Contribution ID: **964** Type: **Verbal**

Influence of Surface Nanotopography on the Adsorption of Europium on Muscovite (001)

Tuesday, 17 May 2022 15:50 (18 minutes)

Radionuclide migration is one of the key problems for the long-term safety of nuclear waste repositories. One possible mechanism to retard or prevent the migration of radionuclides from the repository to the biosphere is the adsorption onto mineral surfaces of the surrounding host rock. Clay rock formations such as the Opalinus Clay are being considered for potential sites for nuclear waste repositories, partly due to the strong sorption potential of clay minerals. Phyllosilicates, such as clay minerals or mica, have shown a high affinity for adsorption of various radionuclides in several experimental studies. However, mineral surfaces in natural environments are often subjected to reactions (e.g., dissolution) that may alter the surface nanotopography and, consequently, affect the overall adsorption process. Recently, it has been reported that the nanotopography of calcite surfaces leads to heterogonous sorption of europium due to differences in the atomic configuration of the adsorption sites [1].
In this study, we investigate the influence of surface site coordination on the adsorption energy barrier and the resulting overall distribution of radionuclide adsorption on the mineral surface. We utilize numerical methods to study the adsorption of Eu(OH)3 on a muscovite (001) surface with different nanotopographic structures. Density Functional Theory (DFT) calculations are performed to obtain the adsorption energy barriers of several surface sites present on muscovite. For each site, the adsorption energy is calculated based on a series of geometry optimizations with increasing Eu–site distance. The values of the site-specific adsorption energy barriers are then implemented in a Kinetic Monte Carlo (KMC) model based on a previous study [2]. In the KMC model, larger surface structures, such as steps or etch pits, are placed on the muscovite surface and a dissolution simulation is performed to create a realistic nanotopography. Based on the adsorption energy barriers obtained with DFT, Eu(OH)3 is adsorbed on the generated muscovite surface in a second KMC model step. The KMC model is then used to predict the distribution of adsorbed Eu(OH)3 and the temporal evolution of the adsorption. Using this combined numerical approach, we show the effect of surface site coordination on radionuclide adsorption reactions and the resulting adsorption heterogeneity on mineral surfaces at large scales.

[1] Yuan, T., Schymura, S., Bollermann, T., Molodtsov, K., Chekhonin, P., Schmidt, M., Stumpf, T. and Fischer, C. (2021) Heterogeneous Sorption of Radionuclides Predicted by Crystal Surface Nanoroughness. Environ. Sci. Technol., 55, 15797–15809.
[2] Schabernack, J., Kurganskaya, I., Fischer, C. and Luttge, A. (2021) Influence of Muscovite (001) Surface Nanotopography on Radionuclide Adsorption Studied by Kinetic Monte Carlo Simulations. Minerals, 11, 468.

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Session Classification: Nuclear Fuel Cycle

Track Classification: Chemistry of Nuclear Fuel Cycle, Radiochemical Problems in Nuclear Waste Management