEM; micro CT

INTRODUCTION

Snow on the ground undergoes complex changes in both structure and physical properties, which impacts energy, mass and chemical exchange between the ground and the atmosphere and relates to polar firn formation and avalanche release (Colbeck, 1991; Schweizer, 2008). The mechanisms for snow metamorphism have been studied for a long time, and have been observed to be strongly dependent on thermal conditions. Kingery (1960) observed the growth rate of ice bonds during the sintering of ice spheres and he pointed out that the process of ice sintering is due to mass transfer by surface diffusion. Kuroiwa (1962) examined ice sphere sintering in both ice saturated air and kerosene and concluded that both volume diffusion and surface diffusion play predominant roles under isothermal conditions. Vapor transport is widely accepted as a significant mechanism during sintering. Snow subjected to a macroscopic temperature gradient experiences a quite different microstructural change from the one in a macroscopic uniform temperature. As such, the current research examined the change of ice microstructure both under low temperature gradients and high temperature gradients and the emphasis was placed on the growth of necks in between ice particles. Both sintering, which is the commonly observed equilibrium behavior under macroscopic equi-temperature conditions (Miller, 2003), and faceted ice crystal growth, which is due to kinetic microstructural changes under a macroscopic temperature gradient, were observed in our research.

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EXPERIMENTAL

Sample preparation was carried out at -10°C in a cold room. We produced spherical ice particles by dropping pre-cooled undoped water or sulfuric acid doped water into liquid nitrogen. The frozen water drops were picked up using a clean sieve and arranged in a sample holder to form a two-dimensional (2D) array (Figure 1). The average diameter of the ice spheres was 4 mm. The holder was made of plexiglass with copper pieces at the top and the bottom for efficient heat transferring. The whole sample was seated vertically into a temperature controlled system, with the top and bottom copper pieces in contact with peltier coolers, whose temperatures were controlled by two temperature controllers independently (Figure 2). To make both the heat flux and vapor flux go from bottom to top, the temperature of the bottom was set to be higher than the temperature of the top. The whole system was set up in a cold room at -10°C. The size of neck between ice spheres was observed and measured using a stereo optical microscope with transmitted light. For acquiring high-resolution images and measuring the microchemistry, an environmental field emission gun scanning electron microscope (SEM) equipped with a Gatan cold stage was used. For examining the inner structure of the necks between the spheres, three-dimensional (3D) models were built using a Skyscan 1172 X-ray computed tomography (micro CT).

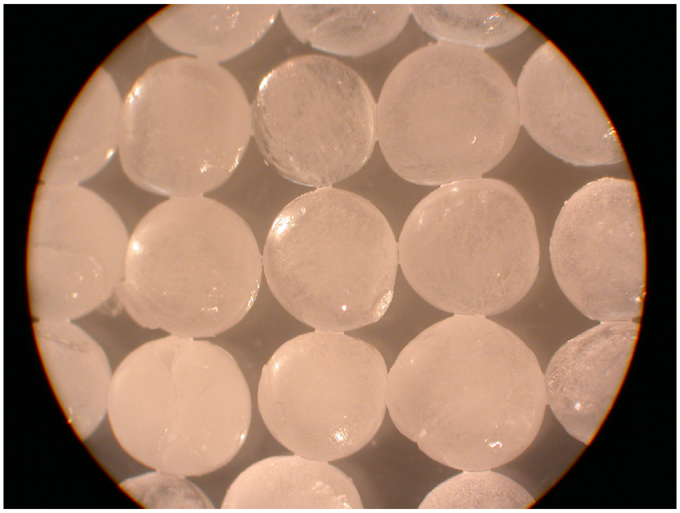

Figure 1. Optical micrograph of 2D array of ice spheres. The average ice sphere diameter is 4 mm.

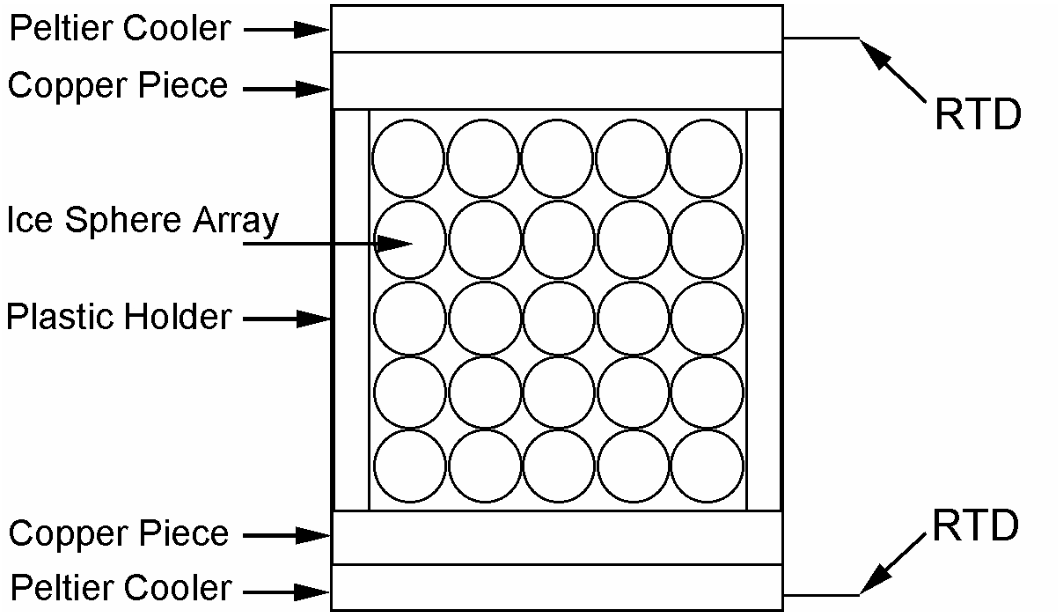

Figure 2. Schematic of temperature gradient control set-up.

RESULTS

The early stages of bonding

The necks between adjacent ice spheres were observed to have their highest growth rate at the earliest stages of sintering. The size of the neck doubled in the first hour after the spheres were laid in contact with each other. As the neck grew, the growth rate decreased significantly. High resolution images of paired ice spheres at the earliest stages of growth show that in between two adjacent ice spheres the ice particles have a preferred growth direction from one ice sphere towards the other (Figure 3). This phenomenon has also been observed by Adams (2008).

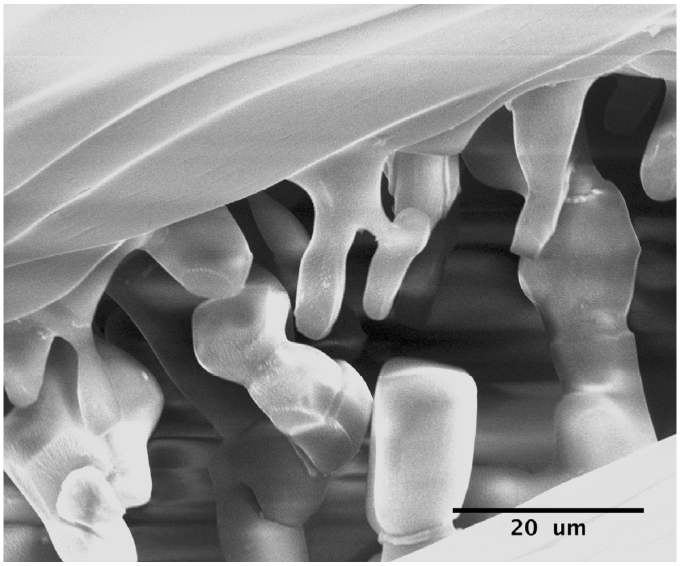

Figure 3. Secondary electron image showing growth of ice particles from one ice sphere towards an adjacent one.

Sintering under low temperature gradient

In the limit of no temperature gradient, a very low temperature gradient can be considered to be an isothermal condition. The temperatures of the bottom and the top of a 2 cm × 2 cm holder were regulated to be -10±0.1°C by a chilled solution and measured through two resistance temperature detectors (RTDs). Ice spheres made by de-ionized (DI) water sintered at -10±0.1°C were examined for two weeks. The bonding process was recorded through photomicrographs taken at various time intervals. Two kinds of necks were observed. One was formed by fine protrusions from adjacent ice particles containing numerous pores inside. After two weeks, the necks tended to densify by filling up the pores with water molecules, but with only a slight change occurring to the neck diameter (Figure 4). The other kind of bonding process began with a solid neck. In two weeks, an increase of neck size was observed. In both the bonding processes, the shrinkage of the ice particles was observed, especially around the neck areas. Together with the neck growth, particle shrinkage resulted in the increase of dihedral angle, a feature which has been reported to impact the sintering rate (Zhang, 1995).

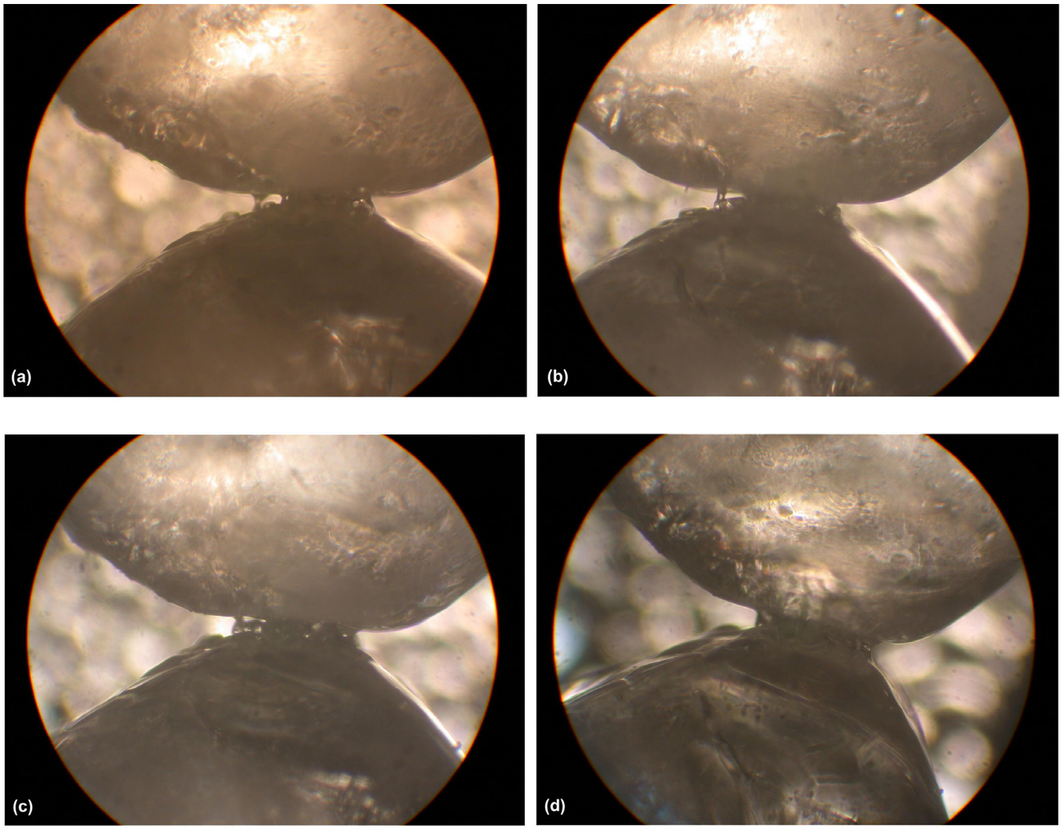

Figure 4. Optical micrographs showing neck growth between two ice spheres by filling up the pores at low temperature gradient. (a) initial stage; (b) after 3 days; (c) after 6 days; (d) after 18 days.

In addition, small faceted protrusions were also observed on the surface of some ice spheres but without connection to adjacent ice particles. The protrusions disappeared and resulted in a smooth ice sphere surface later during the sintering. This rounding process is commonly observed as equilibrium metamorphism occurring at low temperature gradients (Figure 5).

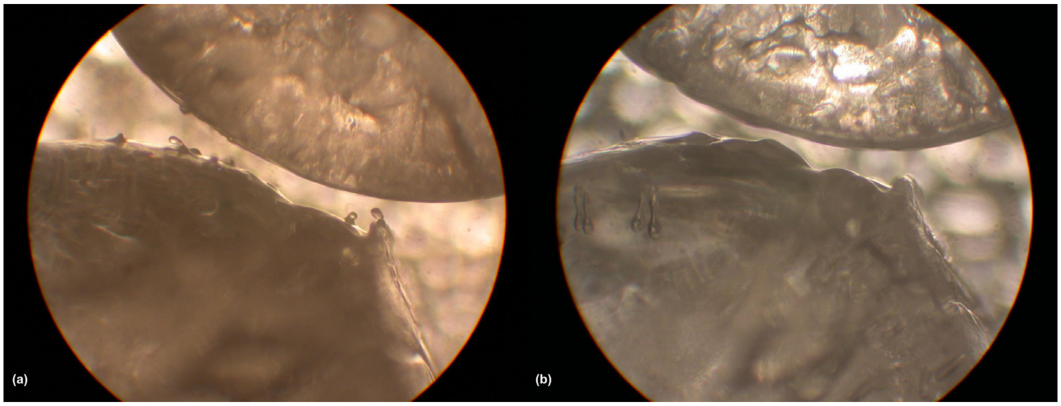

Figure 5. Optical micrographs showing equilibrium rounding process at a low temperature gradient. (a) initial stage; (b) after 18 days.

Impurity impact

Adding some chemical impurities to the ice spheres reduced the growth rate. Figure 6 shows the successive stages of ice sphere sintering. The ice spheres were made from 500 ppm sulfuric acid doped water.

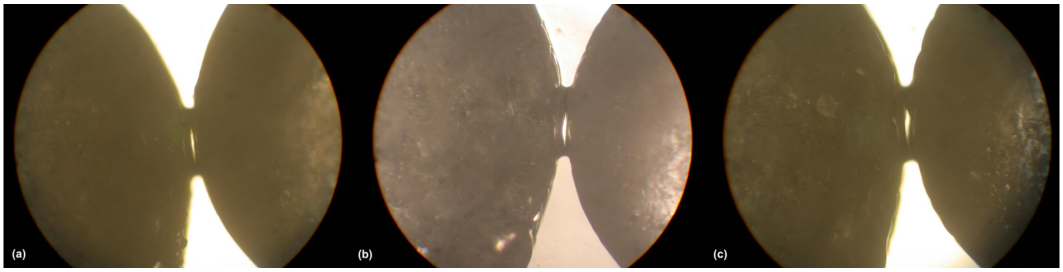

Figure 6. Optical micrographs showing that by adding a chemical impurity little change in the neck occurred during the observation period. (a) initial stage; (b) after 15 days; (c) after 48 days.

Because impurities strongly segregate to grain boundaries (Mulvaney et al., 1988; Cullen and Baker, 2000), it is suggested that the impurity particles decrease the bonding rate by impeding grain boundary diffusion, which is the dominant mechanism during metal powder sintering (German, 1996) and also has a possibility to play a significant role in ice sintering.

Vacancy flow

3D models of ice spheres were obtained through the use of a micro CT. Without physically sectioning the specimen, the inner structure of neck was examined by cutting through the model in a computer program (Figure 7). The inside structural change was also observed by making the 3D models partially transparent by decreasing the opacity coefficient (Figure 8). Both Figure 7 and Figure 8 show that more voids occur inside or on the surface of the ice particles crowded together around the neck during ice sintering. This indicates that vacancies generated at the neck flow to the ice particles and locate inside or on the surface. In fact, vacancy flow is accompanied by volume diffusion and surface diffusion, by which mass is transferred in the opposite direction, i.e. from the ice particles to the neck. This observation resembles the vacancy diffusion in ice sintering noted by Kuroiwa (1962).

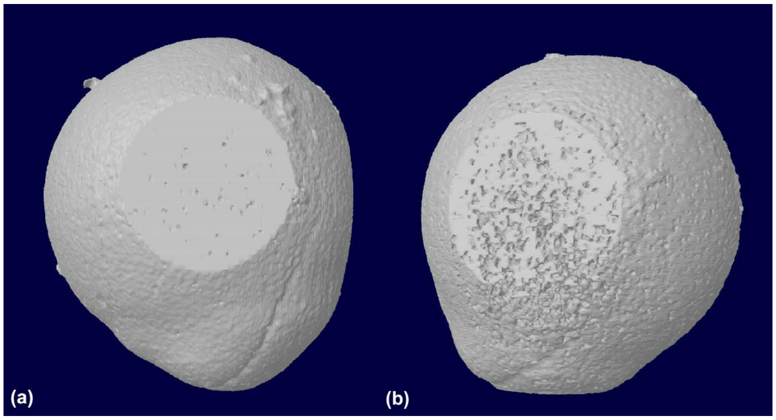

Figure 7. Sectioned micro CT images of an ice sphere showing the inner structure of the neck. (a) after 1 hour; (b) after 75 hours.

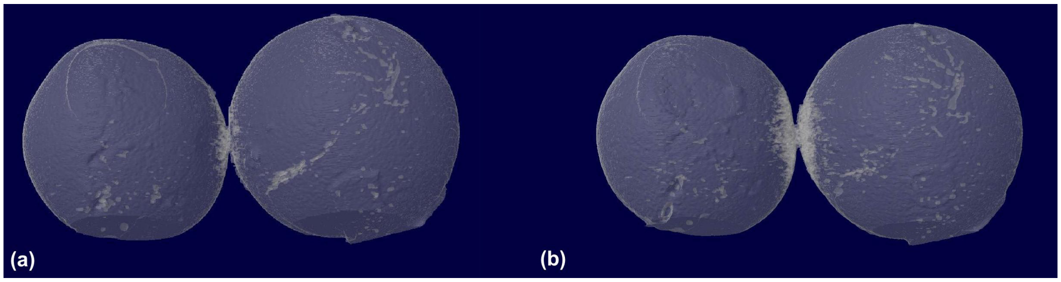

Figure 8. Partially transparent micro CT images of ice spheres showing the voids crowded around the neck. (a) after 1 hour; (b) after 25 hours.

Kinetic growth under high temperature gradient

In this case, the vertical temperature difference was controlled to be 10°C across a 2 cm × 2 cm ice sphere array, by maintaining the temperature of the bottom and the top at -20±0.1°C and -10±0.1°C, respectively. Therefore, the temperature gradient of the ice sphere array was 500°C/m. The necks were divided into two groups (Figure 9). Group 1 contains all the necks whose normal directions were perpendicular to the direction of vapor flow. Group 2 contains all the necks whose normal directions were parallel to the direction of vapor flow. The behaviors of the necks in Group 1 were similar to the ones under a low temperature gradient. While in Group 2, the size of the necks increased rapidly by adding faceted ice particles around the necks. While some of the newly grown ice particles were irregular shapes, most of them were typically hexagonal plates (Figure 10). Such faceted ice particles were also observed underneath the ice spheres. This indicates that a large temperature gradient results in a high water vapor gradient. The water vapor generated from the ice spheres in the lower part is transported along the temperature gradient to the higher part, which condenses around necks and surface of ice spheres.

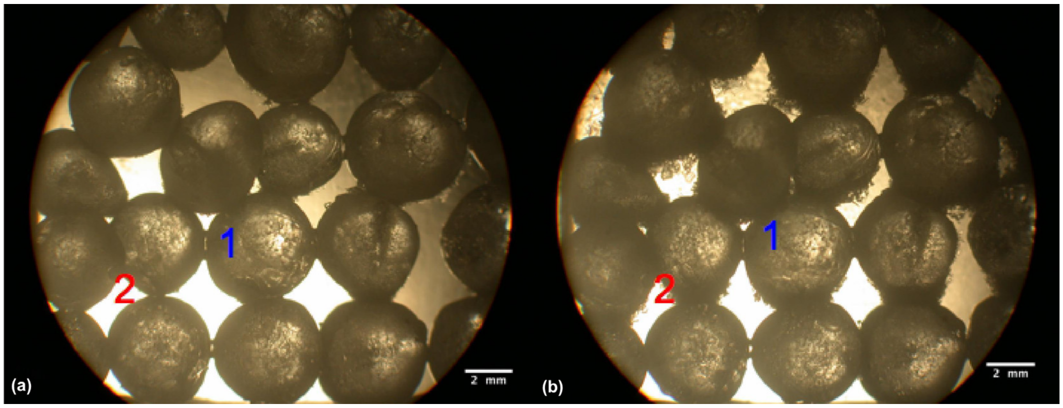

Figure 9. Optical micrographs showing the growth of necks (separated into Group 1 and Group 2 by their normal directions) under a high temperature gradient. (a) initial stage; (b) after 8 hours.

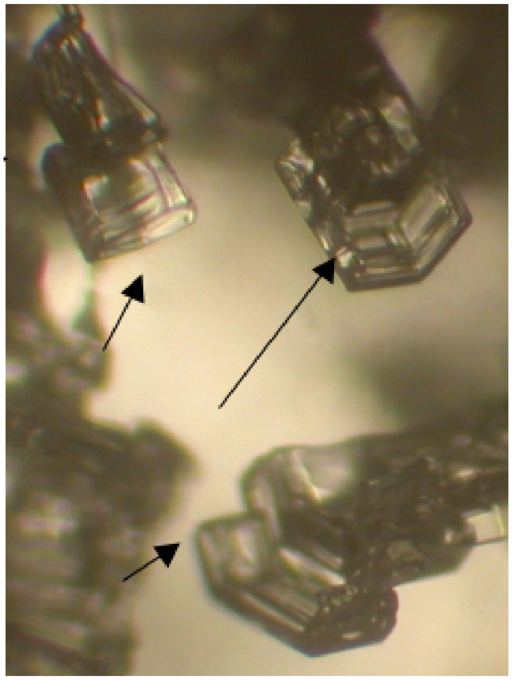

Figure 10. Optical micrographs showing faceted ice particle grown around necks and underneath ice spheres.

SUMMARY

We produced 2D ice sphere array from both DI water and impurity-doped water and set up a temperature control system for examining ice sintering under different conditions. In order to study the structural change of the necks between ice spheres during metamorphism, we combined the use of optical microscope for daily observation, SEM image for detailed microstructural and chemical detection, and micro CT for inner features modeling. Based on the observations and measurements, we have a better understanding of the sintering process of ice particles and investigated the possibility of mechanisms involved.

Under a low temperature gradient, equilibrium metamorphism occurs, besides vapor transport, other mechanisms including volume diffusion, surface diffusion and grain boundary diffusion also play significant roles on neck structure formation and neck growth rate. Under a high temperature gradient, kinetic ice growth takes place, ending with faceted ice particles around necks and underneath ice particles.

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