In-situ Observations of Snow Sublimation using Scanning Electron Microscopy

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ABSTRACT:

The morphology of a snowflake and the sublimation-induced morphological changes under high vacuum are reported. Observations of freshly fallen snow were made using a scanning electron microscope (SEM) equipped with a cryo-system. The snow specimen was initially controlled at -180°C for examining the initial morphology, which consisted of snow crystals in various geometries and dimensions. Specimen sublimation was initiated by increasing the temperature. Severe morphological destructions due to sublimation occurred at round -150°C. *In-situ* structural changes of an individual snowflake were recorded by taking images intermittently. While most of the features commonly reported from field sublimation were occurring, the rounding process from faceted edges was not observed in this high-vacuum environment.

Keywords: fresh snow, SEM, sublimation

INTRODUCTION

Snowflakes generally begin to form by ice nucleation on solid particles in the upper atmosphere. Since their structure is highly sensitive to the local temperature and humidity conditions, snowflakes encompass a huge number of crystal types: plates, (often-hollow) columns, dendritic crystals, needles, prisms capped by plates or dendrites, bullet rosettes, sometimes capped by plates or dendrites (Legagneux *et al*., 2003). Snowflakes can be either individual snow crystals or aggregations of two or more snow crystals, resulting in a size distribution from ~50 μm to several centimeters (Hobbs, 1974; Gray and Male, 1981).

As soon as snow reaches the ground, it begins to undergo continuous metamorphism, in which the earliest stage is “destructive metamorphism” (Colbeck, 1983). In this process, snowflakes are broken apart due to mass sublimation and result in many more individual crystals. In the absence of a temperature gradient, these new seed crystals favor cylindrical or spherical shapes with rounded edges replacing the initially crystalline facets (Colbeck, 1983).

In this paper, freshly fallen snow was collected and studied using a scanning electron microscope (SEM), in which the specimen temperature was controlled using an equipped cryo-system. The initial morphologies of snowflakes were revealed closed to liquid nitrogen temperature. By warming up the specimen, rapid sublimation was induced causing severe destruction to the fine structures, which was observed and recorded *in situ*.

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EXPERIMENTAL

A sample of snow was collected on December 9th, 2008 in Hanover, New Hampshire in a custom-made sample holder during a snowfall. The sample holder, which was made of copper due to its high thermal conductivity, had a trapezoid base to fit onto a SEM specimen stage and a shallow 10 mm x 25 mm cup on the top for holding the snow specimen. After sufficient snowflakes were obtained in the cup, the sample holder was slid into a copper rack, which was maintained at liquid nitrogen temperature (-196°C) in a portable cooler. A teflon cover was used to protect the snow sample from vapor deposition and any contaminants. The snow sample was sealed in the liquid-nitrogen environment until transfer into the SEM chamber, so that there was little, if any, metamorphism occurring before SEM observations. No conductive coating was applied to the snow sample. The advantages of examining uncoated snow have been discussed in earlier work (Iliescu and Baker, 2002; Baker *et al*., 2003; Dominé *et al*., 2003).

Observations of the snow aggregations were performed using a FEI XL30 field emission gun environmental SEM equipped with a Gatan cryo-system. The cryo-system was used to regulate the temperature of the specimen stage through a temperature controller. It also helped to maintain the vacuum in the SEM chamber by applying an airlock component, through which the snow sample was loaded onto the specimen stage in the SEM. A window on the airlock enabled inspection in order to get correct specimen placement. The teflon cover was removed from the sample holder, by rotating the holder vertically while in the airlock. This caused the cover to fall off into the airlock. Hence, the snow sample was exposed to the atmosphere for only less than a second.

To examine the morphology of freshly fallen snow, the sample stage was initially held at -180°C ± 2 °C and the SEM chamber was pre-evacuated to 1×10^{-2} Pa. Under these conditions, the snow suffered little sublimation enabling its initial morphology to be examined for more than two hours. Specimen charging is an issue at such low temperatures, because with little ice sublimation, electrons cannot be effectively removed from the sample surfaces. Therefore, an accelerating voltage of 2 kV was used in order to minimize charging, by producing surface conduction.

Since the rate of snow specimen sublimation is highly affected by the pressure and temperature in the SEM chamber (Waller *et al*., 2005), mass sublimation was initiated by warming up the specimen stage. The temperature was set through the temperature controller, which then automatically turned on the heating element embedded in the specimen stage.

RESULTS AND DISCUSSION

Figure 1 shows the initial morphologies of snowflakes consisted in the snow specimen observed at -180°C. The snow crystals encompass a variety of geometries and dimensions; however, the majority were flat hexagonal plates of 0.4-0.8 mm dia. and 0.04-0.1 mm thick. Narrow grooves were frequently observed on the edges of these plates. This characteristic has also been noted by earlier researchers as a double sheet structure (Nakaya, 1954; Rango *et al*., 1996).

To study the sublimation process, an individual snow crystal was focused on instead of the whole specimen. This snow crystal was first viewed at -180°C when it was found to be a hollow hexagonal column capped with a stellar plate on each end (Figure 2a). To initiate its sublimation, the specimen stage was then warmed up to -100°C over 25 mins. Over this period, the structural evolution was recorded by taking images intermittently, and recording the temperatures and times. The electron beam was only kept on when taking images in order to minimize the sample exposure to the electron beam.

During heating, no structural changes of the snow crystal was observed before the temperature reached -155°C. While it is generally found that solid ice sublimation begins at around -100°C for conventional high vacuum cryo-SEM (Waller *et al*., 2005; Stokes 2003), substantial sublimation of the fresh-snow specimen was observed starting at around -150°C. This decrease in the starting point of sublimation is probably caused by the complicated structures of the snow specimen compared to solid ice. Snow aggregations consist of lots of pore spaces and ice crystals with various geometries, which results in a very large interface between ice surfaces and the atmosphere (around 56 mm^2/mm^3 in this specimen). In addition, water molecules, especially the ones on the edges, are bonded more loosely to the ice matrix in a snow specimen than on flat ice surfaces duo to the complicated surface curvatures of individual snow crystals.

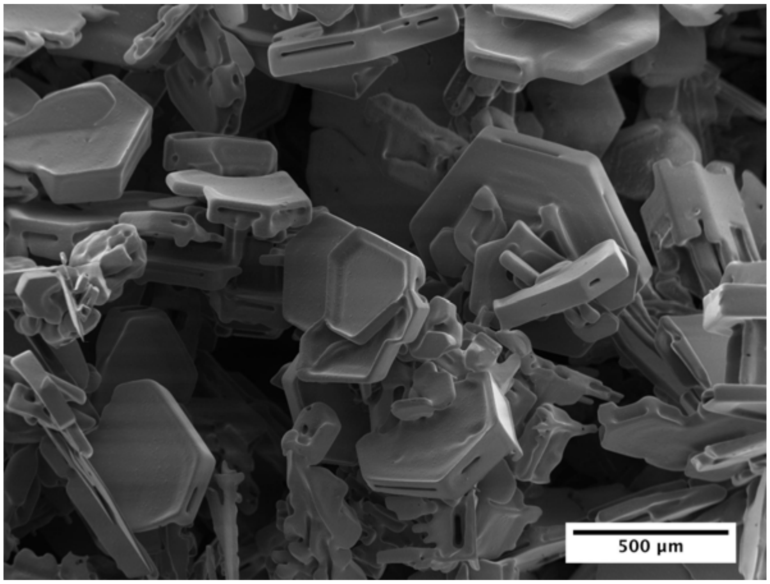
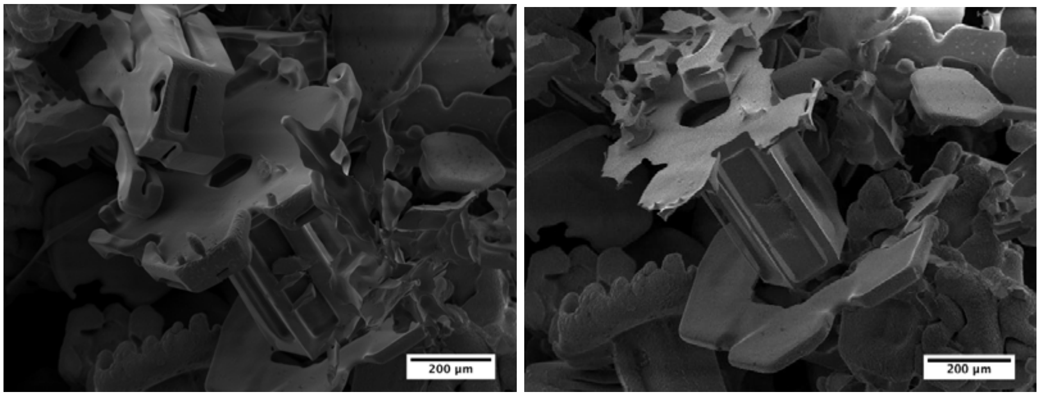


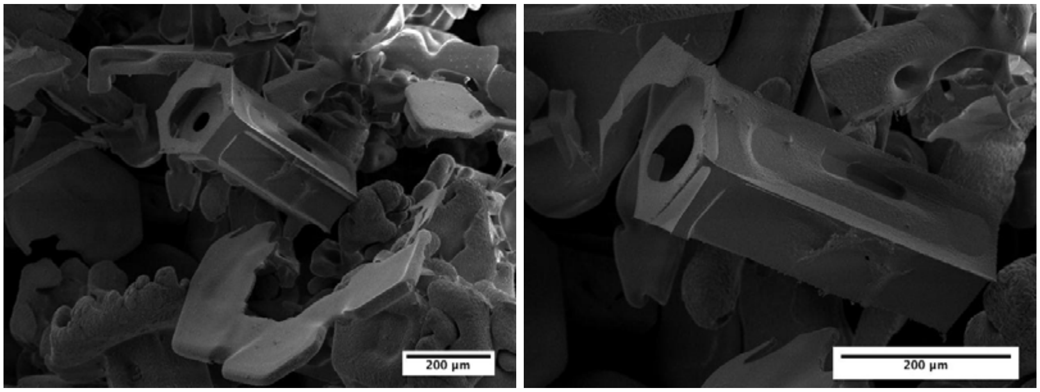
Figure 1. Secondary electron image of a fresh-snow specimen consisting of numerous snow crystals with various geometries, in which the majority are thin hexagonal plates with narrow grooves on the edges. The image was taken at -180°C. The accelerating voltage was 2 kV.

After sublimation initiated at around -150°C, mass loss occurred rapidly causing severe destruction to the snow crystal. Figure 2 are selected ones from the whole set of time series images recording the structural changes over 25 mins. Our observations indicate that compared with the center column, the capping plates were in a higher energy state and more readily sublimated away. While the stellar plate on the top was etched from the edges, the one on the bottom was broken off from the weakest connection to the column. Afterwards, the hollow column was peeled off from the outside stepped layers, accompanied with a slight increase in the dimension of the hole in the top of the column. Continuous surface roughing indicates that water molecules were also evaporated from the flat surfaces, although with a much lower rate. Slight orientation changes were also noticed, which was probably caused by the interaction between this snow crystal and adjacent ones, since they all experienced highly active morphological changes under relatively high temperatures. Another possible reason for particle movements is the specimen drift due to differential cooling of the cold stage (Cullen and Baker, 2002), which could cause crystal rearrangement.

Interestingly during this sublimation process under high-vacuum conditions, no obvious edge rounding was observed, a feature commonly observed from initial field sublimation (Colbeck, 1983; Rango *et al.*, 1996). A possible explanation for the absence of rounding edges is that under high vacuum, the huge difference between the real pressure (around 1 x 10^{-2} Pa) and the equilibrium ice vapor pressure drives not only the molecules on the edge but also those in close proximity to sublimate very rapidly, which overwhelms the difference between their activation energies for evaporation.



(a) T = -180°C, t = 0 min (b) T = -150°C, t = 12 min



(c) T = -130°C, t = 20 min (d) T = -100°C, t = 25 min

Figure 2. Secondary electron images showing the sublimation-induced structural changes of a snow crystal under high-vacuum conditions in the SEM chamber. The temperature was raised up from -180°C to -100°C over 25 mins. The accelerating voltage was 2 kV.

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