

NUCLEAR ENERGY UNIVERSITY PROGRAMS

Atomistic Calculations of the Effect of Minor Actinides on Thermodynamic and Kinetic Properties of UO_{2+x}

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Abstract

The team will examine how the incorporation of actinide species important for mixed oxide (MOX) and other advanced fuel designs impacts thermodynamic quantities of the host UO_2 nuclear fuel and how Pu, Np, Cm and Am influence oxygen mobility. In many cases, the experimental data is either insufficient or missing. For example, in the case of pure NpO_2 , there is essentially no experimental data on the hyperstoichiometric form – it is not even known if hyperstoichiometry NpO_{2+x} is stable. The team will employ atomistic modeling tools to calculate these quantities.

Using density functional theory (DFT), the team will calculate how oxygen defects interact with these substitutional species in the UO_2 lattice. They will employ the cluster expansion method to extrapolate results to finite temperatures and large sizes. The team will use classical models in combination with Metropolis Monte Carlo to generate representative structures of UO_2 containing various concentrations of minor actinides. In principle, these mixtures could contain multiple actinide species, such as the transmutation fuels with proposed compositions of $\text{U}_{0.8}\text{Np}_{0.15}\text{Am}_{0.05}\text{O}_2$. These Monte Carlo simulations will yield an equilibrium structure that will be analyzed and thermodynamic and structural quantities extracted. These include the formation enthalpy, free energy and expansion coefficients. The results of the DFT and classical simulations will then be fed directly to kinetic Monte Carlo (kMC) models that calculate oxygen diffusivity in the fuel as a function of non-stoichiometry, temperature, and concentrations of the various actinide species. The team will thus gain valuable insight into the kinetics of oxygen redistribution and thus microstructural evolution that can guide the design of advanced oxide fuels.