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Formation process of branches of needle ice crystals grown from vapor

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Abstract: *In-situ* observations of needle ice crystals growing in air at 10^5 Pa at -7°C and at about water saturation and above water saturation were performed to study the formation process of the branches of needle ice crystals. At about water saturation, six branches with boomerang-like facets are formed after the formation of a thin wall with basal facet along the edge of the basal plane by bunching of steps formed by two-dimensional nucleation at each corner on the basal plane. As time elapses, these branches change into sheath-like branches. Above water saturation, six branches with round tips grow along the *c*-axis because adhesive growth occurs at each corner on the basal plane. As the ice crystal grows further, these branches change into sheath-like branches through the same process as the branches forming at about water saturation.

1. Introduction

Relatively high supersaturation over ice sometimes occurs at North Pole Station when there is a small amount of aerosol (Konishi, 1999). It is well known that ice crystals grow as dendrites or needles in air at 10^5 Pa and above water saturation (Kobayashi, 1961). The schematic diagrams presented by Frank (1982) and Kobayashi (1984) suggest that ice crystals with solid columnar branches at each corner on the basal plane grow in air at 10^5 Pa at about -7°C and at high supersaturation. However, the primary branches of needle ice crystals observed at Mt. Daisetsu in Hokkaido had skeleton structure and round tips (Nakaya Ukichiro Museum of Snow and Ice, 1999).

Although there have been many studies on dendritic ice crystals (Gonda and Nakahara, 1996, 1997; Nakata *et al.*, 1992), we have few studies on the formation process of branches of needle ice crystals. The purpose of this study is to clarify the formation process of branches of needle ice crystals growing in air at 10^5 Pa and at -7°C by means of in situ observations.

2. Experiments

Figure 1 shows a schematic diagram of the growth chamber for ice crystals growing from vapor. The chamber was cooled to the desired temperature by circulating coolant

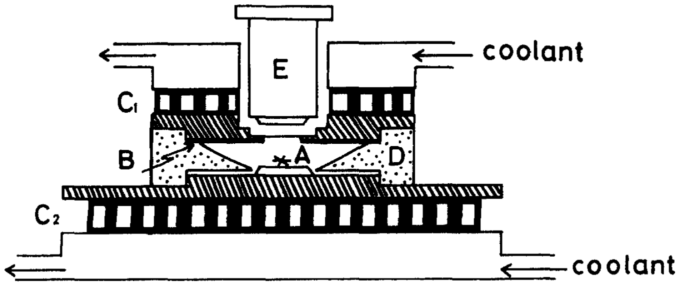

Fig. 1. Schematic diagram of a growth chamber.
A: glass substrate, B: ice plate, C_1, C_2: thermomodules, D: insulator, E: objective lens ($\times 10$).

cooled in advance in a refrigerator. The temperatures of the upper and lower copper plates of the chamber were regulated independently by adjusting electric currents to the thermoelectric modules. Ice crystals were nucleated by injecting sufficiently diluted silver iodide smoke into the chamber. The temperature of the ice plate attached to the upper plate was kept slightly higher than that of the glass substrate in order to supply water vapor to the growing ice crystal. The ice crystal was grown in air at 10^5 Pa at -7°C and at various supersaturations. A reflection type differential interference microscope with a $\times 10$ objective lens and a 35 mm film camera were used to observe the growth of ice crystals. Here, the temperature of the glass substrate and the temperature difference between the glass substrate and the ice plate were measured by using Cu-Co thermocouples. The supersaturation of each experiment was estimated from the measured temperatures.

3. Experimental results

Figure 2 shows an ice crystal growing in air at 10^5 Pa at -7°C and at about 6% supersaturation. The ice crystal had macroscopically flat surfaces in an early stage of growth (a). As the crystal grew, a giant step (arrow ↑) was formed along the edges (b) and advanced toward the center of the basal plane (c). Thin steps were observed

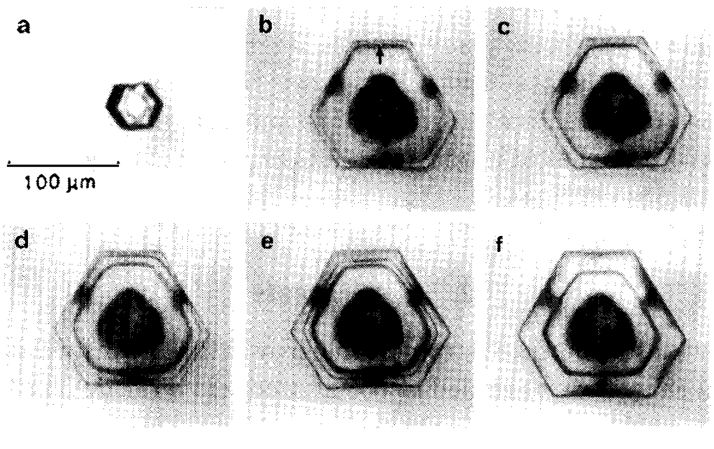

Fig. 2. Ice crystal growing in air at 10^5 Pa at -7°C and at about 6% supersaturation.
(a) 0, (b) 3.0, (c) 3.3, (d) 3.8, (e) 4.0 and (f) 5.0 min.

spreading from each corner on the basal plane but they were too thin and fast to take photographs. These steps advanced preferentially along each edge of the basal plane and contacted each other. As the resulting hexagon-like giant step advanced toward the center of the basal plane (d, e), step velocity gradually decreased. Thus the preceding step was caught by the following step (f). As a result, a hexagon-like terrace was formed by bunching of the giant steps.

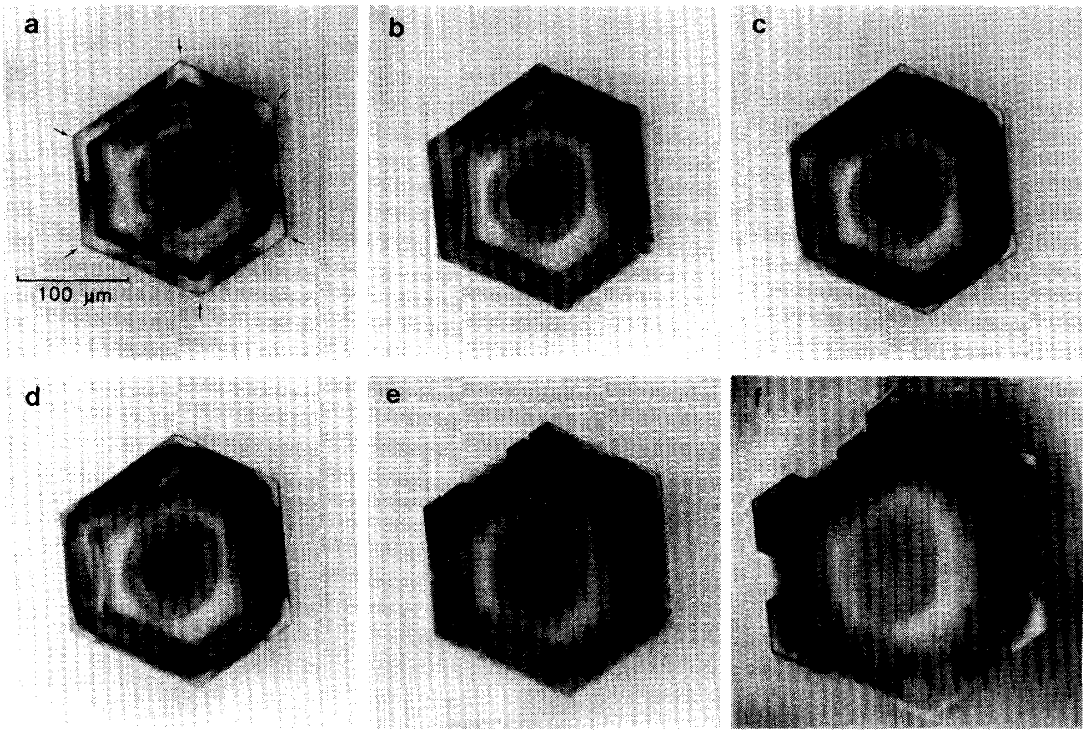

Fig. 3. Further growth of the same ice crystal as shown in Fig. 2. (a) 0, (b) 0.5, (c) 1.0, (d) 2.0, (e) 7.0 and (f) 12 min.

Figure 3 shows the further growth of the same ice crystal as shown in Fig. 2. Bunching of steps occurred near the edge of the basal plane and a thin wall with basal facet was formed along the edge of the basal plane (a). Then the six branches with boomerang-like facets grew along the *c*-axis at each corner on the thin wall (b–d). Figure 4 shows a crystal that was observed near the crystal shown in Fig. 3c. As time elapsed, the branches shown in Fig. 3d changed into sheath like branches (3e). Then, the tips of the sheath-like branches tended to be capped by a basal plane (3f) as seen in ZnO hollow crystals (Iwanaga and Shibata, 1981).

Figure 5 shows a side view of an ice crystal growing in air at 10^5 Pa at -7°C and at about 10% supersaturation. A prismatic plane of the ice crystal is parallel to the glass substrate. The branches with round tips grew along the c-axis at each corner on the basal plane (a). As these branches grew further, prismatic facets were formed at the sides of the branches (b) and then the round tips changed into boomerang-like facets (c, d). As time elapsed, the branches changed into sheath-like branches (e). Thereafter, the supersaturation was increased up to about 13%. Then secondary branches with round tips grew along the c-axis at each corner on the primary branches (f).

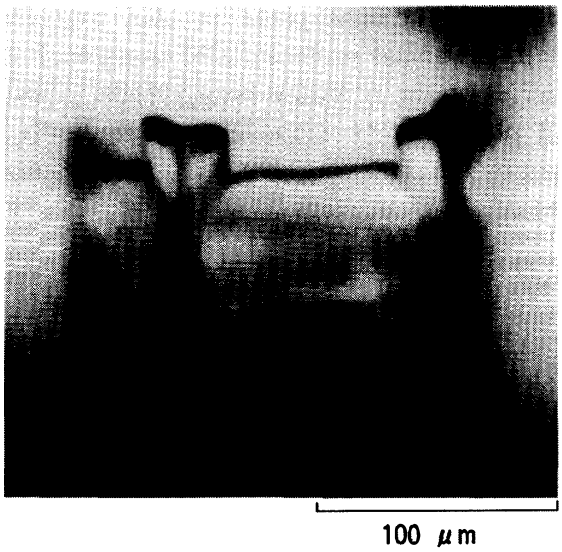

Fig. 4. Ice crystal observed near the ice crystal shown in Fig. 3c.

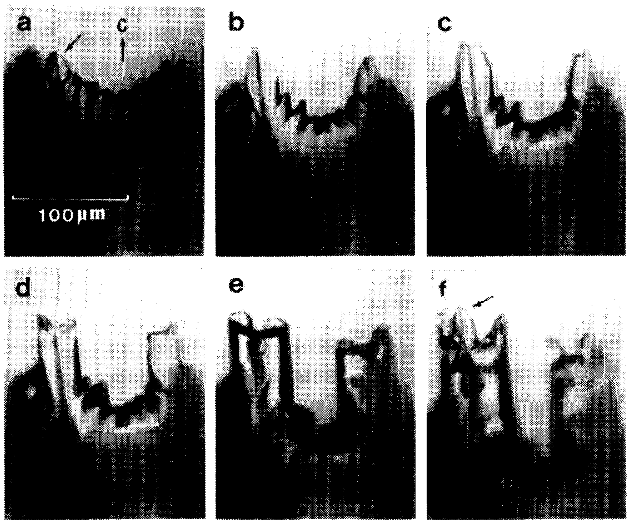

Fig. 5. Ice crystal growing in air at 10^5 Pa and at -7°C and at about 10% (a–e) and about 13% (f) supersaturation. (a) 0, (b) 0.3, (c) 0.5, (d) 1.0, (e) 3.0 and (f) 4.0 min.

Figure 6 shows the further growth of the secondary branch with round tip, indicated by an arrow in Fig. 5f. After preferential growth along the c-axis, the round tip changed into a faceted tip. Then the secondary branch changed into the sheath-like one through the same process as the primary branch. The sheath-like branches are open at the nearest side to the center of the basal plane (Fig. 7e).

4. Discussion

In the present experiments, actual surface supersaturation of an ice crystal depends on the size and shape of the crystal. Usually, the increase of crystal size and the growth

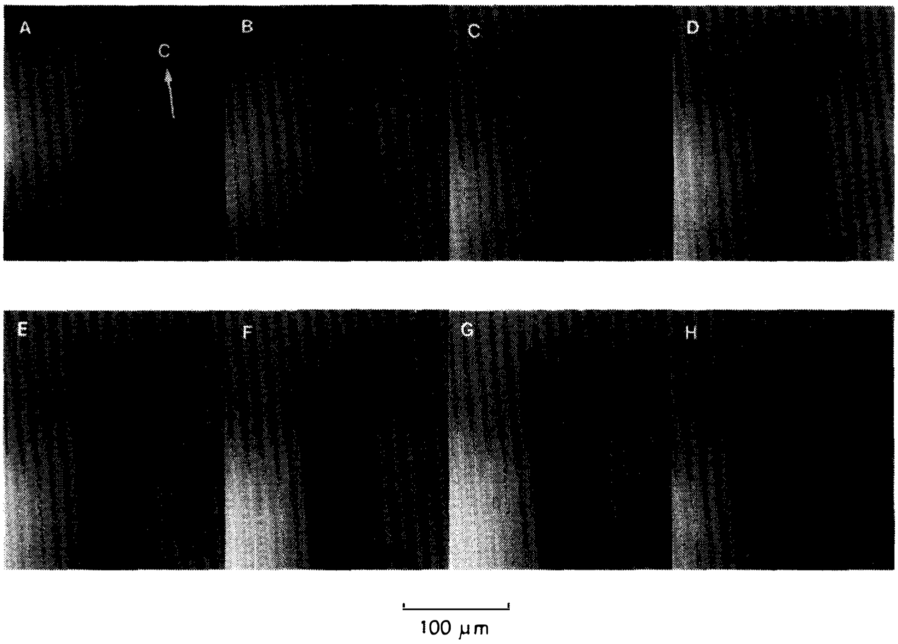

Fig. 6. Further growth of the secondary branch indicated by an arrow in Fig. 5f. (A) 0, (B) 0.5, (C) 1.0, (D) 1.5, (E) 2.0, (F) 2.5, (G) 3.0 and (H) 3.5 min.

of neighboring faces reduce the surface supersaturation.

It is very difficult that screw dislocations which are parallel to the c-axis outcrop at all corners of the basal plane. Accordingly, the formation mechanism of steps observed at the corners on the basal plane in Figs. 2–3 is a two-dimensional nucleation mechanism.

The tips of branches in Figs. 5a, b and f have round shapes. This means that the tips are formed by an adhesive growth mechanism because such round surfaces are rough on the molecular scale.

Based on Figs. 2–4, Fig. 7 shows schematically the formation process of the branches at $-7°C$ and at about water saturation. The thin steps due to two-dimensional nucleation at each corner on the basal plane advance preferentially along the edge of the basal plane and contact each other. As the resulting hexagon-like steps advance toward the center of the basal plane, step velocity gradually decreases and then

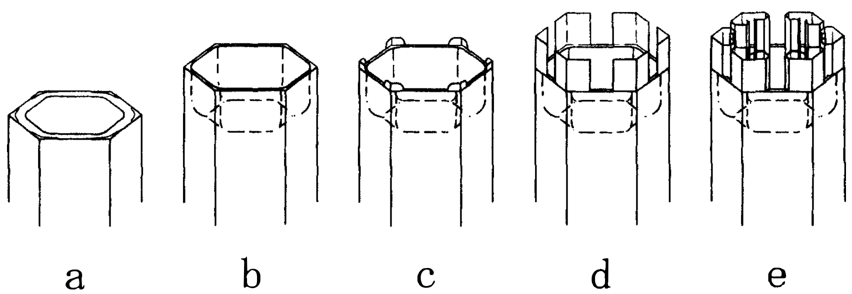

Fig. 7. Growth process of branches of a needle ice crystal growing in air at 10^5 Pa at $-7°C$ and at about water saturation.

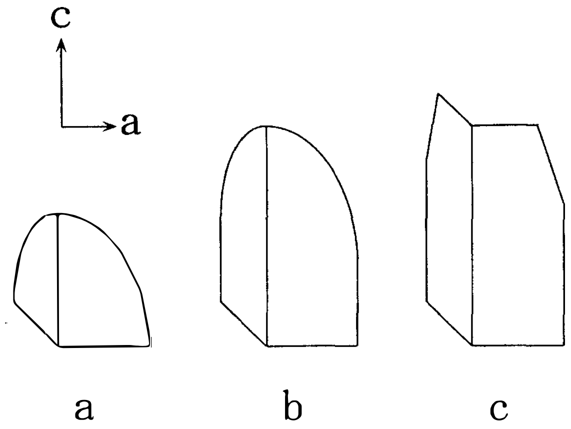

Fig. 8. Growth process of a branch of a needle ice crystal growing in air at 10^5 Pa at $-7°C$ and above water saturation.

the steps bunch up each other (a). Then the bunching of steps occurs near the edge of the basal plane and a thin wall with basal facet is formed along the edge of the basal plane (b). Next, the thin steps spreading from neighboring corners on the thin wall begin to bunch before they contact each other. Then six branches with boomerang-like facets grow along the c-axis at each corner on the thin wall (c). As these branches approach each other, their growth rates decrease (d). Then the branches change into the sheath-like ones (e).

Based on Figs. 5 and 6, Fig. 8 shows schematically the formation process of a branch of the needle ice crystal growing at $-7°C$ and above water saturation. As adhesive growth occurs at the corner on the basal plane, a primary branch with round tip grows along the *c*-axis (a). As the branch grows further, prismatic facets are formed at the sides of the branch (b), and then the tip changes into a boomerang-like facet (c). Then the branch changes into a sheath-like branch through the same process as shown in Figs. 7d and e.

5. Conclusions

In order to study the formation process of branches of needle ice crystals grown from vapor, in-situ observations of ice crystals growing in air at 10^5 Pa at $-7°C$ and at about water saturation and above water saturation were carried out. The results are as follows.

(1) At about water saturation, steps formed by two-dimensional nucleation at each corner on the basal plane form a hexagon-like giant step by bunching of steps at the middle of the basal plane. Then bunching of steps occurs near the edge of the basal plane and forms a thin wall with basal facet along the edge of the basal plane. Then branches with boomerang-like facets grow along the *c*-axis at each corner on the thin wall. As time elapses, these branches change into sheath-like branches.

(2) Above water saturation, as adhesive growth occurs at each corner on the basal plane, branches with round tips grow along the *c*-axis. The round tips of the branches change into boomerang-like facets with increasing crystal size. As the ice

crystal grows further, these branches change into sheath-like branches through the same process as branches forming at about water saturation.

Acknowledgments

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