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Interfacial reactivity of Pu and Th at the muscovite (001) basal plane

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The geochemistry of the actinides is of utmost importance in understanding and predicting their behavior in contaminated legacy sites as well as nuclear waste storage facilities. The unique chemistry of this group of elements including strong hydrolysis, complex redox chemistry, and the potential for polymerization reactions in combination with the actinides'inherent radioactivity and toxicity makes studies challenging. However, especially for artificial elements like Pu and other transuranics, no natural analogues are available and homologues frequently fall short in accurately reproducing the actinides'behavior.
We will present and discuss recent results from in situ resonant anomalous x-ray reflectivity (RAXR) and crystal truncation rod (CTR) experiments, shedding light on the inter-action of Th(IV) as well as Pu(III) and Pu(IV) with the negatively charged muscovite (001) basal plane. The example of Th(IV) demonstrates how the strong hydration of the highly charged cations prevents a close approach to the surface, instead favoring adsorption as a highly hydrated extended outer sphere complex. Subsequently, it will be shown how similar adsorption behavior in combination with the complex redox chemistry of plutonium, leads to a surface-enhanced formation of nanoparticles.
Results from surface x-ray scattering will be supplemented by ex situ alpha-spectrometry quantification and atomic force microscopy (AFM), to yield a more complete understanding of the interfacial structure.

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