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Isotope Distribution in Pure Indium after Liquid-State Centrifugation

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Abstract

An ultracentrifuge experiment was performed on a pure liquid-indium at centrifugal acceleration field of 8.2×10^5 g at 300℃ for 100 hours. The isotope ratio measurements were performed on the centrifuged specimen, which was cooled and solidified before release the acceleration, with Secondary Ion Mass Spectrometer (CAMECA IMS-6f). $^{113}In/^{115}In$ isotope ratio on the specimen changed with negative gradient in the direction of centrifugal acceleration approximately 1.4%. The measurement result indicated that the heavy ^{115}In isotopes moved in the direction of the centrifugal acceleration and the light ^{113}In isotopes moved in the counter direction in the specimen.

1. Introduction

Sedimentation of particles in gas or liquid phase is well-known macroscopic phenomena under a centrifugal acceleration field, and "the higher the relevant centrifugal acceleration, the smaller the minimum size of the sediment particles". And, the sedimentation in a solid is induced in an atomic scale under strong centrifugal acceleration field condition where an influence of centrifugal acceleration on the diffusion cannot be ignorable.

In 1969, the sedimentation of interstitial impurity atoms in a pure solid metal, i.e., the sedimentation of non-constitutional atoms was confirmed by the depth profiling of gold radioisotopes in solids centrifuged potassium under a centrifugal acceleration field of 1.6×10^5 g[1)]. In 1997, the sedimentation of substitutional solute atoms, i.e., the sedimentation of constitutional atoms was verified by evaluating the change of the lattice constant of a graded structure in an all-proportional miscible Bi-Sb alloy processed by centrifugation under a centrifugal acceleration field of almost 10^6 g[2)]. As noted above, sedimentation of atoms occurs in solids under strong centrifugal acceleration filed. And, recently, we were successful in the sedimentation of constitutional isotopes in the solid-state single element of pure Se, where the sedimentation was dependent only on the small mass difference among the relevant isotopes[3)]. The isotope fractionation was confirmed by the precise characterization of the isotope re-distribution in the centrifuged sample using Secondary Ion Mass Spectrometry (SIMS). And, we have succeeded in developing a prototype rotor having two grooves to realize the new isotope separation process based on the phenomenon described above, i.e., the centrifugal isotope separation process in solid-state materials[4)]. In the development, indium has been chosen for the isotope separation experiment instead of selenium from the main reason that natural indium consist of only two isotopes and a relatively simple procedure is expected to verify the mass dependent isotope fractionation in single element.

Not only in the solid-centrifugation, we also observed the isotope ratio gradient in pure selenium that was centrifuged at 820,000g in the molten-state (liquid-state), and it was tentatively concluded that the sedimentation of isotopes occurred in the liquid[5)]. However, in the case of liquid-centrifugation, as the vacuum-tight sedimentation tubes are used in the ultracentrifuge processing, a molten specimen in a sedimentation tube has to be solidified before the release of a centrifugal force to keep the centrifugally fluctuated isotope distribution. So, there is a possibility that the isotope fractionation effect in the recovered specimen contains not only by liquid-centrifugation process but also due to solidification or idle centrifugation time of several hours in solid state to cool the sample enough to near the room temperature.

For the above reason, it is indispensable to measure the recovered two specimens that were separated into a heavy and a light isotope rich part respectively during the process of centrifugation to judge the sedimentation of isotopes apparently occurs in the liquid, and we have planed a series of experiments to do the judgment. The two specimens for the judgment will be obtained by using the developed rotor described before.

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And we are going to use pure-indium as a specimen for same reason as in the rotor development. It is expected that the rotor can be used as is only applying the suit experimental conditions for liquid-indium.

In this study, we have performed an ultracentrifuge experiment on a pure liquid-indium with a sedimentation tube and have characterized the isotope fractionation in the centrifuged specimen to get fundamentals for the experiment to judge the sedimentation of isotopes apparently occurs in the liquid.

2. Experimental Procedure

2.1 Specimen

Indium has been chosen for a series of experiments described above instead of selenium, which had already finished the ultracentrifuge experiment and characterization of isotope fractionation in the specimen, from the following reason that natural indium consists of only two isotopes of ^{113}In and ^{115}In with the relative isotopic abundance of 4.3% and 95.7%, respectively[6], and a relatively simple procedure is expected to verify isotope fractionation than selenium that has many isotopes. In this study, a column shaped with a diameter of 5mm and a height of 7mm and 99.99% high pure indium (Rare Metallic Co., Ltd.) was used.

2.2 Ultracentrifuge experiment

A specialized ultracentrifuge for material science that were developed at Japan Atomic Energy Agency (JAEA) to study sedimentation of atoms or crystal-chemical instability in solid under a strong gravitational filed by a collaborative study with Kumamoto University was used[7]. It can generate a centrifugal acceleration field up to 1 x 10^6 g at elevated temperature. A SUS304 made sedimentation tube containing a specimen of indium was set into a Ti alloy made rotor with an outer diameter of 80 mm. The maximum distance form the axis of rotation in a specimen was about 35.5mm. The ultracentrifuge experiment was performed at a rotational speed of 152,000 rev. min^{-1} ($8.2x10^5$ g) at 300℃ for 100 hours, and it was cooled and solidified before the release of a centrifugal force.

2.3 Isotope Ratio Measurement

The centrifuged indium was cut into halves with cross-sectional surfaces containing the axis of rotation. One piece of them was potted in the acrylic resin and the plane of the resin with a cross-sectional surface of the specimen was well polished with 3 μ m diamond paste. A potted natural indium with well-polished surface was also prepared as same procedure above for isotope ratio measurement of initial state. Both samples were coated by gold evaporation and the preparation of samples for the isotope ratio measurements was finished. The isotope ratio measurements were performed on both a starting material of natural indium and the centrifuged one by a secondary ion mass spectrometer (SIMS, CAMECA IMS-6f). In SIMS analysis, an O_2^+ beam of 15keV with a current of 1.3 nA was used as the primary ion beam. The diameter of the beam, raster size, and image field were a few μ m, 50 μ m and 150 μ m, respectively. The positive secondary ions emitted from the sample were accelerated to 5 keV. An energy slit was adjusted for a band pass of 10 eV. The mass resolution was set at 300. The secondary ions at m/z=113 and 115 were counted in an automatic peak-jumping mode. An electron multiplier was operated in the ion counting mode with a dead time of 25 ns. Pre-sputtering time was set as 600s in each measurement area to remove Au evaporation layer and oxides on the surface. The data correction using the mass discrimination technique was not performed, and all of the isotope ratios in the figures were raw data. The isotope ratios of starting material were measured not only to compare the isotope ratio of initial and centrifuged but also to evaluate the stability of the present isotope measurement.

3. Results and Discussions

Figure 1 shows the ^{113}In/^{115}In isotope ratio distribution of initial-state of natural indium obtained by SIMS. Respective 7 spots on the surface of the specimen were measured. The ^{113}In/^{115}In isotope ratios were distributed around 0.04506, and the fluctuations from the central values were lower than 0.35%. The average value of ^{113}In/^{115}In isotope ratio was 0.04506±0.00018 (the error represent one standard deviation).

Figure 2 is a photograph of the polished surface representing

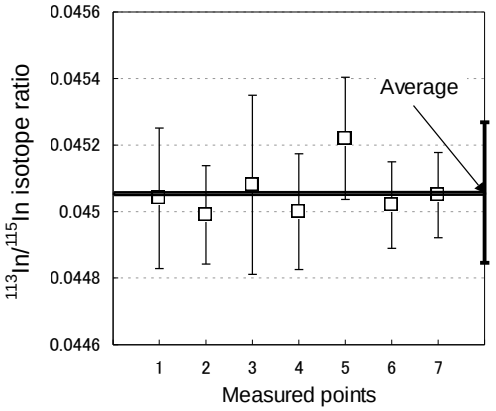

Fig. 1 The ^{113}In/^{115}In isotope ratio distribution of initial-state of natural indium obtained by SIMS. The solid line represents the average value (0.04506±0.00018). The error bars represent one standard deviation.

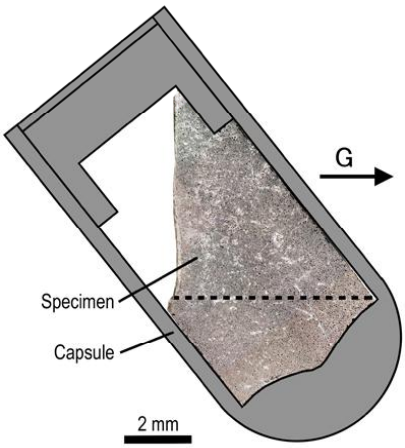

Fig. 2 A photograph of the polished surface of the centrifuged indium. The SIMS measurements were performed along the dotted line.

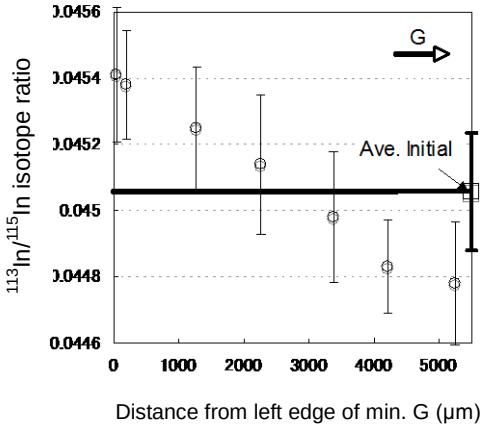

Fig. 3 The ^{113}In/^{115}In isotope ratio distribution of centrifuged indium obtained by SIMS. The solid line represents the average value of initial (0.04506±0.00018). The error bars represent one standard deviation.

a plane in the centrifuged specimen containing the axis of rotation. The sketch is the total complete view of the cross-sectional plane of the centrifuged specimen in the SUS 304 capsule.

Figure 3 shows the distribution of ^{113}In/^{115}In isotope ratio on cross section of centrifuged sample obtained by SIMS from minimum to maximum radius. The SIMS measurements were performed along the dotted line on the photograph in **Fig.2**. The ^{113}In/^{115}In isotope ratio linearly decreases from 0.04541 to 0.04478 in the direction of centrifugal acceleration, and the magnitudes of the isotopic fractionation are about 1.4%. Isotope ratio fluctuations of centrifuged specimen are about 4 times larger than that of initial and there are the apparent tendency that heavy ^{115}In isotope linearly increased and light ^{113}In isotope linearly decreased in the direction of centrifugal acceleration. These observations clearly indicate that a strong centrifugal acceleration filed affects self-diffusion of indium as isotope fractionation effect, i.e., the sedimentation of isotopes occurs.

4. Conclusion

We have performed an ultracentrifuge experiment on a pure indium at liquid state with a sedimentation tube. It has confirmed that the sedimentation of isotopes apparently occurred in the centrifuged specimen, which was cooled and solidified before release the acceleration, by characterizing the isotope fractionation in the specimen. The experiment to judge the sedimentation of isotopes apparently occurs in the liquid will be done using indium as a specimen.

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References

1) L. W. Barr and F. A. Smith: Philos. Mag., **20** (1969) 1293.
2) T. Mashimo, S. Okazaki and S. Tashiro: Jpn. J. Appl. Phys., **36** (1997) 498.
3) T. Mashimo, M. Ono, X. Huang, Y. Iguchi, S. Okayasu, K. Kobayashi and E. Nakamura: Europhysics Lett., **81** (2008) 56002.
4) M. Ono, M. Sueyoshi, S. Okayasu, T. Hao, F. Esaka, T. Osawa, Y. Iguchi and T. Mashimo: Rev. Sci. Instr., **80** (2009) 083908.
5) T. Mashimo, M. Ono, X. Huang, Y. Iguchi, S. Okayasu, K. Kobayashi, E. Nakamura: Appl. Phys. Lett., **91** (2007) 231917.
6) J. Emsley: The Elements, 3rd ed (Oxford Univ. press, Oxford, 1998).
7) T. Mashimo, X.S. Huang, T. Osakabe, M. Ono, M. Nishihara, H. Ihara, M. Sueyoshi, K. Shibasaki, S. Shibasaki, and N. Mori: Rev. Sci. Instr., **74** (2003) 160.

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