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銀河化学進化の研究に向けた同位体希釈-TIMS 法による極微少量 U・Th 定量技術の開発
Determination of extremely small amount of U and Th by ID-TIMS for the study of U-Th cosmochronology

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Primitive chondrites are known to have internal isotope anomalies for various elements due to the existence of presolar grains. These grains provide information on stellar nucleosysthesis where the grains have formed before the onset of Solar System. In particular, abundance ratios of radioactive r-process nuclides such as ^{232}Th, ^{235}U and ^{238}U in presolar grains would provide the age of r-process nucleosynthesis. Of presolar grains, silicon carbide (SiC) can be a promising target because it has relatively larger grain size and contains abundant trace elements compared to the other presolar grains. The amount of U and Th in presolar SiC grains separated from 1 gram of carbonaceous chondrite is expected to be less than 1 pg. Therefore, we need to develop a micro analytical technique that enables to determine extremely small amount of U and Th.

Historically, presolar grains have been analyzed using secondary ion mass spectrometry (SIMS), but spot analysis of trace elements with <ppm abundance is very difficult. In contrast, thermal ionization mass spectrometry (TIMS) is suitable to measure very small amount of trace elements because it provides stable beam intensity relative to the other mass spectrometry, especially when chemical separation has been carried out before analysis. In this study, we have developed the determination of sub-pg amount of U and Th by using TIMS coupled with isotope dilution method.

The performance of TIMS measurement was evaluated by repeatedly analyzing variable amounts of U and Th standard materials with the addition of^{236}U and ^{229}Th enriched spikes. The analytical precisions for determining 1 pg of U and Th were 0.36% and 0.92%, respectively.

In addition to TIMS analysis, we have also developed a separation method for extremely small amount of U and Th utilizing two-step ion exchange column chemistry. In the first column, the sample solution was dissolved in 0.1 mL of 5M HNO_3 and passed through 0.01 mL of U/TEVA spec. Thorium was eluted with 0.1 mL of 5M HCl together with Zr, followed by U elution with 0.1 mL of 0.3M HF-0.1M HNO_3. The Th fraction was dried and dissolved in 0.1 mL of 8M HNO_3 to pass through the second column that consists of 0.05 mL of AG1x8. Using these systems, we were able to separate major elements from U and Th, and achieved U and Th recovery yields of 93% and 82%, respectively.

キーワード: TIMS, 微少量サンプル, ウラン, トリウム, コンタミネーション, 化学分離
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