**301c Molecular Modeling of Crystalline Silicotitanate and Polyoxoniobate Ion Exchangers**

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Permanent disposal of high-level nuclear waste requires efficient separation of trace amounts of radioactive elements from alkaline waste solutions in order to minimize disposal volume required for vitrification. Development of highly selective ion exchangers with increased capacity and kinetics is desired for a more cost-effective separation process. In collaboration with researchers at Savannah River National Laboratories, Sandia National Laboratories, and Texas A&M University, various classes of high-capacity ion exchangers, such as crystalline silicotitanates (CST), polyoxoniobates and pharmacosiderates are under investigation to enhance their ion exchange performance.

Molecular simulations have the potential to be a valuable design tool in guiding future synthesis efforts. The accuracy of molecular simulations is critically dependent upon the quality of the intermolecular force field used to describe the interactions. The methods used to develop and validate force field parameters that are transferable over an entire ion exchanger class are presented. In addition, ab initio methods, such as density functional theory (DFT) simulations, are investigated to provide additional insight into the electronic structure of these materials and improve upon the force field. Once a suitable force field is obtained, molecular dynamics simulations are applied to predict cation and water positions in the CST and polyoxoniobate materials. Moreover, the location of cations in these materials depends critically on the degree of hydration and the framework composition. To estimate the water loading and its variation with cation exchange, we have conducted grand canonical Monte Carlo (GCMC) simulations to directly compute water adsorption isotherms. These results compare well with existing experimental data, thus providing additional molecular-level insight into the origins of selectivity.