Stable Growth of Ice Crystals under Microgravity

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The ice crystal growth experiments were carried out from December 2008 to February 2009 to understand the mechanisms of the morphological transition. However, the transition was not observed at all in space. Hence, we introduce a two-dimensional model to understand the stabilization mechanisms. From the comparison between the numerical and experimental results, it is found that the stabilization mechanism is the Gibbs-Thomson effect. The convection influence on the ground is also investigated. From the estimation of the Nusselt number, it is found that the convection strongly affects the temperature distribution even though the small supercoolings.

Key Words: Ice Crystal, Stable Growth Mechanisms, Microgravity, Numerical Analysis

Nomenclature

ρ_i	:	Density of water ρ_L or ice ρ_S
C_{p_i}	:	Specific heat of water C_{p_L} or ice C_{p_S}
κ_i	:	Thermal conductivity of water κ_L or ice κ_S
T	:	Temperature
r, z	:	Coordinates
ℓ_{SL}	:	Latent heat per unit volume
f	:	r -coordinate of an interface
$\tilde{\alpha}_1, \tilde{\alpha}_2$	:	Stiffness
R_1, R_2	:	Principal radii
T_m	:	Melting point of ice
T_m'	:	Melting point affected by the Gibbs-Thomson effect
θ_i	:	Angle between the normal and datum lines
σ	:	Interfacial tension of side surface
σ_0	:	Interfacial tension of the {1010} plane
u	:	Radial flow rate

Subscripts

S	:	Operation towards the solid phase
L	:	Operation towards the liquid phase

1. Introduction

A disk ice crystal in water is a typical shape of a stable morphology. However, the disk shape changes sooner or later to unstable shapes under supercooling conditions on the earth. First, a radius of one basal plane becomes larger than that of another plane. Then the edge on the larger plane begins to undulate. The undulations become larger and larger like cellular growth. Finally, they change to dendrites. These processes have been investigated experimentally¹⁻⁴⁾ and theoretically⁵⁻⁸⁾ for a long time. However, the trigger of the morphological instability is not clear yet. Temperature

nonuniformity caused by the thermal convection may be one of the main reasons why it is difficult to fully understand the instability mechanisms. Therefore, a microgravity experiment of ice crystal growth was planned in 1993. The experiment was conducted from December 2008 to February 2009 on the International Space Station (ISS)⁹⁻¹¹⁾ to observe the transition from the stable to unstable shapes. The transition, however, was not observed at all. Namely, the stable crystal in small supercooling temperatures was kept stable, while the dendritic crystal grew from the initial stage. The experimental results quite differ from those on the ground. As the first step to reach the final goal, which is to know the transition mechanisms, we investigate the stabilization mechanisms. If the mechanisms are successfully understood, all conditions required for the stable growth will be predicted. We also investigate the convection influence on the temperature distribution because

Fig. 1. Experimental setup for ice crystal growth: the cross sections of the (a) top view and (b) side view.

Fig. 2. Typical experimental results of stable growth: (a) the initial stage and (b) 1952 sec. later from (a). The supercooling is set to 0.03 K.

we should know the data reliability accumulated on the ground.

2. Experimental Setup

The ice crystal experiments were carried out by using the Ice Crystal Cell (ICC), which is set on a cold plate inside the Solution Crystallization Observation Facility (SCOF). The SCOF is installed on the “Kibo” module of the ISS. The ICC has small equipment shown in Fig. 1. This equipment has two main components named the nucleation cell and the growth cell. The size of the growth cell is 26 mm in diameter and 24

Fig. 3. Typical experimental results of unstable growth: (a) the initial stage and (b) 1952 sec. later from (a). The supercooling is set to 0.06 K.

Fig. 4. Typical experimental results on the ground. The supercooling setting is 0.02 K. Upper and lower figures represent the crystals at the initial stage and 25 min. later from the time of upper figure, respectively.

mm in height. Each cell is made of copper blocks for the sufficient temperature stability. These cells are connected with a capillary tube of 300 μm in inner diameter. Heavy water is filled in each cell and the tube due to the higher refractive index than light water.

To initiate the experiment, the nucleation cell is rapidly cooled by up to 10 degrees below the melting point to cause the nucleation surely. On the other hand, the growth cell is maintained at a certain supercooled temperature. Since the growth cell requires the temperature stability as good as possible, we use the thermistor with the highest measurement precisions which the SCOF has. Thus the measurement error is estimated to be less than 0.044 K. After the successful nucleation, the nuclei grow and propagate through the capillary. It may be expected that a single grain is selected in the tube. The grain appears in the growth cell after a certain time, depending on the supercooling condition. The temperature distribution is measured by interferometry. To calibrate the absolute value of the interferometry data, a thermistor is set inside the growth cell, of which precision is within 0.45 K.

3. Experimental Results

The typical experimental results in space are shown in Fig. 2. Figure 2(a) shows the initial stage just after the crystal appears at the capillary tip, while Fig. 2(b) is a snapshot at the end time of this experiment, that is, 1952 sec. later from Fig. 2(a). The supercooling is set to 0.03 K in this case. Please pay attention that the supercooling setting may not be equal to the actual supercooling due to the measurement and control errors. It is found from this figure that the disk shape is maintained over the experimental period. The stable crystals are observed

Fig. 5. Time evolution of crystal thickness and radius. The red and blue lines represent the thickness and radius, respectively. The supercooling is set to 0.04 K.

only under the supercooling settings of 0.04 K or less. In the cases of larger supercooling settings of 0.06 K or more, no disk crystals can be observed. The typical example of this case is shown in Fig. 3. The supercooling setting is 0.06 K. The undulating crystal is observed at the initial stage. The unstable growth is more clearly observed 1952 sec. later from Fig. 3(a), which is the same time as the Fig. 2(b).

On the other hand, the crystal morphology varies from the stable to unstable shapes on the ground even though at small supercoolings. The typical results at the small supercooling setting of 0.02 K are shown in Fig. 4. This figure clearly shows the morphological transition. This is the common phenomenon on the ground from the small to large supercoolings. The reason why the difference in the experimental results between in space and on the earth occurs may be the change of the temperature distribution due to the buoyancy-driven convection. This is discussed later.

4. Model

We describe a two-dimensional model expressing the stable growth under microgravity. Since the convection is negligible, the governing equations are as follows.

$$\rho_i C_{p,i} \frac{\partial T}{\partial t} = \kappa_i \left(\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial z^2} \right) \quad (1)$$

In addition, we should consider the energy balance at interfaces. The balance equations on the prism plane (side surface) and on the basal plane (top and bottom surfaces) are expressed in Eqs. (2) and (3).

$$\ell_{SL} \frac{\partial f}{\partial t} = -\kappa_L \left(\frac{\partial T}{\partial r} - \frac{\partial T}{\partial z} \frac{\partial f}{\partial z} \right)_L + \kappa_S \left(\frac{\partial T}{\partial r} - \frac{\partial T}{\partial z} \frac{\partial f}{\partial z} \right)_S \quad (2)$$

$$-\kappa_L \left(\frac{\partial T}{\partial z} \right)_L + \kappa_S \left(\frac{\partial T}{\partial z} \right)_S = 0 \quad (3)$$

The prism plane does not show the faceted surfaces but shows the rounded surfaces. This indicates that the roughening transition has occurred. The radial growth should be equivalent to the adhesive growth. Therefore, we use Eq. (2) to represent the radial growth. On the other hand, we do not consider the perpendicular growth to the basal plane as represented in Eq. (3) since the crystal thickness is almost

Fig. 6. Comparison between the numerical and experimental results by varying the interfacial tension. The open circles and open squares represent the experimental results under microgravity. The blue, red, green and brown lines represent the numerical results corresponding to the interfacial tensions of 15, 20, 25 and 30 mJ/m².

constant in small supercoolings as shown in Fig. 5. The perpendicular growth should be caused by the two-dimensional nucleation or spiral growth. However, both phenomena require a certain supercooling being more than a threshold to appear. Therefore, this is qualitatively consistent with the experimental results at small supercoolings.

If the side surface at a certain z -position becomes convex towards the liquid, the temperature gradient towards the liquid at that z -position increases. This means that the convex shape becomes more convex. This is similar effect to the Berg effect. Hence, we should consider the stabilization mechanisms. In our study, the Gibbs-Thomson effect expressed in Eq. (4) is used as the mechanism.

$$T_m' = T_m - \frac{T_m}{\ell_{SL}} \left(\frac{\tilde{\alpha}_1}{R_1} + \frac{\tilde{\alpha}_2}{R_2} \right) \quad (4)$$

The stiffness is described as follows.

$$\tilde{\alpha}_i = \sigma (\tan \theta_i) + \frac{\partial^2 \sigma}{\partial \theta_i^2} \quad (5)$$

We assume that the dependency of the interfacial tension on the angle to be negligibly small. Thus we obtain the stiffness as follows.

$$\tilde{\alpha}_1 = \tilde{\alpha}_2 = \sigma_0 \quad (6)$$

Although the interfacial tension of the {1010} plane has been reported, the value varies from 15 to 33 mJ/m² [12-14]. Therefore, we estimate the interfacial tension first. We calculate the crystal growth by varying the interfacial tension and compare the numerical results with the experimental results in space. The comparison is shown in Fig. 6. The supercooling is adjusted in each calculation so that the final crystal size can be almost the same as each other. In the microgravity experiments, the ice crystal did not grow in the supercooling setting of 0.02 K, while it grew in the supercooling setting of 0.03 K. This indicates that the actual supercooling should be 0.01 K or less in the supercooling setting of 0.03 K. Therefore, the interfacial tension of 15 to 20 mJ/m² should be the reasonable values. By comparing the calculations in the cases of 15 and 20 mJ/m², the result of 20 mJ/m² is more consistent with the experiments. Thus we use the

Fig. 7. Comparisons between the numerical and experimental results in the supercooling settings of (a) 0.03 K and (b) 0.04 K. The open symbols represent the experimental results under microgravity. The red, blue and green lines represent the simulation results.

20 mJ/m² as the interfacial tension in this paper.

5. Comparison of Numerical Results

To solve the governing equations, we use the boundary-fitted coordinate (BFC) method. This is one of the difference methods. The BFC method is applicable to the non-rectangular coordinates by transforming the coordinates in the real space to those in the computational space. The typical results in the supercooling settings of 0.03 and 0.04 K are shown in Fig. 7. From this figure, it is found that the simulation results agree well with the experimental ones until about 40 to 50 min. in the case (a) and about 25 to 30 min. in

Fig. 8. Temperature comparisons between the experiments and simulations. The red solid and dashed lines represent the calculations in the supercoolings of 0.01 K and 0.018 K, respectively. The blue solid and dashed lines represent the experiments at the supercooling settings of 0.03 K and 0.04 K.

the case (b). After those time periods, the crystal in the experiments grows more slowly than that in the simulations. This should be caused by the ice growth on the wall of the growth cell. The ice crystal grows not only towards the water but also along the capillary tube. Once the ice crystal growing along the capillary reaches the cell wall, the ice grows on the wall. This means that the supercooling is cancelled and thus the growth rate will become smaller. Therefore, the regions of (a) from 50 min. to the end and from 30 min. to the end are inadequate for the comparison.

In order to verify the simulation reliability, the temperature at the position of the thermistor inside the growth cell is compared. The results are shown in Fig. 8. Although the thermistor installed inside the cell has the measurement accuracy of ± 0.45 K as mentioned previously, the measurement stability is much better. This means that the measured temperature may have some offsets from the reference point but the temperature drift may be small. This expectation should be correct since the temperature behavior from the simulation agrees well with that from the experiments except for the absolute values. The difference of the temperatures, however, is 0.13 K and satisfies the specification of the measurement circuit.

6. Convection Influence

As mentioned at the last in the section 3, we discuss the convection influence here. The temperature distributions in 1 g and 0 g are calculated and are shown in Fig. 9. The supercooling is set to 0.02 K in Fig. 9(a), while it is set to 0.03 K in Fig. 9(b). It is found that the temperature distribution in 1 g is strongly affected by the convection. To verify the convection influence, we estimate the local Nusselt number at the side surface.

$$Nu = 1 + \left| \frac{\rho C_p u T}{\kappa_L \frac{\partial T}{\partial r}} \right| \quad (7)$$

The denominator of the second term of the right side is the heat flux in 0 g. The radial flow rate and the temperature gradient in 0 g are obtained from the simulations. The flow rate of 9.7×10^{-7} m/s and the temperature gradient of 9.6 K/m

Fig. 9. Comparison of temperature distributions between on the ground and in space. The supercoolings in (a) 1 g and (b) 0 g are set to 0.02 K and 0.03 K, respectively.

are obtained. By substituting the density of $1.1055 \times 10^3 \text{ kg/m}^3$, latent heat of 4217 J/(kg K) , temperature of 276.95 K , thermal conductivity of 0.561 W/(m K) , flow rate and temperature gradient for Eq. (2), the local Nusselt number is calculated to be about 230. This value indicates that the convection strongly affects the temperature distribution even though the small supercooling of 0.02 K . Intuitively, such the small supercooling may not affect the temperature distribution so much. However, this supposition is wrong. Although further studies are required to understand the convection influence from small to large supercoolings more deeply, the small dominator due to the small supercoolings may enhance the Nusselt number. It is summarized that the microgravity environment is indispensable for the ice crystal growth experiments.

7. Conclusions

Although the morphological transition from disk shapes to dendrites is common phenomenon on the ground, such the transition was not observed in the microgravity experiments. To understand the stabilization mechanisms of the disk shape, we introduce the two-dimensional model. By comparing the numerical results with experimental ones, it is found that our model explains the stable growth well and the stabilization mechanism should be the Gibbs-Thomson effect. We also investigate the necessity of the microgravity environment in the ice crystal growth. From the investigation, it is clarified that the temperature distribution on the ground is strongly affected by the convection even though the small supercooling conditions such as 0.02 K . This may be one of the reasons why the transition was not observed on orbit.

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