Pattern Formation during Ice Crystal Growth

- ISS KIBO Experiments -

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Crystal growth experiments of ice in supercooled D2O water were carried out in the Japan Experiment Module KIBO of the International Space Station (ISS) in the period between December 2008 and March 2009. Overview of this project and preliminary results are presented.

Key Words: Ice Crystal, ISS KIBO, Crystal Growth, Morphological Instability, Pattern Formation

1. Introduction

Fundamental research of crystal growth mechanism is one of the important topics that should be conducted in the microgravity condition. Especially the morphological instability and the pattern formation are strongly affected by the effect of disturbance such as the free convection occurred in the environment around the growing crystal. An ice crystal growing in supercooled pure water changes from circular disk to perturbed disk and finally to well-developed dendrite with hexagonal symmetry1-3), which is completely different from the dendritic pattern formation observed for the previous research material such as Succinonitrile (SCN)4-6).

The growth of the basal planes of an ice disk is controlled by the kinetic manner, whereas the side plane is completely rough interface and its growth is controlled by the dissipation of the released latent heat7-9). That is, two kinds of growth planes with completely different growth mechanisms coexist in one crystal and its pattern formation is dominated by the interaction between both planes.

Japan Experiment Module “KIBO” was launched by Space Shuttle and attached to International Space Station in 2008. In order to clarify the pattern formation mechanism of ice crystal grown in supercooled water, ice growth experiment (recognized as the code name of ICE CRYSTAL) was carried out in the KIBO as the second experiment in the period between December 2008 and March 2009. In this paper, we give the overview of this space experiment.

2. Properties for Pattern Formation of Ice Crystal

Growth process of an ice crystal is divided in two stages as shown in Figure 1. First stage is the stable-growth in which the ice crystal grows with maintaining the shape of the thin circular disk. The growth of the edge plane of an ice disk is dominantly controlled by the thermal diffusion efficiency of the latent heat released during growth, but the growth of the
basal planes with flat interface is controlled by the layer-by-layer kinetics. Shimada and Furukawa\(^3\) experimentally clarified that this growth stage further fell into two types depending on the growth mechanisms of basal planes, namely, either the spiral growth or the two-dimensional nucleation growth. After the morphological instability, second stage of ice crystal growth (instable growth) starts and develops the dendritic pattern with the hexagonal symmetry. In this stage, the analogous extension of crystal pattern is not observed any more.

Ground experiments have clarified that the transition from the stable to instable growth stages, namely the morphological instability, occurs at the edge plane of the ice disk when the disk thickness reaches a critical crystal thickness, \(h_c\). The clear relationship of \(1 - \Delta \propto Th_c\) has been also found from the measurements of the critical thickness as a function of initial supercooling temperature\(^6\). Furthermore, the instability initially starts in the direction of thickness (namely, along the c-axis), and uninterruptedly occurs in the circumferential direction. It is to be noted that the cross section of edge plane is not symmetrical in an instance of instability occurrence, which means that the symmetry breaking is observed during the development of instability along the edge plane.

On the other hand, Yokoyama et al.\(^8,9\) has developed a new model to understand the growth of ice disk and the morphological instability at the disk edge. The appearance of an asymmetrical pattern that occurs when a disk crystal of ice grows from supercooled water was studied by using an analysis of growth rates for radius and thickness (see Figure 2). The growth of the radius is controlled by transport of latent heat and is calculated by solving the diffusion equation for the temperature field surrounding the disk. The growth of the basal planes is governed by the generation and lateral motion of steps and is expressed as a power function of the supercooling at the center of a basal face. They indicated that the critical thickness was related to the critical condition for the stable growth of a basal face and a difference of growth rates between two basal faces was a possible mechanism for the appearance of the asymmetrical shape. An alternative explanation for the symmetry breaking has been given by Xu and Shimizu\(^10,11\).

The discriminative properties observed during the ice crystal growth are completely different from the general mechanism of dendritic growth based on such the theoretical prediction as the classical model as typified by Langer Müller-Krumbhaar theory\(^12\). Understanding the pattern formation mechanism of ice crystal will give a new perspective for the studies of pattern formation and morphological instability. For the delicate properties of the ice crystal patterns, the experiments under the microgravity condition without any disturbance will give the most ideal observations of pattern developments. Actually, some experimental results for ice crystal growth under the short-term microgravity condition strongly indicate that the effect of thermal convection around the growing crystal is critically large for the crystal growth\(^13,14\). Consequently, it is very important to conduct the experiments of ice crystal growth under the long-term microgravity realized in the ISS.

3. Ice Growth Experiments at ISS-KIBO

3.1 Experimental apparatus

An experimental apparatus for ice growth experiment in ISS-KIBO, which is recognized as “Ice Crystal Cell”, was developed. Figure 3 shows a picture of ICE CRYSTAL CELL, which was composed by the ice growth apparatus and the optical system. Figure 4 is the illustrative cartoon to indicate the basic concept of the ice growth apparatus. The apparatus was configured by two cells which were for the free growth of ice and for the nucleation of ice; namely, a growth cell and a nucleation cell, respectively. Both cells were connected by a thin glass capillary. Temperatures of both cells were separately controlled by peltier module+PID controllers. The ice growth apparatus was fulfilled by pure
heavy water (D₂O) and the free growth of an ice single crystal occurred at the tip of glass capillary inserted into the center of the growth cell. The melting point of D₂O is +3.8°C, which is higher than that of H₂O. Furthermore, temperature dependences of both the density and the refractive index of D₂O around the melting point are larger than those of H₂O. Consequently, D₂O is more convenient for the space experiments than H₂O.

3.2 Procedures of KIBO experiments

The ICE CRYSTAL CELL was launched by the Space Shuttle flight (STS126) on 14 November 2008 (EST). The apparatus was installed in the Solution Crystalization Observation Facility (SCOF) of KIBO and the space experiments were started on 2 December 2008 (JST).

The experiments were carried out by the following procedures. After the temperature of water sample in the growth cell reached the prescribed supercooling temperature and was homogenized, the nucleation cell was rapidly cooled down. Ice crystals were nucleated in the nucleation cell and then the instantaneous rise in temperature of nucleation cell by the release of latent heat was observed. Nucleated ice crystals continued to grow in the glass capillary and then only one ice crystal was able to survive by the growth competition when the ice crystal reached to the end of capillary. In this way, the free growth of a single ice crystal was started at the tip of capillary in the center of growth cell. The growth behaviour of ice crystal was observed by two-axes interference microscopies, which were assembled in the ICE GROWTH CELL and SCOF. The images obtained by these optical systems were downlinked from the KIBO-ISS to the KIBO control center in Tsukuba Space Center of JAXA.

![Image of the free growth apparatus of ice crystal under the microgravity condition.](image)

**Fig. 4.** Basic concept for the free growth apparatus of ice crystal under the microgravity condition. It is composed of the growth cell and the nucleation cell. A thin glass capillary connects both cells. Ice crystals are nucleated in the nucleation cell and the competition among the neighboring ice particles may occur in the capillary. Finally, only one single crystal can survive at the tip of capillary. The nucleation cell is dispensable for the laboratory experiments, because it is possible to nucleate ice particles by various cooling methods. However, this is the most important innovation for the space experiment apparatus.

![Image of time sequence pictures of an ice crystal growing in supercooled D₂O water in space.](image)

**Fig. 5.** Time sequence pictures of an ice crystal growing in supercooled D₂O water in space. Size of each picture is 4.8mm long and 6.4mm wide. The ice crystal has a hexagonal symmetry, but only three branches grow by the effect of glass capillary, which is seen at the right edges of pictures. Initial supercooling temperature was 0.5K.
When the ice crystal sufficiently grew up, both the growth and nucleation cells were heated up to the temperature higher than the melting point of ice. Then the ice crystals were completely melted, and the growth apparatus was easily reset by this way. It is to be emphasized that the growth experiments could be repeated only by a simple sequence of temperature changes.

KIBO experiments were carried out for three months until the beginning of March 2009, and totally repeated 134 times for various supercooling temperatures.

4. Ice Crystal Growth Observed in Space

A typical ice crystal observed in space is shown in Figure 5. The initial temperature of supercooled water in this experiment was +3.3°C (0.5K supercooling). More symmetrical pattern is obtained compared of that of the ice crystal grown on the ground. Though the detailed analysis obtained by KIBO experiments is in operation at the present time, the brief summaries are given as follows.

1) 134 experiments were successfully repeated for various supercooling temperatures using the ICE CRYSTAL CELL.
2) Images by two-axes interference microscopes were separately obtained and used for the analysis of three dimensional pattern developments of ice crystals and the thermal diffusion fields around the growing crystals.  
3) The symmetry breaking during the morphological instability was not always observed for ice crystals growing in space.  
4) The growth rates of basal planes were qualitatively reduced for the ice crystal growth in space.  
5) Detailed quantitative analysis of the experimental data will be continued to complete within a year.

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References