

YIN Zhigang, ZHANG Xingwang, WU Jinling, LI Xiaoya, YU Jianding, YUAN Zhangfu. Solidification and crystal growth on the SJ-10 recoverable scientific experiment satellite. *Chin. J. Space Sci.*, 2018, **38**(5): 836-838. DOI:10.11728/cjss2018.05.836

Solidification and Crystal Growth on the SJ-10 Recoverable Scientific Experiment Satellite

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Abstract The low-gravity environment aboard the space provides a unique platform for understanding crystal-growth-related phenomena that are masked by gravity on the Earth and for exploring new crystal growth techniques. We have characterized the wetting behavior of metal alloys and carried out melt growth of compound semiconductors under the support of materials science program in the SJ-10 recoverable satellite. We found that interfacial reaction plays a significant role in the interfacial evolution of Sn-based alloys. Detached growth of InAsSb was realized under microgravity, whereas during the terrestrial experiment the crystal and the crucible wall contact with each other. Moreover, the suppression of buoyancy-driven convection results in a more uniform composition distribution in the InGaSb and Bi₂Te₃-based semiconductor alloys.

Key words Solidification, Crystal growth, Wetting, Microgravity, Recoverable satellite

Classified index V524

The microgravity environment in space offers a new degree of freedom for materials processing. It provides possibilities to prepare perfect crystals with properties that cannot be obtained on the earth by eliminating the influence of gravity and allow profound understanding of fundamental phenomena that are masked by gravity and therefore are difficult to study quantitatively^[1-4]. The materials science program in the SJ-10 recoverable satellite, which is led by Prof. Zhang X W from the Institute of Semiconductors, Chinese Academy of Sciences, mainly focuses on the following issues: (1) the growth of semiconductor alloys with uniform composition distribution and low defect density, (2) understanding how the gravity-driven phenomena affect the crystal growth, (3) revealing the influence of microgravity on the wetting behaviors of molten metal alloys and clarifying the liquid/solid interfacial structure and its evolution during the solidification process. All the experiments

were carried out successively in the same multiple materials processing furnace with a gravity level of $10^{-4}\sim 10^{-5}g$ on the Chinese recovery satellite SJ-10 that was launched in 2016.

1 Wetting Behaviors and Interfacial Characteristics of Sn-Based Alloys

Due to the development of electronics manufacturing and welding technology, lead-free solder alloys have attracted great attention in recent years. Sn-based alloys are viewed as one of the most promising lead-free solder candidates owing to its excellent properties^[5]. However, the electric sparks caused by the formation of Sn whiskers under the high vacuum space environments can result in disastrous consequences and therefore have a close correlation with the safety issues^[6]. We designed experiments to investigate the

wetting behaviors and interfacial characteristics of Sn-based alloys on the Cu substrates and performed them at the SJ-10 recovery satellite.

We selected Sn-3.5Ag/Sn-17Bi-0.5Cu and Sn-3.5Ag/Sn-Sb as the model systems. By extracting the shapes and dimensions of the molten droplets, the contact angles and their relaxations were analyzed using the empirical formula. Such information is helpful for gaining better insight into the dynamical evolution of the three-phase line. We also modeled and analyzed the dependence of interfacial morphologies on the substrate inclination angle by using the finite-element method. Moreover, it was found that interfacial reactions play a key role in the spreading process. The detailed interfacial structures were studied to clarify the influence of interfacial reactions on the spreading behavior. Our work provides in-depth understandings on the interfacial microstructures of Sn-based alloys and offers opportunities for the development of environment-friendly materials with good solderability.

2 High-quality Bi₂Te₃-based Thermoelectric Semiconductor

Thermoelectric materials that can convert a temperature gradient into electric voltage, or vice versa, have attracted considerable attention due to their potential applications in solid-state cooling, temperature sensing and power generation [7]. To achieve an enhanced figure of merit, a thermoelectric material should have a high electric conductivity while possessing a low thermal conductivity. Bulk Bi₂Te₃ and its solid solutions are important thermoelectric systems, which act as a key role for the state-of-the-art thermoelectric devices operated at room temperature [8,9]. Herein, Bi₂Te₃-based thermoelectric alloy with a diameter of about 7.5 mm was grown by the zone melting method on the SJ-10 scientific platform. For comparison, Bi₂Te₃-based crystal with similar composition was also carried out under the same experimental condition and in the facility on the ground.

We have performed structural, compositional and thermoelectric analyses of the samples. In the X-ray diffraction curves, only the Bi₂Te₃-based alloy was resolved for both space and ground grown samples, and no impurity phase was observed. Moreover, the Full Width at Half-Maximum (FWHM) of the peak for the space-crystal is 0.190°, much lower than that (0.208°) of the ground-based sample. This result

suggests that the crystallization of space crystal is better than that of the ground sample. Furthermore, due to the suppression of buoyancy-driven convection, the composition homogeneity of the space crystal is improved as compared to that of the ground-based specimen. The thermoelectric measurements demonstrated that the space-grown sample has a good dimensionless figure of merit ZT.

3 Detached Growth of InAs_xSb_{1-x} Crystal

Long-wavelength infrared detectors that can operate at room-temperature have important applications in both civilian and military areas. Recently, InAs_xSb_{1-x} has attracted interest as a promising alternative to Hg_{1-x}Cd_xTe, the most prevalent material in high-performance long-wavelength infrared detectors at present [10–12]. The large, positive optical bowing effect due to the ordering in InAs_xSb_{1-x} making possible its applications in the long-wavelength infrared detection. In the SJ-10 project, we performed the InAs_xSb_{1-x} crystal growth by the Bridgman directional solidification. Taking into account the harsh mechanical environment during the launch, we designed an ampoule structure that can fix the charges along both the axial and radial directions. By heating the feed material in the quartz crucible above its melting point and then slowly cooling it from one end where the seed locates, the InAs_xSb_{1-x} crystal was progressively formed along the axial direction of the initial ingot.

During the ground-based InAs_xSb_{1-x} growth, the crystal adheres to the crucible wall which leads to non-ignorable thermal mismatch due to the difference in the thermal expansion coefficients of the crystal and the crucible material. In our case, the thermal expansion coefficient of quartz is about $5 \times 10^{-7} \text{ K}^{-1}$, an order of magnitude lower than that of InAs_xSb_{1-x}. Such a thermal mismatch results in a considerable defect (such as dislocation) density or even worse, macroscopic cracks in the grown crystals (or in the crucible). By sharp contrast, detached growth was observed during the InAs_xSb_{1-x} growth under microgravity, which is featured with a “bottleneck” near the seed/feed interface. In principle, the lack of direct contact between the grown crystal and the container crucible in the detached growth provides an effective way to reduce the defect density and therefore to improve the total crystal quality. Measurements indeed

demonstrated that the etched (dislocation) pit density exhibit a reduction of about 2 orders of magnitudes in the detached regions. Similar results were also observed for Ge and CdZnTe crystals prepared under μg condition^[13]. The reduction of dislocation density is a benefit for achieving higher carrier mobility and therefore has great implications for future long-wavelength infrared detector applications.

4 InGaSb Crystal with Homogeneous Composition Distribution

We also carried out the InGaSb growth experiment by the Vertical Gradient Freezing (VGF) method on SJ-10. The VGF method is a zone-melting method and the most important feature of VGF is that it has a saturated liquidus-zone during the growth process^[14,15]. Homogeneous semiconductor alloys can be grown using VGF by cooling the sample at a fixed cooling rate, on condition that the liquidus temperature at the growth interface keeps constant. The ingot is composed of three parts: the seed, the low temperature zone and the feed^[16]. The feed and seed (both are GaSb) were placed at the high and low temperature regions, respectively. Before growth, the system is heated to a temperature that turns the low temperature material (InSb) into the molten melt. Parts of the seed and feed are dissolved into the liquid zone and thus a solution forms. After that, the crystal growth begins by slowly decreasing the furnace temperature and the solid-liquid interface moves towards the feed.

High-quality $\text{In}_{0.11}\text{Ga}_{0.89}\text{Sb}$ crystal with a diameter of 9 mm and growth length of 6.5 mm was successfully grown on the SJ-10 recovery Satellite. No noticeable reaction between the molten InGaSb and the BN crucible was observed, revealing that the ampoule designing satisfies the microgravity growth of InGaSb. Typically, our electron probing micro-analyzer measurements show that the chemical inhomogeneity is lower than 1% within the whole growth area, both along the axial and radial directions. These results are similar with those obtained at the International Space Station^[14–16] and are much better than those achieved in the experiments under 1 g condition. Our observations have implications for future thermoelectric applications due to the tunable bandgap and good optoelectric properties of InGaSb.

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