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Materials Experiment on Tiangong-2 Space Laboratory

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Abstract During the China's Tiangong-2 (TG-2) flight mission, the experiments of 18 kinds of material samples were conducted in space by using a Multiple Materials Processing Furnace (MMPF) mounted in the orbital module of the TG-2 space laboratory. After the experiments of 12 kinds of samples of the first and second batches were completed successfully, astronauts packed and brought them back to the ground by Shenzhou-Ⅱ spacecraft. By studying processing and formation on semiconductor and optoelectronics materials, metal alloys and metastable materials, functional single-crystal, micro- and nano-composite materials encapsulated in sample ampoules both in space and on Earth, we expect to explore some physical and chemical processes and mechanism of the materials formation that are normally obscured and therefore are difficult to study quantitatively on the ground due to the gravity-induced convection, to obtain the processing and synthesis technology for preparing high quality materials, and lead to the improvement and development of materials processing techniques on Earth, and also develop the experiment device and comprehensive ability for materials experiment in microgravity environment. This report briefly introduces the main points of each research work and preliminary comparative analysis results of 12 samples carried out by scientists undertaking research task.

Key words Solidification, Crystal growth, Wetting, Microgravity, Tiangong-2 space laboratory

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By experimental study of processing and formation on semiconductor and optoelectronics materials, metal alloys and metastable materials, functional single-crystal, micro- and nano-composite materials onboard Tiangong-2 (TG-2) space laboratory, we expect to reveal some physical and chemical processes and mechanism of the materials formation that are normally obscured and therefore are difficult to study quantitatively on the ground due to the gravity-induced convection, to obtain the processing and synthesis technologies for preparing high quality materials, and lead to the improvement and deve-

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lopment of materials processing techniques on Earth. Based on the space experiments during the 2nd step of Chinese manned space flight, we also developed the experiment device and comprehensive ability for materials experiment in microgravity environment and astronauts participated in space materials experiment onboard space laboratory the first time. China's space materials science experiment level can be obviously enhanced through the execution of TG-2 flight mission.

On 15 September 2016, a Multiple Materials Processing Furnace (MMPF) mounted in the orbital module of the TG-2 space laboratory was carried into space. The materials science experiment was carried out after completing the function and performance tests on the furnace. The experiments of 18 kinds of material samples encapsulated in ampoules, which were divided into three batches, were conducted in space, and the astronauts performed in-orbit replacement of the experiment samples of the three batches. The microgravity level during experiments is better than $10^{-3}\,g$. After the experiments of 12 kinds of samples of the first and second batches were completed successfully, astronauts packed and brought them back to the ground by Shenzhou-XI. The thermal behavior measurements for MMPF with another 6 unrecoverable samples were conducted after the astronauts came back to Earth. The comparative analyses and studies on the samples from the space and ground experiments are carried out by scientists undertaking research task in their own laboratory.

1 Preparation of New Type of Metal Matrix Composites in Space

The formation, solidification, wettability and atomic interaction of the new composite are investigated under microgravity. The experiment is expected to clarify the effects of microgravity on the structure and properties of the new composites-SiC/Zr-based alloy composite. The underlying mechanisms of gravity in the solidification process would be proposed, which will lead to the development of technique and the improvement of properties of composites on Earth. It will promote the progress of materials science and the application of the new composites.

The experimental specimens are divided into two groups. One group is composite consisting of Zr-based alloy matrix and SiC particles reinforcement. The size of the cylindrical composite is $\phi 5\,\text{mm}\times 25\,\text{mm}$. The other one is wetting specimens between Zr-based alloy and SiC substrate. The size of the substrate is $\phi 8\,\text{mm}\times 1\,\text{mm}$. The samples are separated using BN slice. The two groups of samples were placed into BN crucible. Then BN crucible is sealed using vacuum quartz ampoule.

The experiment has been carried out using MMPF in space. The sample ampoule was moved to isothermal zone as soon as possible, holding 60 min at 880°C, then the power was switched off. After the temperature decreased to 300°C, the sample ampoule was taken out from the furnace.

The analyses of structures, composition, macro- and microstructural morphologies and phase distribution on samples are performed by using various techniques and devices. The main results of the experiment show that the precursor alloy films with a width of over 60 μm from space samples were formed between SiC substrate and Zr-based alloy in all wetting samples only in microgravity, and the films appear on the lateral side of the solid-liquid-gas interface; the interfacial and microstructures of wetting samples from space experiment are also different from that on the ground; and the more homogenous distribution of reinforced phases of SiC particles in Zr-based alloy matrix were obtained in space samples.

This kind of new metal matrix composite has important applications in defense equipment, and Hofmann and Roberts are going to carry out similar experiments on the International Space Station in the future[1].

2 Study on the Mechanism of Ferroelectric Thin Film Epitaxial Growth

The high quality epitaxial ferroelectric thin film is the key to the development of large scale focal plane infrared detector. $PbZrTiO_3$ (PZT) is a typical ferroelectric material due to its adjustable thermal, dielectric and pyroelectric properties. However, the film growth mechanism of epitaxial PZT thin films by sol-gel technology has not been fully understood.

In the synthesis of PZT thin film, the formation and growth of thin film involve many steps and processes, such as diffusions of Pb, Zr, Ti and other elements, combined to complete the film nucleation and growth with the producing of H_2O and CO_2. In the ground environment, the existence of various factors such as gravity induced convective flows and sedimentation makes it difficult to explore the growth mechanism. Thus, we can attempt to explore the formation of PZT thin film by a sol-gel method in microgravity, and to study the physical and chemical processes of nucleation and growth of PZT thin film by diffusion of elements of Pb, Zr and Ti on substrate accompanying the releases of H_2O and CO_2 which is a possible factor for microscopic convection on Earth. The results will help to reveal the epitaxial growth mechanism of thin film formed by the sol-gel method and optimize conditions for thin-film epitaxy growth on the ground.

The $PbZrTiO_3$ precursor thin films of about 75 nm thickness containing Zr, Ti, and Pb composition were fabricated by means of sol-gel spin coating method on LNO/Si substrates. The pretreatment of the film was carried out at 200°C and 380°C for 5 min, respectively. A group of specimen slices with 5 mm×5 mm were cut from a large silicon substrate with the precursor film and were sealed in quartz ampoule filled with a 0.1 MPa of O_2, and each slice was separated using quartz ring. Space experiment was performed using MMPF. The ampoule was heated to 500°C and held at this temperature for 120 min, and then cooled to 200°C with a given cooling rate and the power was cut off after this.

By observing the microstructural morphologies, analyzing the structure and relative properties of specimens from space and ground, the significant result of the experiment is that the polycrystalline growth is significantly suppressed and the preferential orientation growth is obtained in the microgravity environment, although there are no obvious differences for SEM morphologies on the surface of epitaxial growth films in both cases. This is the first time to carry out the exploring experiment of epitaxial growth of the PZT thin films with important application value.

3 Crystal Growth of ZnTe: Cu Crystal in Microgravity

The ZnTe: Cu crystal, an important Ⅱ-Ⅵ compound semiconductor, has the direct band-gap of 2.2 eV at room temperature. Having a large 2nd order nonlinear susceptibility and electro-optic coefficient, the crystal is one of the best electro-optic crystal candidates for the detection and generation of the THz wave. ZnTe: Cu crystal can also be used in other areas for the fabrication of solar cells, green light emitters, waveguides, and modulators. There are many advantages to the growth of ZnTe: Cu crystal in microgravity. The convection induced by gravity can be eliminated in the microgravity conditions, and it is possible to obtain a more perfect crystal with uniform component and dopant distribution, and very low defect densities. In this experiment, the ZnTe: Cu crystal was grown by a zone-melting technique in MMPF. ZnTe: Cu alloy and Te (as solvent) was loaded into quartz crucible sealed in quartz ampoule under vacuum ($< 10^{-3}$ Pa). The ampoule was heated up to 800°C and kept for 160 min, and then crystallization from the position of contact between ZnTe: Cu alloy and Te melt begun by moving ampoule a rate of 0.5 mm·h^{-1} for 90 h isothermally. After finishing the crystal growth the ampoule was moved out furnace when its temperature was decreased to room temperature.

The investigation on the crystal morphologies, structure and optical properties of grown ZnTe: Cu crystals by various characterization methods shows that an orange ZnTe single crystal (its size is about 10 mm×6 mm×2 mm) was obtained in the space sample. It is found that the crystal size of space sample is larger than that of the ground sample. Because of the capillary phenomenon in microgravity, Te and ZnTe film were grown at the surface of the silicon rod of the space sample. However, a little bit of gaseous product was found at the ground sample. This result underlines the importance of microgravity for crystal growth of Group Ⅱ-Ⅵ semiconductor material. It gives the positive guidance on the growth of infrared semiconductor materials on the ground such as HgCdTe, CdZnTe and ZnTe crystals.

4 Growth of Opto-functional Crystals in Outer Space

Functional Crystals play quite important roles in various instruments and equipment. However, composition segregation exists in some of the crystals grown on Earth, which is seriously impact the properties and utilization rate of the materials, such as $Pb(In_{1/2}Nb_{1/3})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ relaxor ferroelectric single crystal[2], $K_{1/2}Na_{1/2}NbO_3$ lead-free piezoelectric single crystal[3], Ti^{3+} doped CsI scintillation crystal[4] and so on. Therefore, it is quite necessary to explore effective ways to grow these crystals with low or no segregation in order to improve or enhance the properties of crystals. Gravity is one of the main factors that can influence the segregation[4]. CsI-based crystals are very important opto-functional materials. CsI crystal is a traditional and widely used alkali metal halide inorganic scintillator. In this study, we grew the Eu^{2+} doped CsI crystal in microgravity and studied the effect of microgravity on segregation.

The crystal ingots with the composition of Eu^{2+} doped CsI with a size of ϕ9 mm×70 mm were prepared in the ground laboratory, and then sealed in a platinum crucible. After this, the platinum crucible was sealed in a quartz ampoule with a low vacuum of 1 Pa pressure. The crystal growth experiment was conducted in MMPF. The quartz ampoule containing Eu^{2+} doped CsI crystal ingot was firstly heated to 780°C and kept for 2 h isothermally, and thereafter ampoule was moved at a speed of 2.16 mm·h^{-1} until the growth of crystal was completed.

The quality and the uniformity of the crystals grown in microgravity and 1 g were analyzed by optical, structural and property characterization. The results show that the crystal is transparent with the dimension of ϕ9.8 mm×51 mm except for the bottom in space sample, the wettability of CsI (Eu) melt and the platinum crucible is good, and the number of traps in the space sample is smaller than that in the ground sample and the Eu segregation in the space sample is lower than that in the ground sample.

5 Solidification of Immiscible Alloy in Space

The solidification of immiscible alloy, the precipitation of the minority phase droplets/particles and the formation mechanism of micro/macrosegregation attract great attention of material scientists because they are the key issues in the manufacturing of the in-situ particle composite materials with high performances[5−9]. This work investigates the solidification behavior of Al-Sn-Bi alloy in space and on Earth. The study contents and objectives of the work include by directional solidification experiments with Al-Sn-Bi alloys on Earth under general conditions or under the effect of static magnetic field, investigating the precipitation of droplets/particles in the melt, the phase segregation behavior during solidification and the formation of the in-situ particle composite, deepening our understandings of solidification microstructure formation mechanisms of immiscible alloy and the effect of gravity, and building models describing the microstructure evolution in directionally solidified Al-Sn-Bi immiscible alloys and promoting the development of the industrial manufacturing methods for the in-situ particle composite materials with high performances

The Al-Sn-Bi alloy ingot with a size of ϕ 6.9 mm ×57 mm was loaded in a BN crucible sealed in a quartz ampoule with a vacuum better than 10^{-3} Pa. The remelt and solidification of the alloy were conducted in the MMPF. The quartz ampoule containing alloy ingot was firstly heated to 700°C and kept for 4 h isothermally, and then ampoule was moved with a speed of 100 mm·h^{-1} until the end of solidification.

The analysis of the solidified samples of Al-Sn-Bi immiscible alloy presents that there is a more uniform and dispersed solidification structure in space samples. A model was developed to describe the liquid-liquid phase transformation during solidification. The microstructure formation process was calculated and the microgravity effects were analyzed. The results demonstrate that the microgravity affects the microstructure formation mainly through weakening the convective flow of the matrix liquid and diminishing the Stokes settlement of the Minority Phase Droplets (MPDs). All these are favorable for the obtaining the immiscible alloy with a well-dispersed microstructure. This result is of significant value in promoting the development and application of the ground preparation process of immiscible alloy in-situ composites.

6 Solidification of Multicomponent and Multiphase Alloys in Space

The experiment is to investigate the undercooling and solidification process of multicomponent and multiphase alloys under space environment. The thermodynamic and kinetic characteristics of rapid solidification are specified in terms of various conditions of gravity levels, container states, and physical fields, through a systematic comparison between the space experiments aboard the TG-2 space laboratory and the ground experiments performed via different methods. The specimens of Ag-Cu-Ge and Ag-Cu-Sb ternary eutectic alloys were enclosed in quartz crucibles together with a portion of B_2O_3 particles as a denucleating agent, and quartz crucibles also were encapsulated in a quartz ampoule. The alloy specimens are subjected to solidification under the covering of a thin layer of B_2O_3 melts in microgravity condition. High undercooling and rapid solidification of these two alloys are expected to realize in the space environment. A deep understanding is also expected to come to those phenomena occurred during ternary eutectic solidification, such as the competitive nucleation and cooperative growth among three solid phases, the transitions from faceted phase to non-faceted phase, and the transitions from lamellar eutectics to anomalous eutectics.

The two ampoules were heated to 840°C and 700°C respectively and both kept for 1 h isothermally in MMPF onboard the TG-2 space laboratory, and then powers were switched off and ampoules were cooled in-situ to room temperature.

The space solidification of AgCuGe and AgCuSb, indicate that the microgravity conditions can change the distribution of primary phases. For example, the coarse Ge phase is distributed in the middle of the AgCuGe alloy sample, and Sb phase appears inside the AgCuSb alloy sample. Whereas on the ground, the two kinds of phases are located on the top and the surface of the samples, respectively. Microgravity conditions also suppress the separation tendency of the two leading growth phases from each other, which shows a relevance of the two phases. The results are of instructive significance for studying competitive nucleation, cooperative growth mechanism and modulating microstructure and performance of multiphase alloys.

7 Solidification of Al-based Single Crystal Alloy in Space

Taking an AlZnMgCu alloy, which has high similarity in solidification behavior and microstructure with Ni-based single crystal superalloy, as a model material, investigates the effects of gravity on the solidification microstructure of this single crystal alloy. This research analyzes the effect of gravity on the dendrite morphology, and element distribution in multicomponent single crystal alloy, and further, the related mechanisms. The aim is to deepen the understanding of alloy solidification and defect formation mechanism, so as to provide more accurate guidance in avoiding solidification defects of single crystal alloy, and therefore, improve its quality and performance.

The Bridgman method was used for directional solidification experiment. A single crystal sample of about 7 mm in diameter and 90 mm in length was encapsulated in a quartz ampoule under vacuum. When doing the experiment, the ampoule was put at a certain place in the furnace, which permits only partial remelting of the sample, was heated the furnace up to about 750°C, and then let the ampoule move directionally out of the furnace at a rate of 20 mm·h^{-1} for 80 mm. In this way, epitaxial growth from the unmelted seed crystal was realized. For the purpose of comparison, the same experiments were conducted both in space and on the ground.

The results exhibit that epitaxial dendrite growth took place after partial remelting of the samples, the whole samples are composed of seed crystal, initial semi-molten zone, columnar dendrite growth region and equiaxed polycrystal region at the end, the presence of equiaxed grains at the end of samples indicates columnar-to-equiaxed transition happened in the final phase of solidification. the growth morphologies of columnar dendrites and equiaxed grains are different in space sample and ground sample, By contrast, columnar dendrites with fewer high order arms and less obvious cross feather, and grains with more amount and smaller elongations were present in space sample. In addition, micro- and macroscopic segregation of elements occurred in both space and ground samples, however, the extent of segregation, especially along the radial direction, are slighter and the element distributions are more uniform in space sample. Among the solute elements, the segregation of Cu is related to gravity level the most closely, while that of Mg is almost unaffected by gravity. In conclusion, the space environment makes the distribution of alloy compositions more uniform, which would be conducive to reducing defects and improving material properties.

8 Space Preparation and Thermoelectric Properties of High Performance Thermoelectric Semiconductor Crystals

Bismuth telluride is a class of commercial materials used for thermoelectric cooling and power generation. Their thermoelectric figure of merit, the ZT, is around unit. Intensive studies have been carried out to enhance the ZT. Alloying and doping are effective ways to improve the ZT of bismuth telluride. However, it is difficult to get a compositional uniform and high performance multicomponent bismuth telluride crystals on earth ground gravity condition. Space microgravity may provide a favorable melt solidification condition for us to obtain compositional uniform and high performance crystals. Our project is to carry out a multicomponent bismuth telluride (Bi_2Te_3-Sb_2Te_3-Te) crystal growth experiment under the micro-gravity condition on TG-2 space laboratory. By the investigation it is expected to get understanding of the solute transport mechanism during multicomponent crystal growth, to reveal the effect of different components on the properties of the thermal and electrical synergistic transport, to obtain compositional uniform and high performance p-type bismuth telluride thermoelectric material which provides scientific evidence to greatly enhance thermoelectric properties and conversion efficiency of thermoelectric materials.

A multicomponent bismuth telluride ingot with a size of $\phi 8\,\mathrm{mm}\times 55\,\mathrm{mm}$ was synthesized and enclosed inside an evacuated silica ampoule. The sample ampoule was loaded into MMPF and heated to 660°C and kept for 60 min isothermally. After this, the ampoule was moved at a speed of $35\,\mathrm{mm{\cdot}h^{-1}}$ to furnace bottom during the period of 540 min and then cooled to room temperature with given cooling rates.

The comparative analysis of bismuth telluride crystals grown in space and o the ground shows that the uniformity of the axial and radial composition and crystallinity of space samples are better than those of ground samples. The uniformity of thermal conductivity and power factors are better than those on the ground. A deep understanding of the composition, structure, and properties of bismuth telluride based samples in space and on the ground will be helpful to direct the preparation of high quality bismuth-based telluride crystals on the ground.

9 Space Growth of Te- and Mn-doped Semiconductor InSb Crystals

The microgravity environment provides a unique platform to synthesize alloy semiconductors with homogeneous composition distributions, on both macroscopic and microscopic scales, due to the great reduction of buoyancy-driven convection. Moreover, the easy realization of detached solidification in microgravity decreases the formation of defects such as dislocations and twins, and thereby the crystallographic perfection is greatly increased[10,11]. Motivated by these facts, this project aims to growth Te-doped and Mn-doped InSb single crystal alloys with very low defect densities and uniform distribution of composition, the former crystal is an infrared semiconductor and the later one is a diluted magnetic semiconductor. These experiments are expected to provide new insights into the crystal growth mechanism and improve the terrestrial high quality crystal preparation.

The seeding and the feeding materials, with a diameter of 7.5 mm, were encapsulated into a quartz crucible that was covered by graphite caps. A modified Bridgman method was used to grow the Te-doped and Mn-doped InSb crystals. The temperature gradient, ampoule structure and growth rate were carefully designed to allow to grow more perfect single crystals in multiple materials processing furnace.

The observations on the surface of grown InSb-based semiconductor crystals doped with Te and Mn respectively show the appearance of the detached growth of crystals in space. The concentration of defect in the crystal is obviously reduced, and the macroscopic distribution of doped elements along the radial direction is more uniform, and the Hall carrier mobility in space Mn-doped InSb sample is also increased. These results are of great significance in understanding the specific crystal growth mechanism in the microgravity environment, and in studying the mechanism of defect formation and its effective means of regulation. At the same time, it also provides an important guide for the development of high-quality infrared functional crystals.

10 Synthesis of Mesostructured Nanocomposites under the Microgravity Conditions

About nanomaterials, particles size control and particles agglomeration/aggregation are the two major concerns. As an example, nanosized gold-containing composites showed increased optical nonlinearity resulting from the enhancement surface plasmon resonance effect of the metal nanoparticles. To improve the structural stability of nanocomposites, the followed thermal treatment had been conducted. Because nanosized metals would melt at a much lower temperature compared with that of the bulk materials (*e.g.*, the melting temperature of 2 nm Au particle and the bulk counterparts are 600 K and 1337 K, respectively), unfortunately, the original homogeneity of nanoparticles size distributions were destroyed and nanoparticle size increased significantly resulting from the gravity-induced particle aggregation. As a consequence, its optical properties deteriorated. Microgravity conditions in the spacelab offer a special circumstance, in which the gravity effect would be greatly lessened and thermally treated nanocomposites with uniform nanoparticle size and high nonlinear optical response are thus expected. Till now, the related reports about nanocomposites annealed under microgravity conditions and their properties are scarce.

In this project, gold-containing nanocomposites (Au/SiO_2) were firstly synthesized on the ground and sealed in an ampoule. Then, the sample was loaded in the multiple materials processing furnace. The thermal treatment process was finished in the TG-2 under the control of pre-loaded programs. The materials processing temperature and duration were 820°C and 3 h, respectively. During the whole thermal treatment, the sample ampoule was kept statically. After isothermal process ended, the ampoule was cooled to room temperature in-situ.

The microstructures of Au/SiO_2 nanocomposites thermally treated in space and on the ground were analyzed. The experiment results showed that the similar nanoparticle aggregation behaviors and the approximately nonlinear optical response were obtained. Interestingly, according to the idea that microgravity would suppress the nanoparticle aggregation during high temperature thermal treatment[12], in a ground work it was found that the aggregation of gold nanoparticles can be alleviated by the applied magnetic field, and thus the enhanced optical nonlinear response of the resultant materials had been demonstrated. The results are of great importance for the structural design and preparation of new nanomaterials.

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