Spatially-resolved speciation of Eu(III) and Cm(III) on granite surfaces

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The search for a suitable site for a nuclear waste repository in Germany requires linking molecular scale information with the large scale of the repository. Here, we present a novel approach to bridge the gap from the molecular to the millimeter scale.

We complement well-known surface investigation techniques such as Raman microscopy, interferometry and autoradiography with µTRLFS. This newly developed technique allows the investigation of luminescent radionuclides, such as Cm(III) and its chemical homologue Eu(III), on the surface of crystalline rocks with complex mineral composition. The combination of multiple surface investigation techniques allows to draw a correlation between surface mineralogy, topography, radionuclide speciation and the resulting retention behavior.

In an initial µTRLFS study using natural granite from Eibenstock, Germany, it was found that uptake strength, capacity, and homogeneity vary from mineral to mineral. For example, Eu(III) on feldspars adsorbed relatively weakly but in large amounts, whereas only minor sorption was observed on quartz, but with a high sorption strength. In addition, distinct sorption behavior was found on some mineral grain boundaries.[1]

To obtain a more comprehensive picture, granitic drill core samples were obtained from across Europe, from which thin section samples were prepared for µTRLFS experiments. The sorption of Eu(III) and Cm(III) onto these samples was conducted using solutions with defined ionic strength, metal concentration and pH.

We will discuss the speciation differences between varying mineral phases one each rock, as well as differences between the characteristic crystalline rocks from diverse locations and the potential impact of the radionuclide speciation on their migration properties in the geosphere. Additionally the results will be compared to single phase studies from literature to evaluate the validity of an additive component mixing approach.

[1] Molodtsov *et al.* (2019), Sci. Rep., under review.