MORPHOLOGICAL STABILITY OF POLYHEDRAL ICE CRYSTALS GROWING FROM THE VAPOR PHASE

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The morphological stability of polyhedral ice crystals growing in air at 1.0 atm and −15°C has been studied. The stability of the (1010) faces of the ice crystals depends on both bulk supersaturation and crystal size. The stability region of the ice crystals above 100 μm in size coincides qualitatively with the result of the numerical calculations by Kuroda, Irisawa and Ookawa.

1. Introduction

The results of experimental studies on the growth forms of ice crystals from the vapor made by Nakaya [1], Hallett and Mason [2], and Kobayashi [3,4] have been combined by Kobayashi [4] in a supersaturation versus temperature diagram. The effects of diffusion of vapor and heat on the growth forms of ice crystals grown from the vapor have been studied and established by Isono, Komabayasi and Ono [5], Komabayasi [6], and Gonda et al. [7—10]. However, the growth mechanisms of ice crystals growing from the vapor have still not been solved. Recently, Kuroda and Lachmann [11] theoretically studied the variation of the habit of ice crystals with temperature, and claimed in the study that the habit of ice crystals growing below −20°C depended not only on temperature but also on bulk supersaturation. Gonda and Koike [12] experimentally studied the habit of ice crystals growing in air at 1.0 atm below −20°C as functions of temperature and bulk supersaturation, and found that the habit of small polyhedral ice crystals of 15 μm in size depended on both temperature and bulk supersaturation, but the habit of large ice crystals above 100 μm in size depended only on temperature.

On the other hand, Chernov [13] and Kuroda, Irisawa and Ookawa [14] theoretically studied the morphological stability of a crystal growing from solution. Gonda and Koike [12] also experimentally studied the morphological stability of ice crystals growing in air at 1.0 atm at a temperature of −30°C. At a temperature near −15°C, the growth rate of ice crystals is the largest under water saturation, and under these growth conditions dendritic ice crystals grow; so it is important to make a study of the morphological stability of ice crystals at a temperature of −15°C.

2. Experimental procedures

Fig.1 shows a growth chamber for in situ observation of ice crystals growing in air at 1.0 atm. In order to cool independently an ice sheet for the supply of water vapor (B) and a growth substrate (A), a thermal insulator (D) is inserted between the thermoelectric cooling panels set at the top (C1) and at the bottom (C2) of the chamber.

For the sake of growing an ice crystal on the substrate at a constant temperature and at various constant supersaturations, the temperature T1 of the ice sheet (B), the temperature T2 of the substrate (A) and the temperature difference T1 − T2 between them must be held constant. To keep T2 at −15°C, the coolant cooled down to about
$-15^\circ C$ is circulated to the lower part of the cooling panel $C_2$. On the other hand, to keep $T_1$ just above $-15^\circ C$, the coolant cooled down to about $-14^\circ C$ is circulated to the upper part of the cooling panel $C_1$. An electric current which is flowed to the cooling panels is automatically controlled by the use of temperature regulators (not shown). As a result, the temperatures of the ice sheet (B) and the substrate (A) are kept at the specified temperature, respectively. Therefore, we can grow an ice crystal at $-15^\circ C$ and for various constant supersaturations.

An inspection of ice and water saturation at $-15^\circ C$ was made by measuring the temperature at which ice crystals began to deposit on the substrate and by measuring the temperature at which supercooled water droplets began to deposit on the substrate.

Minute polyhedral ice crystals were formed in free fall in air at 1.0 atm and $-15^\circ C$ using another cylindrical cold chamber [15]. An ice crystal fell on a slide glass (A) set in the growth chamber. Thereafter, the ice crystal was grown in air at 1.0 atm and $-15^\circ C$ using the growth chamber shown in fig. 1, and we observed the growing ice crystal in situ using a differential interference microscope. For the measurements of the growth rate of ice crystals, only ice crystals whose prismatic face is parallel to the substrate were chosen.

### 3. Experimental results

Fig. 2 shows an ice crystal, grown in air at 1.0 atm and $-15^\circ C$ and at a supersaturation of 12%, whose prismatic face is parallel to the slide glass.

As shown in the figure, a polyhedral crystal grows just after the crystal had fallen on the slide glass (a). After 1.3 min (b), the crystal edges which are composed of basal and prismatic faces begin to spread. After 40 min (c), the polyhedral crystal is transformed to the crystal with the plates on upper and lower bases (double plate). After 10.8 to 16.5 min (d, e), the distance between upper and lower plates of the double plate decreases. After 64 min (f), the double plate reverts to a polyhedral crystal. Thus, the growth form of an ice crystal changes with increasing crystal size.

Fig. 3 shows the stability limit of the $\{10\overline{1}0\}$ faces of plate-like ice crystals grown in air at 1.0 atm and $-15^\circ C$. In the figure, solid circles show
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The stability limit of the \{10\overline{1}0\} faces of original ice crystals prior to transformation to double plates; open circles show the stability limit of the \{10\overline{1}0\} faces of the plates on upper and lower bases. As shown in the figure, at a supersaturation below about 5%, polyhedral ice crystals grow in the whole region of crystal size. At a supersaturation between 5 and 14%, polyhedral ice crystals grow when the crystal size is below about 20 \mu m; however, at sizes between 20 and 500 \mu m, skeletal ice crystals grow, the \{10\overline{1}0\} faces of the original ice crystals becoming unstable. When the crystal size increases above about 500 \mu m, the skeletal ice crystals revert to polyhedral crystals, the \{10\overline{1}0\} faces of the original ice crystals becoming stable again. Next, at a supersaturation above about 14%, polyhedral ice crystals grow when the crystal size is below about 20 \mu m; however, at sizes between 20 and 50 \mu m, the polyhedral ice crystals are transformed to double plates, the edges of the original ice crystals spreading rapidly. At sizes between 50 and 500 \mu m, double plates are transformed to dendritic crystals, the crystals with dendritic plates on upper and lower bases. When the crystal size increases above about 500 \mu m, the dendritic crystals revert to double plates. At sizes above 1000 \mu m, double plates revert to polyhedral crystals. Thus, it is apparent that the morphological stability of plate-like ice crystals grown from the vapor depends not only on bulk supersaturation but also on crystal size.

Fig. 4 shows the size dependence of the growth rates in the \{11\overline{2}0\} direction at the corners of ice crystals grown in air at 1.0 atm and \(-15^\circ\text{C}\) and at various constant supersaturations. As shown in figs. 3 and 4, when the crystal size is below a few hundred \mu m, the growth rates at the corners of ice crystals growing at a supersaturation above 14% increase with increasing crystal size; in this size region, double plates are transformed to dendritic crystals with increasing crystal size. However, when the crystal size increases above a few hundred \mu m, the growth rates at the corners of the crystals decrease with increasing crystal size; in this size region, the dendritic crystals revert to double plates with increasing crystal size. The growth rates at the corners of ice crystals growing at a supersaturation between 5 and 14% increase with increasing crystal size when the crystal size is below
about 20 μm; in this size region, polyhedral crystals are transformed to skeletal crystals with increasing crystal size. However, when the crystal size increases above about 20 μm, the growth rates at the corners of the crystals decrease with increasing crystal size; in this size region, the skeletal crystals revert to polyhedral crystals with increasing crystal size.

Fig. 5 shows the supersaturation dependence of the growth rates in the <1120> and in the <0001> directions at the corners of ice crystals of 40 μm growing in air at 1.0 atm and -15°C. The supersaturation σ is smaller than σ* (about 5%), the growth rates in the <1120> and in the <0001> directions at the corners of ice crystals slightly increase with increasing supersaturation; in this supersaturation region, polyhedral ice crystals grow. For σ** (about 14%) > σ > σ*, however, the growth rate in the <1120> direction gradually increases with increasing supersaturation, while that in the <0001> direction rapidly increases with increasing supersaturation; in this supersaturation region, a skeletal structure develops on the {1010} faces of the crystals. For σ > σ**, the growth rate in the <0001> direction still increases gradually with increasing supersaturation, while the growth rate in the <1120> direction increases still more rapidly with increasing supersaturation; in this supersaturation region, dendritic crystals grow.

4. Discussions

It is found from the present study that the stability of the {1010} faces of ice crystals grown in air at 1.0 atm and -15°C depends on both bulk supersaturation and crystal size. The stability limit of the {1010} faces of ice crystals growing at -15°C prior to transformation to double plates shows the same tendency as that at -30°C [12].

As shown in fig. 3, the stability limit versus supersaturation and crystal size for ice crystals above 100 μm in size coincides qualitatively with the result of numerical calculations by Kuroda, Irisawa and Ookawa [14].

As clearly shown in fig. 4, when the growth rates in the <1120> direction at the corners of ice crystals increase with increasing crystal size, the growth form of the ice crystals is transformed from stable to unstable. On the other hand, when the growth rates in the <1120> direction at the corners of ice crystals decrease with increasing crystal size, the growth form of the ice crystals reverts from unstable to stable.

Next, as clearly shown in fig. 5, the growth rate in the <1120> direction at the corners of ice crystals is larger than that in the <0001> direction when the supersaturation is high. This fact means that when a plate-like ice crystal grows in air at 1.0 atm, water vapor flux concentrates along the <1120> direction of the crystal rather than along the <0001> direction because of the effect of the crystal shape on the volume diffusion of water vapor.

5. Conclusion

The morphological stability of plate-like ice crystals grown in air at 1.0 atm and -15°C has been studied. The obtained results are as follows:

1) The stability limit of the {1010} faces of the ice crystals grown at -15°C depends on both...
bulk supersaturation and crystal size. The stability limit of the \{10\bar{1}0\} faces of the ice crystals grown at $-15^\circ C$ prior to transformation to double plates shows the same tendency as that at $-30^\circ C$.

(2) The stability limit versus supersaturation and crystal size for ice crystals above 100 $\mu$m in size coincides qualitatively with the result of numerical calculations of a crystal growing from solution by Kuroda, Irisawa and Ookawa [14].

(3) When the growth rates in the \langle 11\bar{2}0 \rangle direction at the corners of ice crystals increase with increasing crystal size, the growth form of the ice crystals is transformed from stable to unstable. On the other hand, when the growth rates in the \langle 11\bar{2}0 \rangle direction at the corners of ice crystals decrease with increasing crystal size, the growth form of the ice crystals reverts from unstable to stable.

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