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Materials Experiment on 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 Tian-Gong-2 (TG2) 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, to obtain the processing and synthesis technologies for preparing high quality materials, and direct the improvement and development of materials processing techniques on the ground. Based on the space experiments during the 2nd stage 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 TG2 flight mission.

On 15 September 2016, a Multiple Materials Processing Furnace (MMPF) mounted in the orbital module of the TG space laboratory was carried into space. The material 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 higher 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 the 12 kinds of samples 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. The main points of each research task and preliminary analysis results are briefly introduced as follows.

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 on solidification process would be proposed, which will be used to direct the development of technique and the improvement of properties under the condition of the earth. It will promote the progress of materials science and 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 Φ5×25 mm. The other one is wetting specimens between Zr-based alloy and SiC substrate. The size of the substrate is Φ8×1 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 cut off. When the temperature decreases to 350°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 study 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.

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 optical properties. However, the film growth mechanism of epitaxial PZT thin films by sol-gel technology has been not fully understood.

In the synthesis of PZT thin film, the formation and growth of thin film crystal 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 ground environments, due to factors such as gravity and convection, makes it difficult to the exploration of film growth mechanism due to the existence of micro-convection as a possible factor on the earth. 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 PZT thin film nucleation and growth by diffusion of elements of Pb, Zr and Ti on substrate accompanying the releases of H_2O and CO_2. The results will help to reveal the epitaxial growth mechanism of a thin film formed by sol-gel method, and direct optimization conditions for thin-film epitaxy growth on the ground.

The $PbZrTiO_3$ precursor thin film of about 75 nm thickness containing Zr, Ti and Pb composition was 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 5×5 mm^2 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 given cooling rate and the power was cut off.

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 growth of a specific orientation direction of epitaxial growth on the substrate under gravity is significantly restrained in the microgravity environment although no obvious differences for SEM morphologies of epitaxial growth and characteristic relative properties are found between space and ground samples. This is the first time to carry out the exploring experiment of epitaxial growth of the PZT thin film material 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. The growth of ZnTe: Cu crystal in space has many advantages. The convection induced by gravity can be eliminated in the microgravity conditions. It is possible to get materials with uniform component, good surface, and integrated structure with fewer defects. 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 graphite crucible sealed in quartz ampoule under vacuum($<10^{-3}$ Pa). The ampoule was heated up to 800°C and kept for 160 mins, 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 for 90 hours isothermally. After finishing the crystal growth the ampoule temperature was decreased to room temperature with given cooling rates.

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 very important roles in various instruments and equipment. However, segregation exists in some of the crystals, which is seriously impact the properties and utilization rate of the materials, such as $Pb(In_{1/2}Nb_{1/3})O_3$- $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$, $K_{1/2}Na_{1/2}NbO_3$, Tl^{3+} doped CsI 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. The CsI-based crystals are very important opto-functional crystals, and CsI crystal is a traditional and widely used alkali metal halide inorganic scintillator. For the growth of the crystals most investigations focus on the continuous adding of raw materials, and the growth processes are very complex. Until now, there isn't any report about the influence of microgravity on composition segregation. In this study, we grew the Eu^{2+} doped CsI crystal in microgravity and studied the effect of microgravity on segregation. The Eu^{2+} doped alkali halides have wide practical applications such as X-ray storage phosphors and solar ultraviolet-B detectors.

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 experiment of crystal growth was conducted in MMPF. The quartz ampoule containing Eu^{2+} doped CsI crystal ingot was firstly heated to 780°C and kept for 2 hrs isothermally, and then ampoule was moved with a speed of 2.16 mm/h to the end of crystal growth.

The quality and the uniformity of the crystals grown in microgravity and 1g was analyzed by optical, structural and property characterization. The results show that the crystal is transparent with the dimension of F9.8 mm′51 mm except for the bottom in space sample. The wettability of CsI (Eu) melt and the platinum crucible is good. The number of traps in the space sample is smaller than that in the ground sample and the segregation of Eu in the space sample is lower than that in the ground sample.

5. Solidification of Al-Sn-Bi Alloy in Space

The solidification interface stability, the formation mechanism of micro/macrosegregation and the precipitation of the minority phase droplets/particles attract great attentions of material scientists because they are the key issues in the manufacturing of alloys with high performances, *e.g.* single crystal alloy and the in-situ particle composite materials, etc. This work investigates the solidification behavior of Al-Sn-Bi alloy on the earth and in space. The study contents and objectives of the work include: by directional solidification experiments with Al-Sn-Bi alloys on the earth under general conditions or under the effect of static magnetic field, deepen our understandings on the stability of the solid/liquid interface, the dendrite growth and the effect of gravity or convective flow; investigate the precipitation of droplets/particles in the melt, the phase segregation behavior during solidification and the formation of the in-situ particle composite, and develop models describing the microstructure evolution in directionally solidified Al-Sn-Bi alloys; develop industry manufacturing methods for alloys with high performances, *e.g*. single crystal alloy and the in-situ particle composite materials *etc*.

The Al-Sn-Bi alloy ingot with a size of ϕ7 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 hrs isothermally, and then ampoule was moved with a speed of 100 mm/h to the end of solidification.

The analysis of the solidified samples of Al-Sn Bi monotectic alloy presents that there is a more uniform and dispersed solidification structure in space samples. This reveals that the Marangoni convection can be suppressed by adding active elements to reduce the interphase interfacial energy. Thus, the second phase dispersion distribution is obtained in space. This result is of significant value in directing the development and application of the ground preparation process of monotectic alloy in-situ composites.

6. Solidification of Multi-component and Multiphase Alloys in Space

The scientific goal of this project is to investigate the undercooling and solidification process of multi-component 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 TG2 space laboratory and the ground experiments performed by different methods. The specimens of Ag-Cu-Ge and Ag-Cu-Sb ternary alloys were enclosed in quartz crucibles together with a portion of B_2O_3 particles as the decontaminating 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 hr isothermally in MMPF onboard the TG2 space laboratory, and then powers were switched off and ampoules were cooled in-situ to room temperature.

The preliminary analyses on solidification experiments of two ternary eutectic alloys, AgCuGe and AgCuSb, indicate that the microgravity conditions can change the distribution of primary phases. For example, the larger size Ge phase is distributed in the middle of the AgCuGe alloy sample, and Sb phase appears inside the AgCuSb alloy sample, while 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 each other of the two leading growth phases, which showing a relevance of the two phases. It has important guiding value for studying nucleation and cooperative growth mechanism of multiphase competition and for effectively designing and controlling the microstructure and macroscopic properties of multiphase alloys.

7. Solidification of Al-based Single Crystal Alloy in Space

Taking an Al-based 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, element distribution, and defect (porosity, mixed crystal and freckle) formation, and further, the related mechanisms, especially the effect of convection. Clarify the relationship between gravity and solidification defects by studying space growth of single crystal alloy. Research results will be used to improve the production of single crystal alloys on the ground.

The Bridgman method was used for directional solidification experiment. A sample of about 6—8 mm in diameter was encapsulated in a quartz ampoule under vacuum. When doing an experiment, heat the furnace up to about 800°C, and then let the ampoule move directionally in the furnace at some rate and realize the single crystal growth from the seed crystal.

The study on the growth of the model alloy in space and on the ground exhibits that the dendritic to equiaxed crystal transition occurs at the end of the samples grown by seed epitaxial growth, but the epitaxial growth morphologies of columnar dendrites and terminal equiaxed crystals are different. The higher secondary dendrites of columnar dendrites in space sample are less than those in ground ones, and the inter-dendritic structures are thicker. In addition, the micro- and macroscopic segregation of elements in space samples is lower, and the distribution of elements is more uniform. The weak convection in microgravity makes the distribution of alloy elements more uniform during the growth process, and thus changes the dendrite morphology, which is helpful to reduce solidification defects. The results are of guiding significance for improving the properties of single crystal superalloys.

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 multi-component 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 samples. Our project is to carry out a multi-component bismuth telluride (Bi_2Te_3-Sb_2Te_3-Te) crystal growth experiment under the micro-gravity condition on TG2 space laboratory. By the investigation it is expected to get understanding of the law of solute transport during multi-component 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, and to provide scientific evidence to greatly enhance thermoelectric properties and conversion efficiency of thermoelectric materials.

A multi-component bismuth telluride ingot with a size of $\phi 8$ mm × 55 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 mins isothermally. After this ampoule was moved with a speed of 35 mm/h to furnace bottom during the period of 540 mins, and then cooled to room temperature with given cooling rates.

The comparative analysis of bismuth telluride crystals grown in space and 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 suppresses the formation of defects such as dislocations and twins, and thereby the crystallographic perfection is greatly increased. Motivated by these facts, this project aims to synthesize 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 near diluted magnetic semiconductors. These experimental results are expected to provide new insights into the crystal growth mechanism and direct the terrestrial high quality crystal preparation.

The seeding and the feeding materials, with a diameter of ~8 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 homogeneous 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 phenomena and laws in 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 other functional crystals.

10. Synthesis of Mesostructured Nanocomposites under the Microgravity Conditions

Owing to the characteristic pore structures of ordered mesoporous materials, its discovery not only has a great influence in the traditional industrial fields, such as adsorption/desorption, separation, and catalysis *et al* but also opens up a new opportunity for the booming nanomaterials research. In the latter case, particles size control and particles agglomeration/aggregation are the two major concerns. As "a natural micro-reactor" for nanomaterials preparation, the large surface area and uniform pore dimension of ordered mesoporous materials would benefit the good dispersion of nanoparticles with controllable sizes, which satisfy the prerequisite for the relationship research between nanomaterials features (*i.e.* particle sizes, size distribution, particles spatial dispersion) and their properties. As an example, gold-incorporated mesostructured materials showed increased optical nonlinearity resulting from the NonLinear Optical (NLO) enhancement of the Surface Plasmon Resonance (SPR) of the metal nanoparticles and the nonlinearities of electrons in the metal nanoparticles. To improve the structural stability of mesoporous frameworks, the followed thermal treatment had been conducted. Because nanosized noble 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 on are 600 K and 1337 K, respectively), during the thermal treatment, 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. Therefore, stable nanocomposites experienced thermal treatment under microgravity conditions was expected, in which homogeneously distributed nanoparticle with uniform sizes and high NLO response would be obtained. Till now, no report has been found about the structural evolution of mesostructured-based nanocomposites annealed under microgravity conditions and also its influence on materials properties.

In this project, gold-containing mesostructured nanocomposites (Au/SiO_2) were firstly be synthesized under normal conditions and then sealed in an ampoule. Then, the sample was loaded in the multiple materials processing furnace. The thermal treatment process was finished in the Spacelab under the control of pre-loaded programs. The designed 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 microstructure of Au/SiO_2 nanocomposites processed at high temperature in space and on the ground was observed and analyzed. In space samples, the microstructure of Au particles in the SiO_2 matrix is obviously different from that of ground samples. However, the third order nonlinear optical response properties calculated for space samples are similar to those on the ground. According to the idea that microgravity in space would suppress the aggregation of gold nanoparticles during densification at high temperature. In our early work we found that the aggregation of gold nanoparticles can be controlled by the applied magnetic field on the ground, and thus the improvement of the third order optical nonlinear polarization response of this kind of composite has been demonstrated in our previous work. Mesoporous nanocomposites have important potential applications in the field of optical communication and optical computing. The results are of great scientific and practical value for the structural design, preparation and performance control of these novel materials.