Reliable atomistic modeling of monazite and pyrochlore ceramics as matrices for immobilization of nuclear waste

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Conditioning of radionuclides in ceramic waste forms and deep geological disposal is one of the considered long-term nuclear waste management options. Among other materials, monazite- and pyrochlore-type ceramics are of potential interest for immobilization of radiotoxic fission products and plutonium. A detailed (refined) characterization and understanding of the structural, thermochemical and thermodynamic properties of these ceramics is essentially needed to predict reliably their long-term behavior as waste forms. *Ab initio* atomistic modeling is an excellent tool for investigation of materials properties that are determined on the atomic scale, which are often difficult or inaccessible to experimental techniques. We present an overview of our recent computational activities in the field [1]. In particular we will discuss various structure-property relationships obtained through systematic studies of the considered ceramics and the first results from the virtual radiation damage experiments performed for monazite-type ceramics. These studies complement the relevant and recently acquired experimental data, which results in superior materials characterization.

We will show that carefully chosen computational approaches result in very reliable prediction of thermochemical and thermodynamic parameters of monazite- and pyrochlore-type ceramics. In particular we will discuss the thermodynamic stability of monazite-type solid solutions with actinides, including full characterization of solid solutions with tetravalent actinides, and the energetics of defect formation and the activation barriers for oxygen diffusion in pyrochlore-type ceramics. These results help in assessment of long-term durability and stability of these ceramics as nuclear waste forms.

[1] Beridze & Kowalski, J. Phys. Chem. A, 2014, 118, 11797; Blanca-Romero et al., J. Comput. Chem. 2014, 35, 1339; Li et al., J. Solid State Chem. 2014, 220, 137; Li et al. Scripta Mater. 2015, 107, 18; Kowalski et al. J. Nucl. Mater. 2015, 464, 147; Kowalski & Li, J. Eur. Ceram. Soc. 2016, *in press*.