

A study of U(VI) sorption on illite under carbonate-rich environments: Batch experiments, TRLFS and modeling

*Huiyang Mei¹, Noboru Aoyagi¹, Takumi Saito², Naofumi Kozai¹, Yukio Tachi³

1. Advanced Science Research Center, Nuclear Science Research Institute, Japan Atomic Energy Agency, Ibaraki, 2-4 Shirakata, Tokai-mura, Ibaraki 319-1195, Japan, 2. Nuclear Professional School, School of Engineering, The University of Tokyo, 2-22 Shirakata Shirane, Tokai-mura, Ibaraki 319-1188, Japan, 3. Nuclear Fuel Cycle Engineering Laboratories, Japan Atomic Energy Agency, 4-33 Muramatsu, Tokai-mura, Ibaraki 319-1194, Japan

The safety assessment for radioactive waste repositories largely relies on understanding the mechanisms involved in radionuclide retention by minerals in host rocks. Argillaceous rocks have been considered as one of the potential host-rock formations for geological repository in several countries. Carbonate ion is contained at relatively high concentrations in sedimentary groundwater in Japan ($> 2,000$ mg/L as HCO_3^- in some cases) and exerts a significant influence on the fate of radionuclides by formation of aqueous complexes. However, it is not clear whether the ternary surface complexes containing carbonate will be formed, which may affect the distribution of surface species and the subsequent construction of the predictive model. Thus, we investigate U(VI) sorption on illite (one of the main constituents of argillaceous rock) as functions of pH and HCO_3^- concentrations, by combining batch experiments, cryogenic time-resolved laser fluorescence spectroscopy (cryo-TRLFS), and surface complexation modeling using the 2 Site Protolysis Non-Electrostatic Surface Complexation and Cation Exchange sorption (2SPNE SC/CE) model. The cryo-TRLFS uses near liquid helium temperature to enhance the quality of fluorescence spectra (e.g., spectral intensity, resolution), which helps identify the surficial uranyl complexes in complicated environmental systems. In the talk, we will present the macroscopic and microscopic results of the (surface) speciation of U(VI) with illite in the presence of HCO_3^- and discuss the modeling of the sorption data, reflecting the speciation data.

*This work was part of “The project for validating near-field assessment methodology in geological disposal (FY2020)” supported by the Ministry of Economy, Trade and Industry of Japan.

Keywords: U(VI), illite, carbonate, surface complexation model, TRLFS