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Formation of Graded Structure in Se-Te Solid Solution prepared by an Ultra-Strong Gravitational Field

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The mega-gravity field experiment was performed on the all-proportion miscible Se-Te system (70:30 in mol%) semiconductor by a new ultracentrifuge apparatus in JAERI. The specimen was prepared under an ultra-strong gravitational field of 1,020,000 G in maximum acceleration at a temperature of 260℃ for 100 hours, which was below the melting point of the starting sample. The Te content increased in the direction of gravity from about 0 to 60 at.%, while the Se content decreased from about 100 to 40 at.%. The crystal growth along the direction of gravity was observed. The large composition gradient was observed even in the large grown single crystals. This showed that the graded structure was continuous in atomic scale and caused by the sedimentation of substitutional atoms.
Key words: Ultra-strong gravitational field, Atom sedimentation, Atomic-scale gradient, Crystal growth

1. INTRODUCTION

Materials science research under a very strong gravitational field of up to higher than 1,000,000 G (1G=9.8 m/s^2) has now remained as an unexploited field. Unlike under high pressures, under a gravitational field, each atom is displaced by one-dimensional body force, and as a result, a unique molecular-crystal state or sedimentation of atoms can be realized. Such strong gravitational field has direct effects in atomic scale such as sedimentation or structure change [1]. It is suggested that the sedimentation of atoms will be used as a new materials processing, to concentrate elements or even isotopes, and to form an atomic-scale graded structure or nano-mezo composite structure in condensed matter, and so on.

To study the sedimentation of atoms or crystal-chemical instability in solids under a strong gravitational field, we developed two ultracentrifuge apparatuses in Kumamoto University [2] and Japan Atomic Energy Research Institute (JAERI) [3], which can generate an acceleration of over 1 million G for a long time at high temperature. The new developed ultracentrifuge apparatus in JAERI can generate a larger potential energy of over two times and a wider temperature range up to over 500 ℃ compare with that in Kumamoto University [3]. Up to now, we achieved the atomic-scale graded structures in an all-proportion miscible alloy in bismuth (Bi)-antimony (Sb) (70:30 in mol%) system [4, 5] and an intermediate-phase alloy in indium (In)-lead (Pb) system [6] by the sedimentation of substitutional solute atoms.

The selenium (Se)-tellurium (Te) system is an all-proportion miscible system [7], and the solid solution shows semiconductor property. In this study, we performed the ultra-strong gravitational field experiment on the Se-Te (70:30 in mol%) solid solution, to investigate the sedimentation of atoms and to form an atomic scale graded structure with a graded band gap structure.

2. EXPERIMENTAL PROCEDURE

The Se-Te (70:30 in mol%) alloy lump was prepared by melting pure Se and pure Te shots in a Pyrex test tube at about 640℃ for 30 minutes under an argon atmosphere. The purities of Se and Te shots were 99.99%. The lump was crushed to a powder and was melted again at about 430℃ under vacuum in a hollow glass rod with an inner diameter of 4 mm for several minutes. The uniformity of the specimen was confirmed by the EPMA analysis. The specimen was cut to be a column with a height of about 5 mm, and was set into a SUS304 capsule with an inner diameter of 4 mm. The sample capsule was fixed into a titanium rotor with an outside diameter of 80 mm.

The ultracentrifuge experiment was performed by the new developed ultracentrifuge apparatus in JAERI [3], which can generate an ultra-strong gravitational field even larger than 1 million G over a wide temperature range up to over 500℃ with a high stability control. Figure 1 shows the schematic of the layout of the ultracentrifuge apparatus and the photograph of the titanium alloy rotor and the sample capsules (see the right

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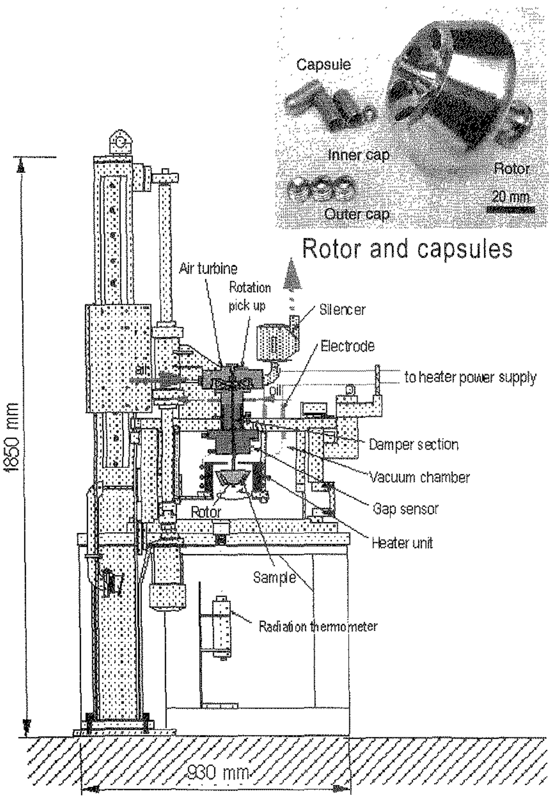

Fig. 1 Schematic of the layout of the ultracentrifuge apparatus and the photograph of the titanium alloy rotor and the sample capsules.

upper part of Fig. 1). For the Se-Te system (70:30 in mol%) solid solution, the ultracentrifuge experiment was performed at a rotation rate of 160,000 rpm and a temperature of 260 ℃ for a duration of 100 hours. The experimental temperature was below the melting point of the starting sample, which is about 280℃ [7]. The acceleration field at the maximum distance (35.5 mm) from the rotor axis in the specimen was 1,020,000 G.

3. RESULTS AND DISCUSSION

The EPMA mapping photographs of Se and Te at the polished surface cut at a plane containing the rotation axis of the ultracentrifuged specimen are shown on the upper part of Fig. 2, and the linear composition profiles of Se and Te along the direction of gravity from the right edge of the ultracentrifuged specimen are shown in the left lower part of Fig. 2. The maximum acceleration field at the right edge of the specimen was 1.02×10^6 G, and the minimum one at the left edge of the specimen was 0.85×10^6 G. In the low gravity field region of the ultracentrifuged specimen, the content of Te with a heavier atomic weight (127.6) greatly increased in the direction of gravity from about 0 to 60 at.%, while the content of Se with a lighter atomic weight (79.0) greatly decreased from about 100 to 40 at.%. The polarmicroscope photograph of the polished surface cut at a plane containing the rotation axis of the ultracentrifuged specimen is shown on the right lower part of Fig. 2. The large and long crystals oriented along the direction of gravity can be observed in the low gravity field region. The grain sizes of the large and long crystals were several mm long and hundreds of μm wide, while those of the small crystals in the high gravitational field region were tens of μm. This crystal growth might be related to the ultra-strong gravitational field because the orientation of the long grown crystals was consistent with the direction of gravity. The large composition gradient was observed even in the large grown single crystals. This showed that the graded structure was continuous in atomic scale and the sedimentation of substitutional atoms occurred.

The large composition gradient was obtained in the low gravity region, which was caused mainly by the change of the diffusion coefficient. In the low gravity region, the diffusion coefficient became larger with the increase of the Se content because the melting point of Se was lower than that of Te. Therefore, the sedimentation proceeded faster in the low gravity region than that did in the high gravity region. If the ultracentrifugation was continued, the sedimentation might progress in the high gravity region as well.

The position of the boundary between the large crystals in the low gravity region and the small crystals in the high gravity region was consistent with that of the boundary where the compositions of Se and Te changed greatly. This indicated that the crystal growth under the ultra-strong gravitational field was strongly related to the sedimentation of atoms. The crystal structures of Se and Te are hexagonal, which shows large anisotropy. Therefore, the diffusion velocity of atoms in a certain orientation of the hexagonal structure might be faster than those in other orientations for the Se-Te alloy under the ultra-strong gravitational field, which caused the preferential crystal growth along the direction of gravity.

4. CONCLUSIONS

An atomic-scale graded structure was formed in the all-proportion miscible Se-Te system (70:30 in mol%) solid solution by the sedimentation of substitutional solute atoms under an ultra-strong gravitational field of 1 million G level in maximum acceleration at 260℃. The crystal growth in the direction of gravity was observed, while it is difficult to grow a single crystal in this system alloy by ordinary method [8]. The graded band gap structure can be expected because of the semiconductor property of the Se-Te system solid solution. The functionally graded material in atomic scale is expected to show unique electronic or optical properties [9, 10]. It is expected that the strong gravitational field will offer us new and powerful options in

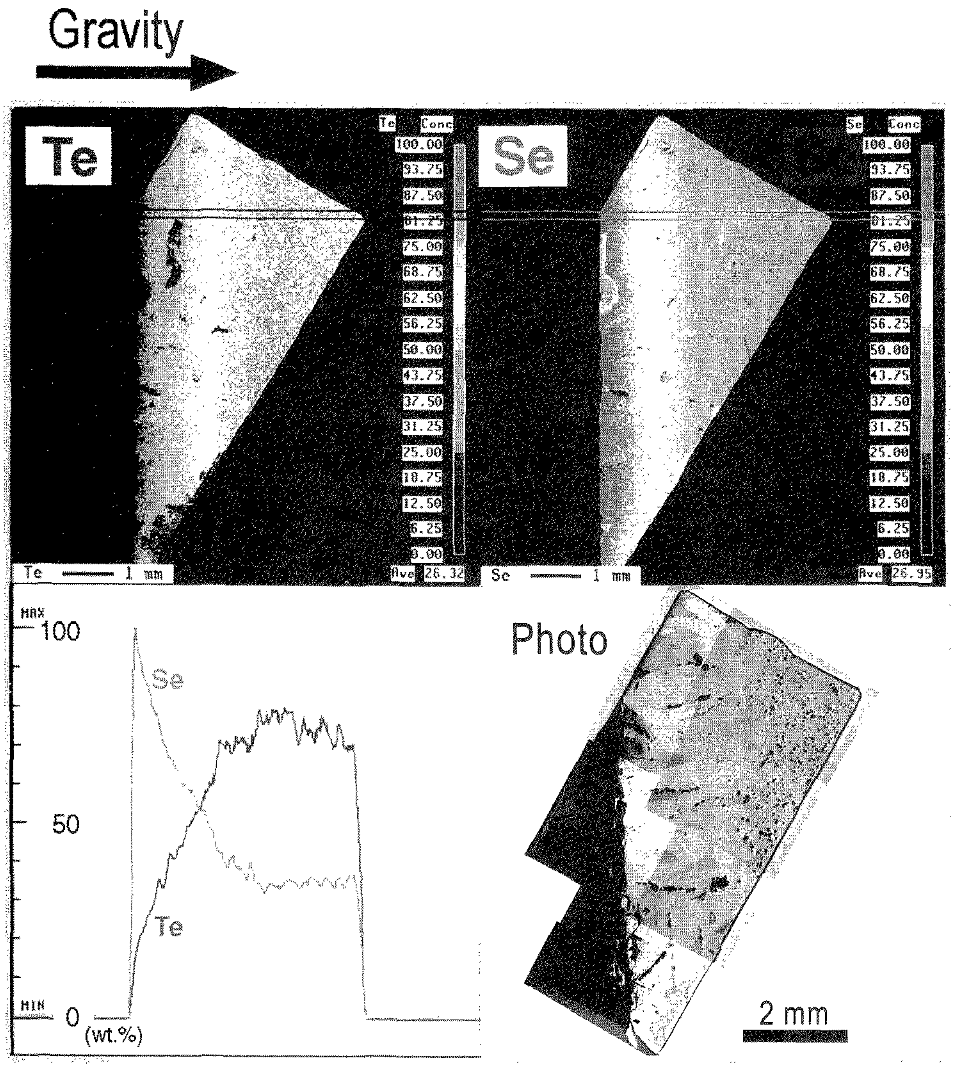

Fig. 2 EPMA mapping photographs of Se and Te, linear composition profiles Se and Te along the direction of gravity and polarmicroscope photograph of the polished surface cut at a plane containing the rotation axis of the ultracentrifuged specimen.

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