

Isotopic determination of ultra trace U in meteorite samples by HR-ICP-MS

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Uranium is of considerable interest in geochemistry and cosmochemistry. Its absolute and relative abundances in geochemical and cosmochemical samples have been discussed widely and deeply due to its significance for understanding the origin and evolution of our solar system and the earth. The disintegration of extinct nuclide ²⁴⁷Cm ($T_{1/2}=16$ Ma) to ²³⁵U by α decay leads to changes in the ratio of ²³⁵U/²³⁸U. The chemical behavior also implies that the fractionation between Cm and U enlarge the ratio. This variation may provide strict constraints on the time interval between the last r-process nucleo-synthetic event and the formation of the solar system.

The possibility of ²⁴⁷Cm as a short chronometer was firstly calculated 30 years ago by Blake and Schramm (1973). Since then, precise and accurate isotope ratio measurements have been carried out by thermal ionization mass spectrometry (TIMS). However, some studies showed a large anomalies of U isotopes up to several percent [1], others could not testified these findings as Cm was not abundant in the early solar system [2]. The presence of live ²⁴⁷Cm was not yet settled.

Therefore in this study an inductively coupled plasma mass spectrometry (ICP-MS) instrument with a magnetic sector mass analyzer and a single collector configuration had been used in the measurement of U isotope ratio. To search for the abnormalities of U isotope ratio in specific mineral phases in meteorites will require not only careful chemical operations but also monitoring the instrumental mass bias. In this experiment, an anion-exchange procedure was introduced to remove the matrix elements to eliminate the matrix effects, and a high quality ²³⁵U-²³⁸U standard solution was used to monitor the instrumental mass bias. All the experiments were performed in the clean rooms in Japan Atomic Energy Research Institute (JAERI), in which the process blank could be controlled within 1 pg level. Our results show that using an ICP-MS (ThermoQuest instruments, Element), we are able to resolve variations in ²³⁵U/²³⁸U at the order of ± 4 permil on sample solution of 100 ppt or 200pg of uranium.

This method will be applied to the analysis of ultra trace U (<500 pg) isotopic measurements in specific mineral phases in meteorites. The data will be claimed whether ²⁴⁷Cm were abundant or not in early solar system.

References

- [1] Tatsumoto M. et al., (1980), *Nature*, 286, 118-122
[2] Chen J. H. et al., (1981), *Anal. Chem.*, 53, 2060-2067

Microbial polymer templation of iron oxyhydroxides

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Biopolymers have been shown to control the nucleation, orientation, and growth of crystals at the nanometer- and micron-scale and thus have been used in efforts to create functional nanomaterials. Microbial mineralization can have a significant effect on environmental chemistry by producing minerals with phases, morphologies, and reactivities that differ from inorganically precipitated minerals.

We have found evidence of a novel form of polymer-assisted biomineralization: extremely thin (few-unit cell wide), microns-long akaganeite (β -FeOOH) filaments, or nanowires, that are templated on natural microbial polymer fibrils. These filaments were identified in iron oxide-encrusted microbial mat samples in the flooded Piquette Mine, WI, USA using high resolution transmission electron microscopy. These pseudo-single crystals are composed of assembled elongate nanocrystals oriented end-to-end in the [010] direction. The presence of akaganeite is unusual, since it is generally considered to form in high salinity environments, suggesting that polymer templation has altered iron oxide phase stability.

In order to verify the presence of the polymer, filaments were imaged by synchrotron X-ray photoelectron emission microscopy (PEEM). The carbon 1s X-ray absorption near edge spectrum (XANES) of the filaments matches best with alginate, a well-characterized microbial polysaccharide, when compared with XANES of various microbial components, including proteins, lipids, and nucleic acids.

To test the hypothesis that polymer fibrils can template akaganeite filament formation, we performed experiments simulating the postulated mineralization pathway. We are able to reproduce a similar structure in the laboratory using model biopolymers (a mix of chitosan, chondroitin sulfate and alginate), providing evidence of the polymers' role in akaganeite filament formation.

In the natural samples, mineralized filaments are sometimes found in association with microorganisms, and appear to be extruding outward from the cells. Thus, we infer that organisms that are oxidizing iron (or are living in environments where rapid iron oxidation is occurring) extrude polymer strands to localize precipitation reactions. Iron oxyhydroxide precipitation generates protons; localization of this reaction near the cell wall may increase the proton motive force, conferring a metabolic advantage to these neutrophilic organisms.