Sounding rockets are suitable platforms to provide the opportunity to carry out research in materials science, fundamental science, and biology. However, no sounding rocket developed in Japan has been served for microgravity experiments after termination of TR-1A rocket launching, while TEXUS and MAXUS rockets are frequently used as platforms for microgravity research in Europe. The present paper briefly describes the following in situ observation experiments under microgravity during free-fall flights of ISAS sounding rockets S-520-24 and S-520-28: (1) faceted cellular array growth, (2) diamond synthesis, (3) nucleation of WO$_3$ and Fe in gas phase, and (4) nucleation of calcium carbonate. All the experiments were successfully performed in the 7 min microgravity conditions as a result of active control of spin of payloads. In the crystal growth experiments, heat and mass transport process near the crystals were optically visualized and then the kinetics on the growing crystal surface was quantitatively evaluated. In the nucleation experiments, nucleation rate under microgravity was drastically decreased compared with that under the terrestrial condition.

Key Words: Sounding Rocket, Free-Fall Flight, Materials Science, Microgravity Experiments

1. Introduction

From a viewpoint of research in microgravity sciences, the advantages of utilizing the ISS over the other platforms are as follows: (1) repeated experiments with a long duration, (2) operation and maintenance of experimental facilities by astronauts. On the other hand, sounding rockets are suitable platforms to provide the opportunity to carry out research in materials science, fundamental science, and biology. Their cost-benefit performances are superior to the ISS for testing new instrumentation or conducting precursor experiments for the other long duration flight, if a few minutes of experiments under microgravity are sufficient to obtain the required results.

However, no sounding rocket developed in Japan has been served for microgravity experiments after termination of TR-1A rocket launching, while TEXUS and MAXUS rockets are frequently used as platforms for microgravity research in Europe. S-520 sounding rockets have been developed and launched by ISAS/JAXA for dedication to astrophysical observation, upper atmosphere exploration, space plasma physics, etc., for scientific research. The rocket has a flexible payload capability for launching of a 100 kg with 520 mm diameter payload far above 300 km. Moreover, it has a possibility to provide more than 5 minutes for a microgravity environment in its parabolic trajectory.

In materials processing with fluid, temperature and concentration gradients become driving forces of convection under the terrestrial condition. An application of a microgravity environment is a promising method to suppress the complex convective heat and mass transfer and make deeper insight into phenomena during the materials processing. The present paper briefly describes in situ observation experiments of crystal growth and nucleation processes under microgravity using sounding rockets S-520-24 and S-520-28. The former experiment was to investigate the influence of heat and mass transfer upon crystal growth, and the latter was to evaluate homogeneous nucleation phenomena.

2. Experimental Procedure

The S-520-24 sounding rocket was launched on August 2, 2008 for the following two crystal growth experiments; (1) FCT: morphological change in a faceted cellular array growth, and (2) DIA: diamond synthesis in hydrogen gas on a silicon substrate. These were collaborative projects proposed by ISAS and Teikyo University of Science & Technology. The S-520-28 rocket was launched on December 17, 2012 for the following two nucleation experiments; (1) DUST: in situ visualization of nucleation environment in gas phase, and (2) CAL: detection of homogeneous nucleation of calcium carbonate. These were collaborative projects proposed by ISAS and Tohoku University.

In both cases of S-520-24 and S-520-28, the spin rate of the rocket was reduced by a yo-yo de-spin mechanism after combustion of the rocket motor was completed. Subsequently the spin rate of the payload, which was connected to the motor even in the parabolic flight, was adjusted to the desired value by a side-jet system in order to strongly dump centrifugal force. As a result of the active control of the spin, all the experiments were successfully performed in the 7 minutes
microgravity conditions, which were measured by accelerometer installed in payloads. All the experiments were carried out successfully and the telemetry data were transmitted to Uchinoura Space Center, JAXA.

2.1. FCT experiment

Some works on production of high quality semiconductor devices have reported break-down from a planar solid-liquid (S/L) interface to a faceted cellular array. Since this morphological development of the interface causes segregation of dopants, great interest in understanding the pattern formation in faceted cellular array growth has been taken. However, temperature and concentration gradients in the liquid become driving forces of convection in the liquid and the convection influences the morphological change of the S/L interface. Therefore, application of a microgravity environment is considered to be a promising method to investigate the morphological stability of the S/L interface.

An in situ observation experiment of faceted cellular growth was carried out in a purified phenyl salicylate crystal (so-called salol), which is known as a faceting material, under a microgravity condition. Temperature of the specimen was measured by two thermocouples and was controlled by Peltier devices and thermocouples. A common-path type microscopic interferometer with the same design as SFU/MEX, as shown in Fig. 1 (left) was mounted in the rocket. The obtained telemeter data were as follows: (1) temperature in the specimen cell, and (2) interference fringe image.

2.2. DIA experiment

Takagi et al. developed a new diamond synthesis method from vapor phase, the so-called graphite rod heating method, in which diamond could be deposited by Joule heating of a graphite rod in hydrogen atmosphere. This method enables us to produce industrial diamonds for a heat sink, nano diamonds for bone tissue engineering, and carbon nano materials such as fullerene and carbon nanotube. The advantages of this process over the other methods are rapid reaction rate and low pressure of reaction gas during deposition of the nanoparticles on a target substrate. However, the synthesis mechanism of the nanoparticles is still not clear. Thus, nucleation and growth processes should be investigated in situ from the view point of growth kinetics on the substrate as well as heat and mass transfer in the gas phase.

Figure 1 (right) shows the DIA setup. In this experiment, a graphite rod was heated by a DC power amplifier, which was controlled by a sequencer. Temperature of the graphite rod was measured by a pyrometer. Excited gas species were produced around the heated rod. The excited species were detected by a spectrometer as some peaks in an emission spectrum from the gas, because the excited species became luminescent at the process temperature. A silicon substrate was heated and the surface reaction was activate by radiation from the graphite rod during the Joule heating. The obtained telemeter data were as follows: (1) temperature on a silicon substrate, (2) temperature of the graphite rod, and (3) output of the spectrometer.

2.3. DUST experiment

Determination of supersaturation in gas phase from temperature and pressure at the nucleation and difference with results of the ground-based experiment in WO3 and Fe vapor were the main subjects of the this experiment.

Nucleation is a fundamental event to determine character, such as size, number density and morphology, of produced crystals. Therefore, understanding and control of nucleation process are crucial in various fields. Nucleation theories have been used to understand the nucleation temperature, number density and size of produced particles.

In the ground-based experiment, a hot evaporation source generates heterogeneity of nucleation temperature and concentration caused by strong convection of gas atmosphere. In addition, there is a possibility of that produced nuclei collides and were mixed by a micropump just before microgravity. The obtained telemeter data were as follows: (1) temperature in the specimen cell, and (2) interference fringe image.

2.4. CAL experiment

The aim of CAL experiment was to detect of homogeneous nucleation and to determine incubation time in aqueous solution of calcium carbonate with low concentration, which was difficult to achieve in airplane parabolic flight.

Aqueous solutions of sodium carbonate in 1 mL syringe and equivalent amount of aqueous solutions of calcium chloride were mixed by a micropump just before microgravity condition established, and consequently nucleation of calcium carbonate occurred in a specimen cell. Eleven specimen cells with different initial concentration from 3 mMol to 15 mMol were served for microgravity experiment, while the five cells were used for checkout of the CAL setup. The incubation time in each cell was simultaneously measured by an impedance measurement device at a frequency of 1 kHz and by a light-scattering measurement device, which were installed in the each cell. The specimen cell array unit, as shown in Fig. 2
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(right), including the sixteen cells, a control circuit for the unit and an amplifier circuit for the data acquisition were installed in an aluminum closed vessel, in which the aqueous solution were prevented from evaporation in space. The obtained telemeter data were as follows: (1) output of the impedance measurement devices, (2) output of the light-scattering measurement devices, (3) temperature at the specimen cell array, and (4) pressure in the vessel.

Fig. 1.  Experimental setups onboard S-520-24: (left) FCT, (right) DIA.

Fig. 2.  Experimental setups onboard S-520-28: (left) DUST, (right) specimen cell array unit of CAL.

3.  Results and Discussion

3.1.  FCT experiment

The morphology of the S/L interface and the interference fringe pattern corresponding to temperature distribution in the liquid were simultaneously measured by a microscopic interferometer as shown in Fig. 3 in order to evaluate the morphological instability taking account of released latent heat in salol. The obtained results provided basic data for a crystal growth experiment under a long-duration microgravity, which was carried out in Japanese Experiment Module “Kibo” of ISS from 2009 to 2010 5).

Fig. 3.  Growth interface of salol crystal under microgravity. Displacement of interference fringes corresponds to change of temperature change in liquid.

3.2.  DIA experiment

When diamond is synthesized by using the graphite rod heating method in hydrogen ambience, peaks of C2 at 383nm, CH at 397nm, C3 at 405nm, C2- at 486nm, and H at 656nm appear in an emission spectrum from the gas phase. In this experiment, nanodiamonds were synthesized in hydrogen gas on a silicon substrate. Some gas species were activated at 2000°C by the Joule heating of a carbon rod during the process. Active species H and H, which were difficult to measure on the ground due to the strong thermal convection, were confirmed by the onboard spectrometer. The peak of H, obviously increased as the heating time advanced, as shown in Fig. 4 6).

Fig. 4.  Emission spectrum from gas phase during diamond synthesis at X+99 second under microgravity.

3.3.  DUST experiment

Evaporated vapor was concentrically diffused uniformly, cooled and condensed in the gas atmosphere. The temperature and concentration at the nucleation site were determined from the movements of the interference fringes. The nucleation occurred very far from the thermal equilibrium and the supersaturation ratio was extremely high, more than 1010. Kimura et al. succeeded to determine surface free energy and sticking probability of manganese nanoparticle from condensation temperature and size of produced particles, which was determined by transmission electron microscopy, based on nucleation theories 7). Recently, a new project was started to determine the physical parameters of nanometer sized particles and evaluate nucleation theories by homogeneous nucleation experiments in vapor phase.
Nanoparticles were formed from a highly supersaturated vapor, supersaturation ratio was as high as the order of $10^4$, after evaporation by electrical heating in a gas atmosphere. The temperature and concentration at the nucleation sites were obtained by an in-situ observation system using interferometry as shown in Fig. 5.

3.4. CAL experiment

Tsukamoto et al. showed a possibility by aircraft parabolic flights that nucleation rate of calcium carbonate from aqueous solution under low gravity condition of approximately 20 seconds might be $10^4$ times slower than that under the terrestrial condition at the same supersaturation $^8)$. The reason of the difference of the nucleation rates is that homogeneous nucleation on a container surface and/or impurity particles overtakes heterogeneous nucleation under the terrestrial condition while the reverse is also taking place under microgravity. However, the initial concentration of the aqueous solution was high under the parabolic flight, because the maximum incubation time was limited within 20 seconds. Therefore, it was not clear that whether the nucleation rate and the incubation time for nucleation followed the same functions of supersaturation even with lower concentration.

In this microgravity experiment, nucleation rate and incubation time for nucleation of calcium carbonate from aqueous solution were measured as functions of supersaturation up to 6 minutes, and the nucleation rate under microgravity gravity was $10^4$ times slower than that under the terrestrial condition, as same as the result of the airplane experiment. Moreover, the incubation time for nucleation with low concentration followed the same function of supersaturation as that with high concentration. This result means that nucleation occurred from one of the following four solid phases: amorphous, vaterite, aragonite, or calcite. And it was derived from a slope of the function that the amorphous phase didn’t appear at the homogeneous nucleation which was realized under microgravity.

4. Conclusions

Four kinds of microgravity experiments using S-520 sounding rockets were successfully performed to investigate mechanism of crystal growth and nucleation. In the crystal growth experiments, heat and mass transport process were visualized and it became possible to evaluate the kinetics on the growing crystal surface quantitatively. In the nucleation experiments, nucleation rate under microgravity was drastically decreased compared with that under the terrestrial condition.

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