Recent Crystal Growth Experiments onboard the ISS KIBO

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Since the launch of the Japanese Experiment Module KIBO in 2008, three crystal growth experiments have been performed. For each experiment, an Experiment Unit (EU) was developed based on the discussion with the Principal Investigator (PI). Using the EU, ground control experiments were conducted before flight. After launch, experiments were run repeatedly to check the reproducibility of the measurements. Experiment coordination process is introduced together with the overview of the experiments.

Key Words: Crystal Growth, SCOF, ISS, Microgravity

1. Introduction

Five years have passed since the Japanese Experiment Module KIBO started its operation in 2008. Microgravity experiment onboard the International Space Station (ISS) is no longer an extra-special event, it is rather one of the experiment facility/opportunity for scientists. Especially for crystal growth researchers, it is a unique environment where convection or sedimentation is highly suppressed. In this paper, we would like to summarize the process of crystal growth experiment coordination. Experiment coordinators are responsible to develop the hardware, support the space experiment operation, work together with the PI (Principal Investigator) group to obtain ground control data and etc. Overview of the experiments are also introduced.

2. Preparation Before Launch

Once the flight experiment proposal is approved, the experiment coordinators have an intense discussion with the PI to clarify the objectives, hypothesis and expected results. Next, the specifications of the Experiment Unit (EU) is discussed. EU is developed for each experiment according to its scientific requirements.

The EU will be connected to the Solution Crystallization Observation Facility (SCOF) which provides electric power and communication to the EU. Cooling water is provided to the cold plate on which the EU is installed. Therefore, the size and resource of the EU is strictly restricted. In order to meet the unique scientific requirements such as temperature control, motor control, and observation system, each EU is customized and designed carefully. Fig. 1 shows the various EUs.

SCOF is also equipped with an amplitude modulation microscope, bright field microscope, Mach-Zehnder type interference microscope with two wavelength light sources, and two cameras (x2 and x4). The field views are 2.4 mm x
The EU developed specifically for this growth of disk shaped/dendrite ice crystals. The experiment is named Ice Crystal Experiment. The first experiment was “Ice Crystal” which focuses on the observation of the crystal growth process. We check the function of the EU by actually growing and observing the crystal. Many try-and errors occur during the development. Through these tests, we finalize the experiment requirement, condition, and EU specification. Ground control data is also obtained before launch. Vibration test, off gassing test, EMC (Electro-Magnetic Compatibility) tests are also preferred to verify the safety of the EU.

In a nominal case, this preparation phase is about 3~5 years. However, the first two experiments were selected in 1993, so it was more than 15 years waiting for the KIBO launch.

3. Experiments on the ISS

After launch, the crew member installs the EU to SCOF. Then all the experiments can be operated from ground at the “Space Station Integration and Promotion Center (SSIPC)” located in Tsukuba Space Center, Japan. The MPEG-2 compressed video images and temperature data are simultaneously downloaded from ISS. During times when the satellite position is at LOS (loss of signal), if necessary, the video images are recorded on a hard disk onboard so that the whole crystal growth process could be reproduced by connecting the video image files. Before a long LOS (ex. 30min), we have to be careful because no images, no telemetry data are downloaded realtime. Of course, no commanding can be done during LOS, so the experiments have to be carefully scheduled to avoid critical commanding during LOS. All telemetry data (including temperature data) during LOS is downloaded later.

The microgravity environment is measured using MMA (Microgravity Measurement Apparatus) developed by JAXA. The gravity level is $1 \times 10^{-3}$ to $1 \times 10^{-5}$ g below 1Hz frequency during ISS night time. In order to avoid the effect of g-jitter, the experiments are conducted during the ISS night time when the crew members are asleep.

The experiments are run repeatedly to check the reproducibility of the measurements under various experiment conditions. Experiment coordinators always stay in the Users Operation Area (UOA) in SSIPC during the experiment, and inform the next experiment condition to the ground operator. When the PI group is not in the UOA, the coordinator has to decide the condition. Thus, the coordinators are required to have a deep understanding of the experiment objectives.

4. Ice Crystal Experiment

The first experiment was “Ice Crystal” which focuses on the growth of disk shaped/dendrite ice crystals. The experiment was proposed by Professor Yosshinori Furukawa (Hokkaido University). The EU developed specifically for this experiment was named Ice Crystal Cell (ICC). There is a crystal growth cell and a nucleation cell connected with a glass capillary inside the ICC (Fig.3). The growth cell has a cylindrical shape with a 26mm inside diameter and a 24mm inside length. The nucleation cell has a disk shape with a 6mm diameter and 1.2mm length. The cells and the capillary are completely filled with the sample (Heavy water, D$_2$O). D$_2$O was selected because of its higher melting temperature (i.e. 3.82 degrees) and its higher temperature dependence on refractive index when compared with H$_2$O. When the nucleation cell is cooled to -10 degrees by a Peltier device, the ice crystal nucleates in the nucleation cell. The ice crystal grows towards the crystal growth cell through the capillary glass. The crystal growth cell is temperature controlled to a certain undercooling level by Peltier devices. The operational range of the Peltier devices is from 1.82 degrees ($\Delta T$=2K) to 3.79 degrees ($\Delta T$=0.03K). The temperature control was stable in the range of ±0.02 degrees. This allows the study of the ice crystal growth under various undercooling conditions by observing the growth cell. After each experiment, the crystal is dissolved by heating the nucleation cell and the growth cell. The ICC is also equipped with a bright field microscope and Mach-Zehnder type interference microscope perpendicular to the light path of the SCOF (Fig. 3). Therefore, it is possible to observe the growing ice crystal from two axial directions. The ICC camera (x1) has a 4.8mm X 6.4mm field of view and a 100 pixels/mm resolution.

![Fig. 3. Schematic diagram of the Ice Crystal Cell (ICC).](image_url)

The experiment was run from December 2008 to February 2009 (total 134 experiment runs). Crystal growth was observed in various supercooling rate. First of all, it is noticed that the crystal shape is highly symmetrical in microgravity because the latent heat is transferred symmetrically by diffusion (Fig. 4). Next, The velocities of dendrite tips parallel to the a axis and the growth rates of basal faces parallel to the c axis were both analyzed under supercooling ranging from 0.03 to 2.0 K. Growth rates of the c axis was calculated using the interferometer fringes of the ICC (Fig. 5). From the analysis, it was concluded that the growth process of the basal faces has a significant influence on the tip velocities $^1$. The dendrite tip velocity was also shown to be smaller than that of...
terrestrial experiment results, because of the absence of convection. Dendrite tip curvature was measured, and discussed with comparison to succinonitrile crystal growth 2).

In very low supercooling such as 0.03K, the ice crystal forms a disk like shape (Fig. 6). The disk did not transform to dendrite during the experiment. A new growth model to understand the disk growth was reported 3).

5. Facet Experiment

The second experiment was “Facet” proposed by Professor Yuko Inatomi (JAXA). “Facet” aims to elucidate the mechanism of a facet-like crystallization (a crystal with flat surfaces) by precisely observing the phenomena at the solid-liquid (S/L) interface. Since it is well known that crystals tend to develop structural defects and capture impurities when a new facet is generated, it is necessary to examine the facet growth process to grow high-quality semiconductors and superconductors. This study may also contribute to the creation of new functional materials. Melt of salol (phenyl salicylate) and t-butanol was used as a model material (Fig. 7). Since the melt is optically transparent with a low melting point, it is suitable for in-situ observation of the facet growth.

As mentioned in Section 2, SCOF is equipped with a Mach-Zehnder type interference microscope with two wavelength light sources. Since the refractive index has a correlation with temperature and solution concentration, by obtaining images of the interferometer fringes at the growing crystal interface such as Fig. 8, the temperature and butanol concentration at the S/L interface can be calculated.

From April 2009 to June 2009 and September 2010 to October 2010, total 60 runs were carried out with various growth conditions. The results are now being analyzed 4,5). The brief summary of the obtained results at this point are as follows. (1) Temperature and concentration distributions in the vicinity of the S/L interface were successfully obtained with high resolution by using the in situ observation method. (2) The relationship between the growth rate and a kinetics undercooling at the S/L interface, which is known as a driving force of crystal growth, was obtained. (3) Breakdown of the growth interface occurred at the point with maximum kinetics undercooling. Splitting of the S/L interface was caused by the dense t-butyl alcohol, which decreased the melting point. (4) The numerical simulation based on a phase-field model validated the effect of latent heat on the morphological change.

Fig. 4. Ice Crystal on ground (a) and in microgravity (b) Photo size is 4.8mm X 6.4mm.

Fig. 5. Ice Crystal interferogram. Photo size is 4.8mm X 6.4mm.

Fig. 6. Disk shape Ice Crystal. Photo size is 1.2mm X 1.6mm.

Fig. 7. Faceted cellular array growth (salol/t-butyl alcohol alloy) Photo size is 2.4mm X 3.2mm.

Fig. 8. 532nm Interferogram of the Faceted cellular array growth. Photo size is 2.4mm X 3.2mm.
6. NanoStep Experiment

The third experiment was “NanoStep” which focuses on protein crystal growth mechanism. The experiment was proposed by Professor Katsuo Tsukamoto (Tohoku University). The perfection of space-grown protein crystals is generally known to be better under microgravity. However, the mechanism responsible for such conclusion is yet to be understood from the fundamental point of view. The objective is to measure the crystal growth rate as a function of supersaturation and observe the growing crystal surfaces by Michelson-type interferometer at a molecular level resolution. Such a detailed measurement has not been made in space. Lysozyme was used as a model protein.

Three sample cells were prepared with different protein/impurity concentrations. Experiment duration was 35 days per cell. In the first week, all the crystals were dissolved by increasing the temperature except for the seed crystal which was chemically fixed by glutaraldehyde. In the next week, the temperature was controlled to let the seed crystal grow slowly. After the whole seed crystal was covered by epitaxial growth layer, the angle of the crystal direction was precisely controlled from ground to obtain a good image of laser interferogram showing the surface topography of a protein surface (Fig. 9). Then, the temperature was controlled to change the supersaturation, and the crystal growth rate was measured in a nanometer order by calculating the movement of the interferometer fringes. Using SCOF Mach-Zehnder type interference microscope, the protein concentration gradient at the crystal interface was obtained (Fig. 10) simultaneously.

The experiment was carried out from August 2012 to December 2012, and the analysis is now ongoing.

7. Conclusions

“Ice Crystal” “Facet” and “NanoStep” experiment were conducted onboard the ISS KIBO using the SCOF. The experiment objectives, hardware, operation and brief results were introduced. In order to successfully carry out the experiment, the experiment coordinators work together with the PI group to support the preliminary ground experiments, develop the EU by try-and error, and operate the space experiment.

In 2013, another crystal growth experiment EU “Ice Crystal 2” will be launched by HTV4. PI is Professor Furukawa. Ice Crystal 2 examines the growth rates and stability of ice crystals in supercooled water including antifreeze glycoprotein (AFGP). The preferential adsorption of AFGP molecules at the ice/water interface controls the growth of ice crystals. Ice crystal oscillatory growth interacting with the interfacial adsorption of AFGP will be precisely observed in the microgravity environment where gravity-based convection is completely removed. The operation is planned from September to December, 2013.

References