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THE GROWTH MECHANISM OF ICE CRYSTALS GROWN IN AIR AT A LOW PRESSURE AND THEIR HABIT CHANGE WITH TEMPERATURE

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Résumé - La dépendance avec la température de la morphologie d'un cristal de glace formé dans l'air à 4.0×10 Pa avec une sursaturation inférieure à 5 % et le mécanisme de cette croissance ont été étudiés en faisant des cycles croissance-évaporation du cristal de glace. La modification de morphologie du cristal de glace avec la température dans les conditions précédentes coïncide grossièrement avec celle des cristaux de glace formés dans l'air à 1.0×10^5 Pa et on a trouvé que les faces $\{0001\}$ et $\{10\bar{1}0\}$ du cristal de glace formé avec une supersaturation inférieure à 5% croissent par un mécanisme de dislocations vis. La dépendance avec la température de la morphologie du cristal de glace formé dans ces conditions peut être qualitativement expliquée par l'anisotropie du coefficient de condensation et de sa dépendance avec la température.

Abstract

The temperature dependence of the habit of an ice crystal grown in air at 4.0×10 Pa under a supersaturation below 5% and its growth mechanism were investigated by repeating the growth and evaporation of the ice crystal. As a result, the habit change of an ice crystal with temperature under this growth condition roughly coincides with that of an ice crystal growing in air at 1.0×10^5 Pa, and it was found that the $\{0001\}$ and $\{10\bar{1}0\}$ faces of the ice crystal grown at a supersaturation below 5% grew by a screw dislocation mechanism. The temperature dependence of the habit of the ice crystal grown under this growth condition can be qualitatively explained by the anisotropy of the condensation-coefficient and its temperature dependence.

1. Introduction

It is well-known that the habit of snow crystals growing in air at 1.0×10^5 Pa varies with temperature [1-3]. The mechanism of the temperature dependence of the habit change of snow crystals has been studied by several authors [4-6]. Recently, a new interpretation of the temperature dependence of the habit change of snow crystals growing in air at 1.0×10^5 Pa at water saturation has been proposed [7,8].

Generally speaking, the snow crystals growing in air at 1.0×10^5 Pa depend on three processes described below. (1) volume diffusion process of water molecules to the crystal surfaces (2) surface kinetic process for incorporation of water molecules into the crystal lattice (3) transport process of the latent heat of sublimation by conduction. Here, it is considered that the temperature dependence of the habit of ice crystals growing from the vapor phase is closely related with the surface kinetic process for incorporation of water molecules into the crystal lattice. Therefore, the study of ice crystal growth under such circumstances as the resistance of the volume diffusion of water molecules and the resistance of the transport of the latent heat of sublimation are ignored is very important because under this growth condition, only the effect of the surface kinetic process of water molecules can be studied. Accordingly, in this study, ice crystals were grown on a cover glass at various temperatures in air at 4.0×10 Pa.

The studies on ice crystal growth on a substrate at a low air pressure have been carried out by different researchers [9-12]. Recently, Gonda and Sei [13] studied the habit of ice crystals grown on a cover glass in air at 4.0×10 Pa and found that

it depended not only on air temperature but also on supersaturation over ice and crystal size. The purposes of this paper are to study the temperature dependence and the mechanism of the habit change of an ice crystal growing on a substrate in air at 4.0×10 Pa and at a relatively low supersaturation.

2. Experimental

A growth chamber and the method of its measurement are described in detail in a previous paper [14]. Ice crystals were nucleated in air at 4.0×10 Pa at -7 , -15 and -30°C and at 1.9% supersaturation by inserting a small amount of silver iodide smoke into the growth chamber. The temperature dependence of the habit change of an ice crystal was measured at $300 \mu\text{m}$ in size by repeating the growth and evaporation of the same ice crystal.

Next, to investigate the growth mechanism of the $\{0001\}$ and $\{10\bar{1}0\}$ faces of the ice crystal, the supersaturation dependence of the normal growth rate of these faces and the supersaturation dependence of the advance rate of macrosteps (step velocity) on these faces were measured, and the surface microstructure of the $\{0001\}$ and $\{10\bar{1}0\}$ faces was also observed in situ using a differential interference microscope.

3. Experimental results

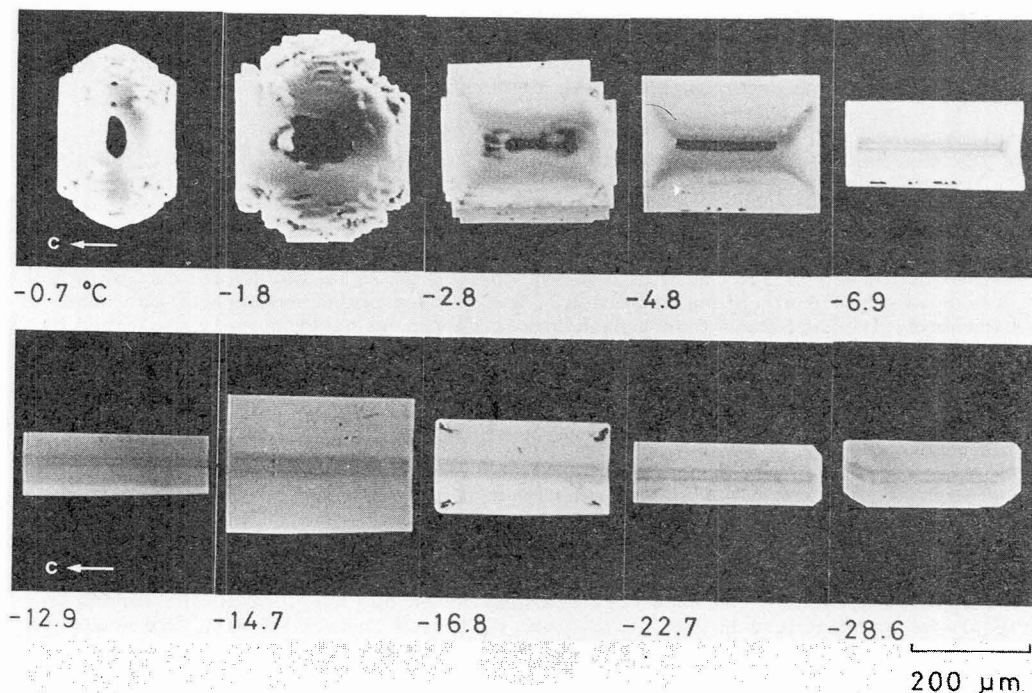


Fig. 1 Temperature variation of the habit of an ice crystal formed under air pressure of 40 Pa and 1.9% supersaturation

Figure 1 shows the temperature variation of the habit of an ice crystal at $300 \mu\text{m}$ in size formed by repeating the growth and the evaporation of the same ice crystal. In the present experiment, it is found that the transition temperatures from hexagonal column to hexagonal plate are almost -6.5 , -13.5 and -15°C .

Figure 2 shows the temperature dependence of the ratio of growth rates of the ice crystal shown in fig. 1.

From figs. 1 and 2, at a temperature above -6.5°C , a hexagonal plate grows; at the temperature range -6.5 to -13.5°C , a hexagonal column grows; at a temperature near -14°C , a hexagonal plate grows; and at a temperature below -15°C , a hexagonal column grows. The habit change of an ice crystal growing in air at 4.0×10^5 Pa with temperature roughly coincides with the temperature dependence of the habit change of an ice crystal growing in air at 1.0×10^5 Pa. Here, in the case of present experiment, the ratio of growth rates $R\{0001\}/R\{11\bar{2}0\}$ of an ice crystal is in the range of 0.5 to 2.0, while the size ratio c/a of ice crystals growing in air at 1.0×10^5 Pa is in the order of 10^{-2} to 10 [15].

Next, the supersaturation dependence of the normal growth rate of the $\{0001\}$ and $\{10\bar{1}0\}$ faces and the supersaturation dependence of the advance rate of macrosteps on the $\{0001\}$ and $\{10\bar{1}0\}$ faces of an ice crystal growing at each temperature were measured to study the growth mechanism of the ice crystal.

Figures 3 and 4 show the results at 4.0×10^5 Pa at -7 , -15 and -30°C . In the fig. 3, each dotted straight line shows Hertz-Knudsen equation, and each solid curve shows the theoretical curve when an ice crystal grows by a screw dislocation mechanism. α is the condensation coefficient which means the rate to incorporate water molecules impinging on the crystal surfaces from the vapor phase into the crystal lattice. From the comparison of the experiment with the theory, it is considered that the $\{0001\}$ and $\{10\bar{1}0\}$ faces of the ice crystal grown at each temperature and at a supersaturation below 5% grow by a screw dislocation mechanism.

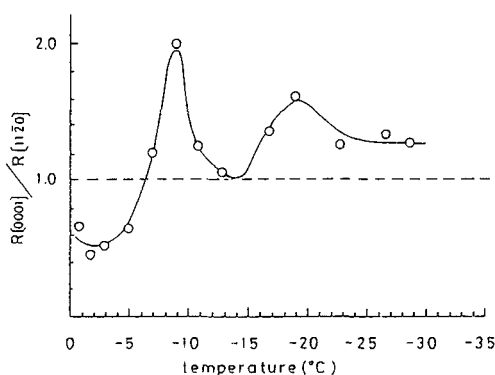


Fig. 2 Temperature dependence of the ratio of growth rates $R\{0001\}/R\{11\bar{2}0\}$ of an ice crystal grown in air at 4.0×10^5 Pa and at 1.9% supersaturation.

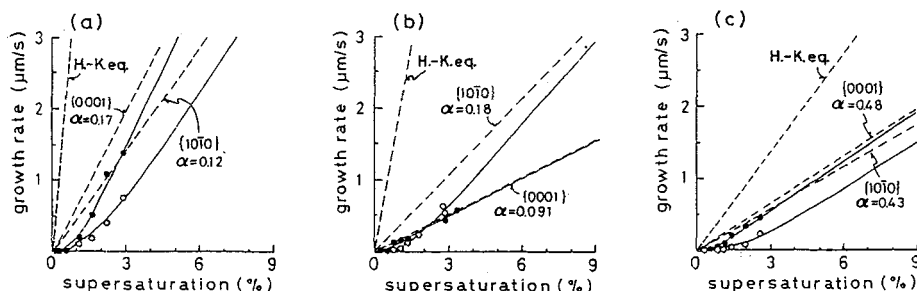


Fig. 3 Supersaturation dependence of the normal growth rate of the $\{0001\}$ and $\{10\bar{1}0\}$ faces of an ice crystal grown at (a) -7 , (b) -15 and (c) -30°C . Solid and open circles show the experimental values of the growth rates of the $\{0001\}$ and $\{10\bar{1}0\}$ faces, respectively. α is the condensation coefficient.

On the other hand, the growth steps originated from screw dislocations were observed on the $\{0001\}$ and $\{10\bar{1}0\}$ faces of ice crystals grown at -7 , -15 and -30°C . In the fig. 4, solid and open circles show the experimental values of the step velocity on the $\{0001\}$ and $\{10\bar{1}0\}$ faces, respectively, and solid and dotted curves show the theoretical curves of the step velocity originated from a screw dislocation. It is

considered from the figure that the $\{0001\}$ and $\{10\bar{1}0\}$ faces of the ice crystals grown at each temperature grow by a screw dislocation mechanism.

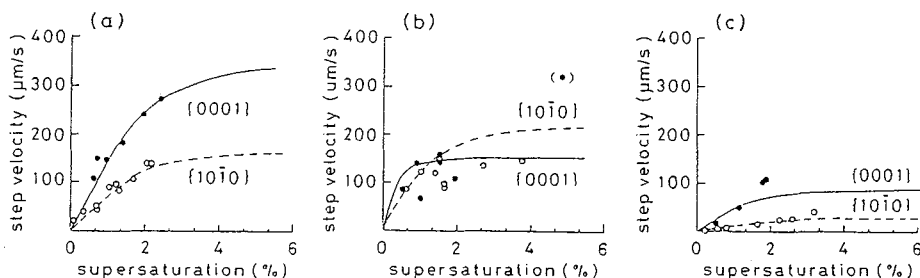


Fig. 4 Supersaturation dependence of the advance rate of macrosteps (step velocity) on the $\{0001\}$ and $\{10\bar{1}0\}$ faces of an ice crystal grown at (a) -7°C , (b) -15°C and (c) -30°C . Solid and open circles show the experimental values of the step velocity on the $\{0001\}$ and $\{10\bar{1}0\}$ faces, respectively.

4. Discussion

It was found from figs. 3 and 4 that both the $\{0001\}$ and $\{10\bar{1}0\}$ faces of an ice crystal grown at -7°C , -15°C and -30°C and at a supersaturation below 5% grew by a screw dislocation mechanism. In spite of this experimental fact, as shown in figs. 1 and 2, how does the habit of an ice crystal grown at 1.9% supersaturation systematically varies with decreasing temperature three times at a temperature between -0.8°C and -30°C ?

Figure 5 shows the temperature dependence of the condensation coefficient of the $\{0001\}$ and $\{10\bar{1}0\}$ faces of an ice crystal grown at -7°C , -15°C and -30°C and at 5% supersaturation. As shown in the figure, it is considered that the temperature dependence of the habit change of an ice crystal grown in air at $4.0 \times 10^4 \text{ Pa}$ and at a supersaturation below 5% can be qualitatively explained by the anisotropy and the temperature dependence of the condensation coefficient α . That is to say, at -7°C , a columnar ice crystal grows as a result of $\alpha_{\{0001\}} > \alpha_{\{10\bar{1}0\}}$; at -15°C , a plate-like ice crystal grows as a result of $\alpha_{\{10\bar{1}0\}} > \alpha_{\{0001\}}$; at -30°C , a columnar ice crystal grows as a result of $\alpha_{\{0001\}} > \alpha_{\{10\bar{1}0\}}$.

Generally speaking, the condensation coefficient α increases with decreasing temperature, but the condensation coefficient α on the $\{0001\}$ face of an ice crystal grown at -15°C becomes smaller than that at -7°C because the number of growth hillocks on the $\{0001\}$ face of the ice crystal at -15°C decreases more than that at -7°C . This fact was found by observing in situ the crystal surfaces using a differential interference microscope. The growth mechanism of an ice crystal growing at a temperature above -6.5°C is under investigation.

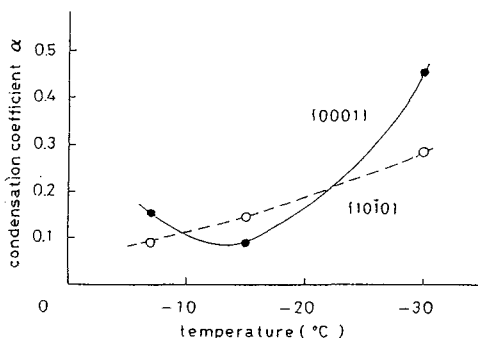


Fig. 5 Temperature dependence of the condensation coefficient α on the $\{0001\}$ and $\{10\bar{1}0\}$ faces of an ice crystal grown at -7°C , -15°C and -30°C and at 5% supersaturation. Solid and open circles show the values on the $\{0001\}$ and $\{10\bar{1}0\}$ faces, respectively.

5. Conclusions

The temperature dependence and the mechanism of the habit change of an ice crystal grown in air at 4.0×10 Pa and at 1.9% supersaturation was studied by repeating the growth and evaporation of the ice crystal. The results obtained are as follows:

(1) The habit of an ice crystal grown in air at 4.0×10 Pa and at 1.9% supersaturation depends on temperature. This experimental result roughly coincides with the temperature dependence of the habit change of ice crystals grown in air at 1.0×10^5 Pa.

(2) The $\{0001\}$ and $\{10\bar{1}0\}$ faces of the ice crystal grown at a supersaturation below 5% grow by a screw dislocation mechanism.

(3) The temperature dependence of the habit of an ice crystal growing in air at 4.0×10 Pa and at a relatively low supersaturation can be qualitatively explained by the anisotropy and the temperature dependence of the condensation coefficient α on the $\{0001\}$ and $\{10\bar{1}0\}$ faces.

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COMMENTS

N.FUKUTA

Question :

I again have a difficulty of understanding your use of screw dislocation mechanism for snow crystal growth. Do you have any direct evidence of screw dislocation existence in the crystal ?

Remark :

Your measurement of condensation coefficient seems to be in good agreement with ours which was done earlier.

Answer :

In our experiments, ice crystals grow on a substrate. It can be considered that screw dislocations arise when ice crystals nucleate in air by the role of silver iodide smoke or ice crystals grow on the substrate. Therefore, we can observe the growth hillocks on the ice crystal surfaces, in a center of which screw dislocations emerge. In addition, we can observe that the spiral growth steps spread continuously from the center of the growth hillocks.