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大陸地殻におけるウランの物質移行の長期変動予測
Challenges to Predict the Long-Term Uranium Migration in Deep Terrestrial Crust

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Our understanding of uranium mobility in the near surface environment has been rapidly expanding in the past decades, especially due to problems associated with environmental remediation of U-contaminated sites. After Fukushima Daiichi nuclear disaster, it is becoming important to study the environmental behavior of U even in Japan, because meltdown debris mainly composed of UO_2 need to be disposed of in a deep geological repository. Recently, it has been demonstrated that our understanding of U in deep granitic rocks is limited, as exemplified by an unexpected finding of high concentrations of uranium in reducing deep granitic groundwater at a Swedish geological disposal site. As Japan is located at tectonically active plate boundaries, geological factors influencing the mobility of uranium remain to be clarified.

Biogeochemical processes mediated in the deep granitic aquifer at the Mizunami underground research laboratory (URL) were studied in details and published in Suzuki et al. (2014). Briefly, large extents of sulfur isotopic fractionation between sulfate and sulfide and zinc sulfide precipitation clearly indicate that the aquifer is under sulfate-reducing conditions. Groundwater concentrations of U were measured to be exceedingly low at the ppt level. To constrain factors determining the low U concentrations, fracture-filling minerals were investigated by scanning electron microscopy (SEM) coupled to energy dispersive X-ray spectroscopy (EDS) for focused ion beam (FIB) fabricated samples. At 300 meter below ground level (mbgl), pyrite and chlorite were associated with U-bearing loci. One of the samples was FIB fabricated for transmission electron microscopy coupled to EDS, and selected area electron diffraction (SAED) patterns, chemical compositions and spacings of lattice fringes were all indicative of the formation of a U(IV) silicate mineral called coffinite. High-resolution TEM observations also revealed that coffinite was crystalline. The solubility of crystalline coffinite was thermodynamically calculated by Phreeq C with our selected database and consistent with the U concentrations in the deep granitic aquifer. It is therefore concluded that U is stable as its reduced state and immobile in the present groundwater.

Tono uranium ore deposits are adjacent to the Mizunami URL, and geological settings of the two sites are almost the same. 10 Ma U mineralization occurred in the overlying sedimentary formation where lignite is a reducing agent for U(VI). It is well established that U is mobilized from granite under oxidizing conditions and transported to downstream ore bodies with abundant reductants such as organic matter and pyrite. However, the current redox state of granitic groundwater at the Mizunami ULR is under stably reducing conditions, and the established concept involving oxidizing groundwater appears not to be a case. As a significant transgression event is known to have occurred 10 Ma, there is the possibility that the intrusion of seawater into the freshwater aquifer might be important as suggested for the Swedish repository site after the last glacial maximum. We investigated the paragenesis of cofinite, pyrite and chlorite within calcite grains formed during the 10 Ma transgression by SEM-EDS, carbon and oxygen stable isotope measurements of microdrilled calcite and field emission elemental probe microanalysis (FE-EPMA). Results will be shown to discuss about the reconstruction of geological events and their influences on U migration in the deep granitic rock.

Suzuki, Y, Konno, U., Fukuda, A., Komatsu, D. D., Hirota, A., Watanabe, K., Togo, Y., Morikawa, N., Hagiwara, H., Aosai, D., Iwatsuki, T., Tsunogai, U., Nagao, S., Ito, K., Mizuno, T. (2014) Biogeochemical Signals from Deep Microbial Life in Terrestrial Crust, PloS one, 9(12), e113063.