Chemical separation of Mo, W and HFSEs for the study of multi-elemental isotope anomalies in meteorites

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Introduction: Recently, developments of mass spectrometry have made it possible to detect small isotope anomalies in some heavy elements present in a variety of meteorites. Some of the anomalies are only marginal, with <10 ppm deviations from terrestrial samples. To make highly-precise and "accurate" isotope analysis, it is essential to develop chemical separation of the target element from various type of meteorites with the following objects: 1) Achieve of nearly 100% recovery to avoid mass fractionation during chemical separation. 2) Complete removal of unwanted elements that can interfere with isotope analyses of target elements. 3) Maintain high sample/blank ratios. 4) Simultaneously separate as many elements as possible from a single sample. The last point is important not only to preserve precious meteorites, but also for performing multi-elemental isotope analysis on heterogeneous samples. Molybdenum, W and HFSEs (High Field Strength Elements, e.g. Hf, Zr) are very curious elements for the study of isotope anomalies in the early solar system. In this study, we have developed a chemical separation method for Mo, W and HFSEs from meteorite samples. We also show our preliminary analysis of Mo isotope compositions in bulk chondrites and their acid leachates.

Experiments: The separation method consists of a two-stage column chemistry using anion exchange resin (BioRad AG1X8, 200-400 mesh). We have evaluated the performance of our technique by using a synthesized multi-element solution (containing Mo, W, Hf, Zr, Nb, Ta and Zn) and some terrestrial rock samples (JB-3, JA-2, JG-1a and JP-1) dissoloved by HF. A quadrupole-type ICP-MS (*ThermoFisher X SERIES II* at Tokyo Tech.) was used to determine the elution profile, as well as the recovery yield of Mo during the column chemistry. Molybdenum isotopes were analyzed by N-TIMS (TRITON *plus* at Tokyo Tech).

Results and Discussion: In the first column (1 mL), HFSEs, W and Mo were successively eluted by 9M HCl-0.05M HF, 9M HCl-1M HF, and 6M HNO₃-3M HF, respectively. Unwanted elements in the Mo fraction (Zn, Nb) were further separated in the second column (0.1 mL). Both elution profiles are shown in Fig. 1 and Fig. 2. The column size and total amount of acids were dramatically reduced compared to previous techniques. The amount of Zr and Ru remaining in the Mo fraction were below detection limits. The Mo recovery was 94.5 ± 2.9 % in the first column and 101.3 ± 5.0 % in the second column. By applying this tech-

nique, we have determined Mo isotope compositions in bulk samples of Murchison (CM2) and Allende (CV3), as well as their acid leachates. The Mo isotope anomalies in bulk Murchison and a leachate indicate deficits of *s*-process Mo isotopes, consistent with previous studies [1-3]. In contrast, it is very difficult to explain the anomalies in bulk Allende by a mixture of only three distinct nucleotynthetic components (*s*-, *r*- and *p*processes). These results may require more nucleosynthetic sources, or a reassessment of the isotopic compositions of endmember components, calculated by current nucleosynthetic theories.

Reference: [1] Burkhardt et al. (2011a) *EPSL* **312**, 390-400. [2] Burkhardt et al. (2011b) *LPSC* **42**, #2592. [3] Dauphas et al. (2002) *ApJ* **565**, 640-644.

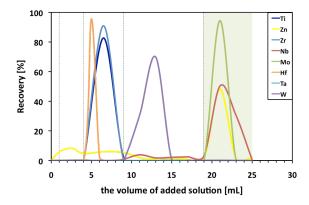


Fig. 1. Elution profile (JB-3) of the first column. Individual steps of chemical separation are divided by dashed lines. Light green range is the Mo fraction.

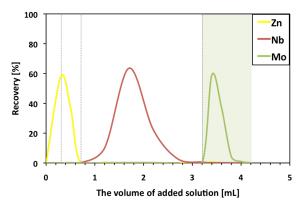


Fig. 2. Elution profile (JB-3) of the second column. Leagents are the same as Fig. 1.