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Brief of the doctral thesis (論文要約)

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author: Yuichiro NAGAI

表題: 負イオン表面電離型質量分析計による精密モリブデン同位体分析法の開発
および隕石における元素合成由来の同位体異常の研究

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In Chapter 1, we introduce the application of Mo isotopes in cosmochemistry, and define the objective of this study. Molybdenum is one of the promising elements to investigate chemical or physical processes in various fields, including cosmochemistry. It has seven stable isotopes (^{92}Mo , ^{94}Mo , ^{95}Mo , ^{96}Mo , ^{97}Mo , ^{98}Mo , ^{100}Mo) synthesized via nucleosyntheses of the s-process, the r-process, and the p-process. Development of techniques utilizing state-of-the-art mass spectrometry instruments in the last two decades made it possible to detect marginal mass-independent fractionations in bulk meteorite. The fractionations are nucleosynthetic in origin, which are named “isotope anomalies”. Molybdenum isotope anomalies for various meteorites provide strong constrains on studies concerning stellar nucleosynthesis and the origin and evolution of the Solar System, although the cause of anomalies remains unclear due to the lack of the precision for Mo isotope analysis. In this study, we developed the analytical methods

for Mo isotope analysis including chemical separation and highly precise isotope analysis by utilizing N-TIMS for the study of Mo isotope anomalies in meteorites.

First, we developed a sequential chemical separation technique for Mo, W, and the group of Ti-Zr-Hf by evaluating the obtained distribution coefficients using an anion exchange resin (Eichrom AG1X8) and HCl-HF and HF-HNO₃ mixtures (Chapter 2). Total volumes of resins and reagents were reduced during the chemical separation procedure. The total recovery yields of Mo, W, Zr and Hf were nearly 100% from several terrestrial and extraterrestrial samples.

Next, highly precise Mo isotope analysis was established by utilizing N-TIMS (Chapter 3). Molybdenum was analyzed as Mo-trioxide ion (MoO₃⁻) to obtain efficient ion beam. In this study, the optimal condition to ionize Mo was to load Mo on a Re Single filament together with La(NO₃)₃ activator so that La/Mo = 5. To achieve highly precise Mo isotope analysis, it is important to determine the in-situ O isotope composition for each measurement and use the data to correct for the O isotope interferences. As a result, the reproducibilities have been improved at the level of 47, 16, 10, 13 and 33 ppm for ⁹²Mo/⁹⁶Mo, ⁹⁴Mo/⁹⁶Mo, ⁹⁵Mo/⁹⁶Mo, ⁹⁷Mo/⁹⁶Mo, ¹⁰⁰Mo/⁹⁶Mo, respectively.

Finally, Mo isotopic compositions in seven non-carbonaceous chondrites, and five iron meteorites were obtained by utilizing the technique of Mo isotope analysis developed in this study (Chapter 4). Molybdenum isotopic compositions for these meteorites are obviously discriminated from those for terrestrial rocks. Our Mo isotopic data reveal that carbonaceous chondrites and non-carbonaceous chondrites exhibit

different Mo isotopic compositions, which reflect the existence of at least two isotopically distinct reservoirs in the solar nebula. Molybdenum isotopic compositions for iron meteorites were also identical to those for non-carbonaceous chondrites, except for IVB iron meteorites. Molybdenum isotope anomalies for IVB iron meteorites similar to those for carbonaceous chondrites indicate that these iron meteorites derived from the more oxidized conditions than that for the other iron meteorites. A combination of Ni-Mo isotope anomalies supported two distinct reservoirs in the solar nebula. The different isotopic compositions reflect physicochemical processes by which existed presolar materials were affected in each reservoir. Molybdenum isotope anomalies in non-carbonaceous chondrites are not simply correlated with the Ru isotope anomalies, which suggests that the Mo and Ru anomalies were not caused by a single process, but probably coincided with the fractionation of Mo and Ru. The positive correlations between Mo isotope anomalies and Ga/Ni ratios would reflect the behavior of the carriers enriched in r-process and p-process isotopes in the early Solar System.