INCORPORATION OF RADIONUCLIDES INTO NUCLEAR WASTE-RELATED MATERIALS: JOINT ATOMISTIC MODELING AND EXPERIMENTAL APPROACH

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The ability of radionuclides to dissolve in the specific solid phases to form thermodynamically stable solid solutions and the formation of actinide-bearing secondary phases are important aspects when considering the long-term nuclear waste management options. Conditioning of Pu or minor actinides in ceramic waste forms, such as lanthanide-orthophosphates and pyrochlore compounds, and the retention of radionuclides by the secondary phases that are expected to form under disposal conditions, are two topics that are intensively researched to setup a scientific basis for nuclear waste disposal. In our research we use a joint computational and experimental approach as a path toward superior characterization of materials and their properties. We will show the results of selected joint efforts that aim into characterization of ceramic materials as prospective nuclear waste forms [1] and $SrUO_4$ as a potential secondary phase [2]. In particular, we will discuss the results of investigation of structural incorporation of actinides into ceramic materials, namely Cm incorporation into the rhabdophane and Pu, Am, Np and U incorporation into pyrochlore. Here we will show the usefulness of atomistic modeling for the understanding of distribution of actinide elements between different cation sites and the related crystalline lattice response. We will discuss $SrUO_4$ as an example of a joint atomistic modeling and experimental investigation of the phase transformation under reducing conditions and the related oxidation state chemistry of uranium. We will demonstrate that a combination of the modeling and experimental efforts results in a superior characterization of the investigated materials and processes that are important for the assessment of long-term durability and stability of disposed nuclear waste.

[1] Huittinen et al., J. Nucl. Mater. 486, 148 (2017); Kowalski et al., Nucl. Instr. Meth. Phys. Res. B 393, 68 (2017).

[2] Murphy et al. Inorg. Chem. 55, 9329 (2016).