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Experimental Studies of Dendritic Ice Crystals Growing from the Vapor Phase

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Abstract. Dendritic ice crystals were grown in air at 1.0×10^5 Pa at –15°C. In this paper, the formation mechanism of primary branches, the tip shape of primary branches, the formation mechanism of side branches and the spacing of side branches are discussed.

1. Introduction

The morphology of snow crystals, ice crystals growing from the vapor phase has been studied since earlier times (Nakaya, 1951; Mason, 1953; Kobayashi, 1961), but there are many unsolved problems even now.

Recently, the morphological instability of vapor-grown ice crystals or the pattern formation of dendritic ice crystals as the supersaturation increases has been studied (Gonda *et al.*, 1990; Yokoyama *et al.*, 1990). On the other hand, the qualitative studies on dendritic ice crystals growing from melt have been done (Arakawa *et al.*, 1952; Tirmizi, 1989).

As the tips of the primary branches of dendritic ice crystals growing from melt and solution (Gonda *et al.*, 1991) is rough in a molecular scale, every water molecule reached on ice crystal surfaces is crystallized.

Accordingly, an anisotropy of the surface tension is important for the formation of dendritic ice crystals growing from melt and solution. On the other hand, as the tips of dendritic ice crystals growing from vapor is smooth in a molecular scale, all water molecules reached on ice crystal surfaces are not crystallized. Accordingly, an anisotropy of the kinetic coefficient is important for the formation of dendritic ice crystals growing from vapor. In this paper, the formation mechanism of primary branches, the tip shape of primary branches, the formation mechanism of side branches and the spacing of side branches are discussed.

2. Experimental Apparatus

Figure 1 shows the experimental apparatus for the growth and in situ observation of dendritic ice crystals. The apparatus is composed of a growth chamber (a) and its temperature regulators (b, c, d), a vacuum system (f, i) and an optical system

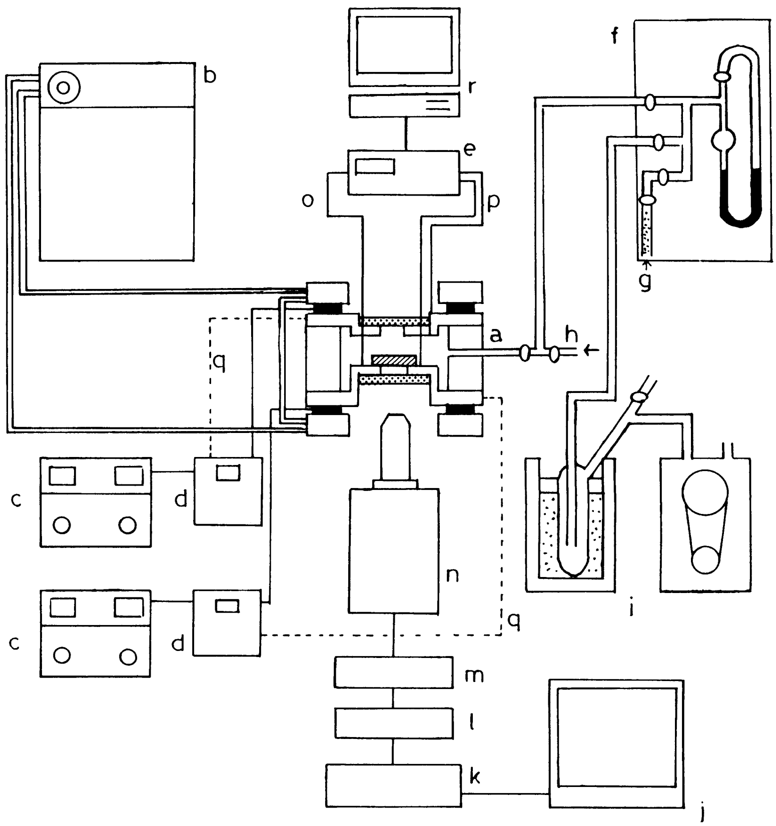

Fig. 1. Experimental apparatus for the formation and in situ observation of dendritic ice crystals. (a) growth chamber, (b) thermostatic refrigerated bath, (c) DC power supply, (d) temperature controller, (e) digital thermometer, (f) mercury manometer, (g) air inlet, (h) silver iodide smoke, (i) cold trap and vacuum pump, (j) video monitor, (k) video recorder, (l) video timer, (m) camera control unit, (n) video camera and microscope, (o, p, q) thermoelectric couples, (r) microcomputer.

(j, k, l, m, n). An ice sheet for water vapor supply and a growth substrate (sapphire) in the chamber were cooled down to about –15°C using thermoelectric modules. After the air in the chamber had been evacuated to about 10 Pa, water vapor was supplied by keeping the ice sheet at a slightly higher temperature than that of the growth substrate. After that, a small amount of diluted silver iodide smoke was inserted into the chamber together with clean air at 1.0×10^5 Pa. A minute ice crystal nucleated in air fell onto the growth substrate and was grown in air at 1.0×10^5 Pa at about –15°C and a controlled supersaturation. Dendritic ice crystals were observed using a transmitted microscope (n) and recorded by a video camera.

3. Experimental Results

3.1 Tip shape of primary branches

Figure 2 shows a time sequence of the growth form of a dendritic ice crystal with primary branches grown in air at 1.0×10^5 Pa at –15°C and about 12% supersaturation. In photograph (a), a hexagonal plate with the facets on the prismatic faces grows. The concaves arise on the prismatic faces with time elapsed (b), that is, the morphological instability takes place in this stage. Then, the concaves develop with time elapsed (c) and the facets on the prismatic faces disappear (d).

That is to say, the primary branches are formed as a result of the morphological instability of the $\{10\bar{1}0\}$ faces of an ice crystal (e, f). The off-faceted tips are formed immediately after primary branches were sprouted (e, f). Then, the facets are formed on the tips of primary branches with time elapsed (g, h).

Figure 3 shows the formation process of a primary branch of a hexagonal plate

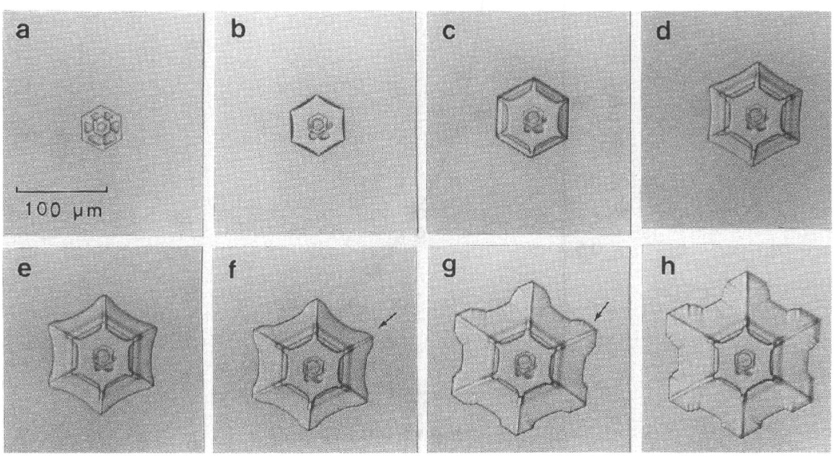

Fig. 2. Time sequence of a dendritic ice crystal grown in air at 1.0×10^5 Pa at –15°C and about 12% supersaturation. (b) 0, (c) 0.3, (d) 0.6, (e) 0.8, (f) 1.0, (g) 1.4, and (h) 1.6 min.

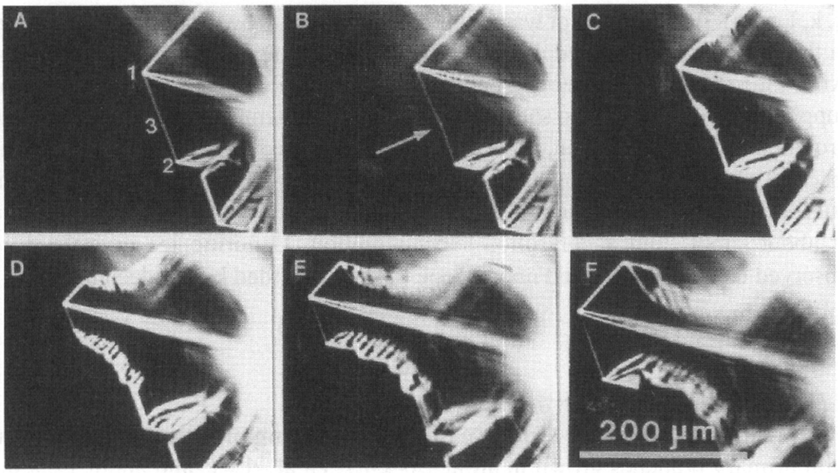

Fig. 3. Formation process of a primary branch of a hexagonal plate grown in air at 1.0×10^5 Pa at −15°C and near water saturation. (A) 0, (B) 0.6, (C) 1.3, (D) 2.8, (E) 4.5, and (F) 6.1 min.

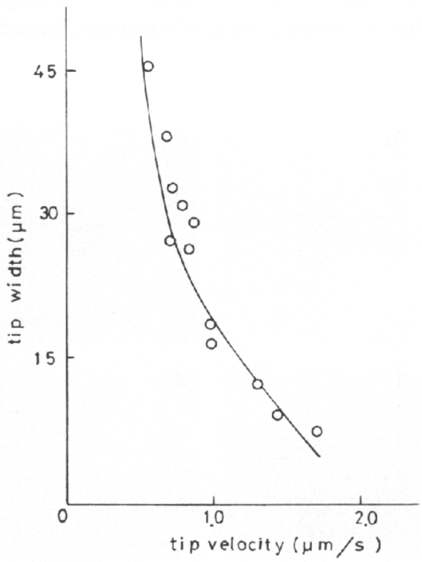

Fig. 4. Relationship between the tip width and the tip velocity of the primary branches of dendritic ice crystals grown in air at 1.0×10^5 Pa and −15°C.

grown in air at 1.0×10^5 Pa at −15°C and near water saturation. It is inferred that steps originated from two-dimensional nuclei nucleated at corners (1, 2) of a prismatic face successively advance to the center (3) of the face. The central part (arrow ↑) of the prismatic face began to be unstable when advancing steps bunched with time elapsed (photo B). A primary branch was formed as a result of morphological instability at two prismatic faces on both sides of the corner 1 (D, E, F).

Figure 4 shows the relationship between the tip width and the tip velocity of the primary branches of dendritic ice crystals grown in air at 1.0×10^5 Pa and −15°C. As shown in the figure, the tip width of primary branches decreased with increasing tip velocity as the supersaturation increases. That is to say, the primary branches with broad tip width grow at relatively low supersaturation, while the primary branches with narrow tip width grow at higher supersaturation.

3.2 Spacing of side branches

Figure 5 shows a time sequence of the growth form of a dendritic ice crystal grown in air at 1.0×10^5 Pa at −15.2°C and 15% supersaturation. In photograph (a), a hexagonal plate with facets on prismatic faces grows. The primary branches are formed with time elapsed as a result of the morphological instability of the $\{10\bar{1}0\}$ faces of the ice crystal (b). The off-faceted tips are formed immediately after the primary branches were sprouted. The facets appear at the tips of primary branches even as the supersaturation is constant (c, d). As the morphological instability (arrow ↑ in photo e) appears on the faceted tips of primary branches, the embryos of side branches are formed (arrow ↑ in g). That is to say, the side branches are formed as a result of the morphological instability of the faceted tips of primary branches.

Figure 6 shows a time sequence of the growth form of a dendritic ice crystal grown in air at 1.0×10^5 Pa at −15.5°C. Photographs (a)–(e) are of an ice crystal grown at water saturation. In (a), the morphological instability has already been generated on prismatic faces (arrow ↑). Six primary branches were formed as a result of morphological instability of six prismatic faces (c). Although the tips of primary branches are initially off-faceted (c, d), facets were formed at their tips after time had elapsed (e). Photographs (f)–(h) are of an ice crystal at about 20% supersaturation over water. The side branches were formed at each corner of primary branches when the supersaturation increased (f), and the tips of primary branches became off-faceted again (f, g). Thereafter, facets were formed again at the tips of primary branches with time elapsed (h). Photograph (i) is of an ice crystal at about 40% supersaturation over water. Secondary side branches were formed again at each corner of primary branches when the supersaturation further increased (i). As shown in Fig. 6, it is important for the formation of side branches of an ice crystal growing from vapor that the facets are formed at the tips of primary branches and then, the supersaturation increases. Here, the spacing of side branches is given in Fig. 6(i).

Figure 7 shows the relationship between the spacing of side branches and the

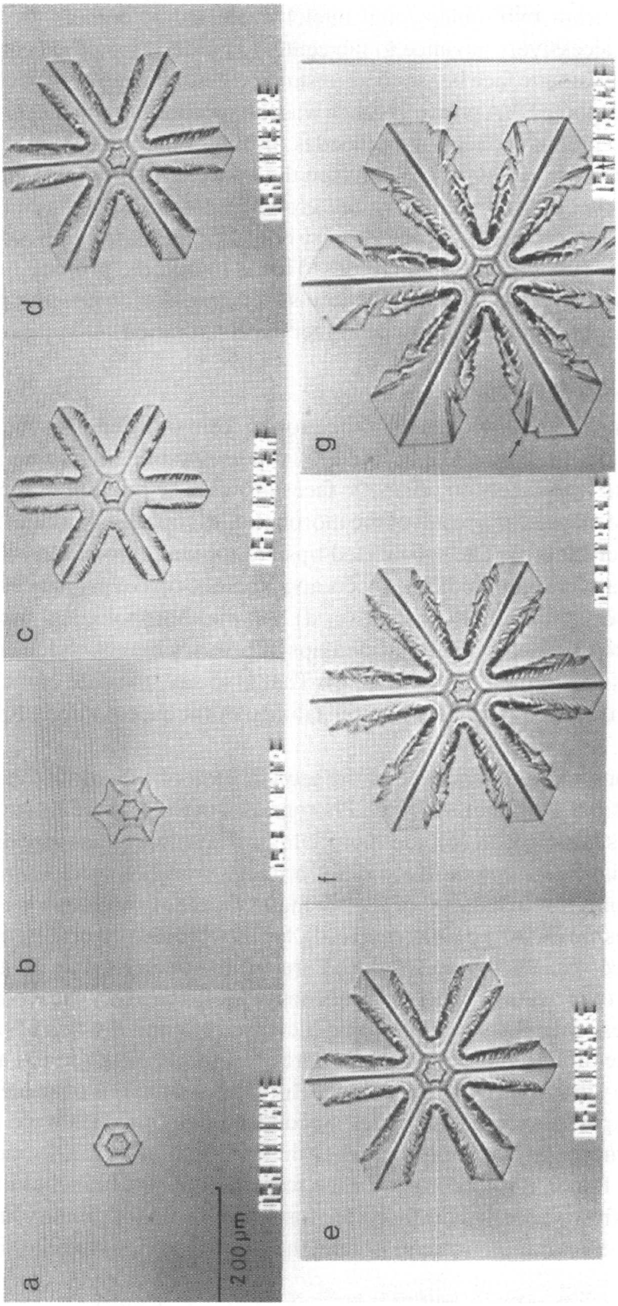

Fig. 5. Time sequence of a dendritic ice crystal grown in air at 1.0×10^5 Pa at −15.2°C and 15% supersaturaion. (a) 0, (b) 0.3, (c) 2.2, (d) 2.5, (e) 2.7, (f) 3.6, and (g) 5.1 min.

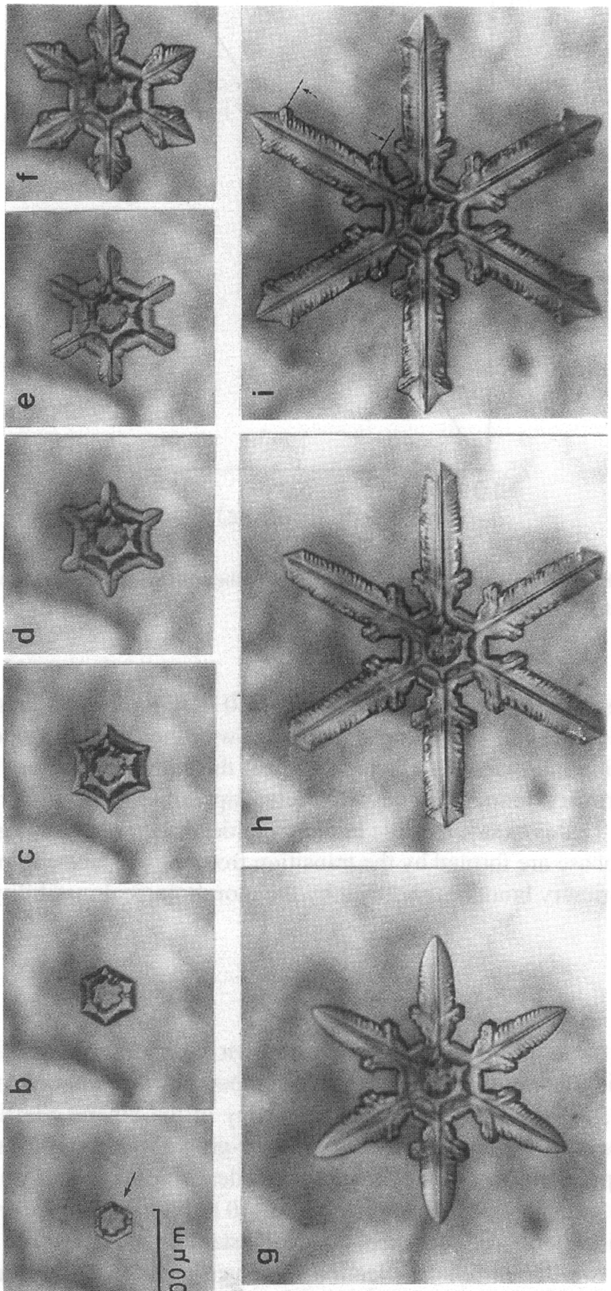

Fig. 6. Time sequence of a dendritic ice crystal grown in air at 1.0×10^5 Pa at −15.5°C. (a) 0, (b) 0.2, (c) 0.3, (d) 0.4, (e) 0.5, (f) 0.7, (g) 1.0, (h) 1.4, and (i) 1.7 min.

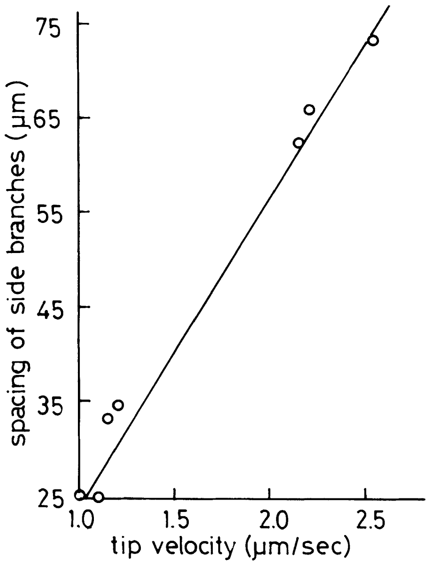

Fig. 7. Relationship between the spacing of the side branches and the tip velocity of dendritic ice crystals grown in air at 1.0×10^5 Pa at -15°C.

tip velocity of dendritic ice crystals grown in air at 1.0×10^5 Pa at -15°C. As shown in the figure, the spacing of side branches increases with increasing tip velocity as the supersaturation increases. The fact means that the side branches are hard to sprout as the primary branches with off-faceted tips are formed under higher supersaturation. That is to say, the side branches of dendritic ice crystals growing from the vapor phase are formed by the transition from the off-faceted tips to the faceted tips of primary branches and then by the morphological instability of the faceted tips.

4. Discussion

In general, the surface structure of vapor-grown ice crystals is smooth as the supersaturaiton is relatively low, except that the temperature is just below a melting point (Sei *et al.*, 1989). Accordingly, an anisotropy of the kinetic coefficient is important for the rate determining process of vapor-grown ice crystals. As shown in the present experiment, the primary branches of a dendritic ice crystal are formed as a result of the morphological instability of the $\{10\bar{1}0\}$ faces of an ice crystal as the supersaturation increases. The morphological instability of the $\{10\bar{1}0\}$ faces is produced by the two-dimensional nucleation at each corner of these faces and then by the bunching of the steps which advance from each corner to a center of these faces. Although off-faceted tips are formed immediately after the primary branches were sprouted, the facets appear on the off-faceted tips with further growth is important, too.

This fact means that not only the volume diffusion process of water molecules towards ice crystal surfaces is important for the growth of dendritic ice crystals growing from the vapor phase, but also the surface kinetic process of water molecules on ice crystal surface.

In the next place, the side branches of dendritic ice crystals are formed as a result of the transition from the off-faceted tips to the faceted tips of primary branches, and then the morphological instability of faceted tips as the supersaturation increases.

It is important for the successive formation of side branches, that the transition from the off-faceted tips to the faceted tips of primary branches arises in sequence with further growth.

The side branches are not formed as the off-faceted tips are maintained during the growth under higher supersaturation. That is to say, in the case of the vapor growth, side branches are not formed as the growth is controlled by only the volume diffusion process of water molecules. In other words, the surface kinetic process of water molecules on ice crystal surfaces is important for the formation of the side branches of dendritic ice crystals growing from the vapor phase.

5. Conclusions

Dendritic ice crystals were grown in air of 1.0×10^5 Pa at -15°C. Obtained results in the present experiment are as follows.

(1) The primary branches of a dendritic ice crystal are formed as a result of the morphological instability of the $\{10\bar{1}0\}$ faces of a hexagonal plate with increasing supersaturation. The tip width of primary branches decreases with increasing tip velocity as the supersaturation increases.

(2) The side branches of a dendritic ice crystal are sprouted as a result of the transition from the off-faceted tips to the faceted tips of primary branches and then the morphological instability of the faceted tips. The side branches are hard to sprout as the supersaturation increases under higher supersaturation.

(3) It is concluded from the present experiment, that the volume diffusion process of water molecules and the surface kinetic process of water molecules are both important for the formation of the dendritic ice crystals growing from the vapor phase.

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