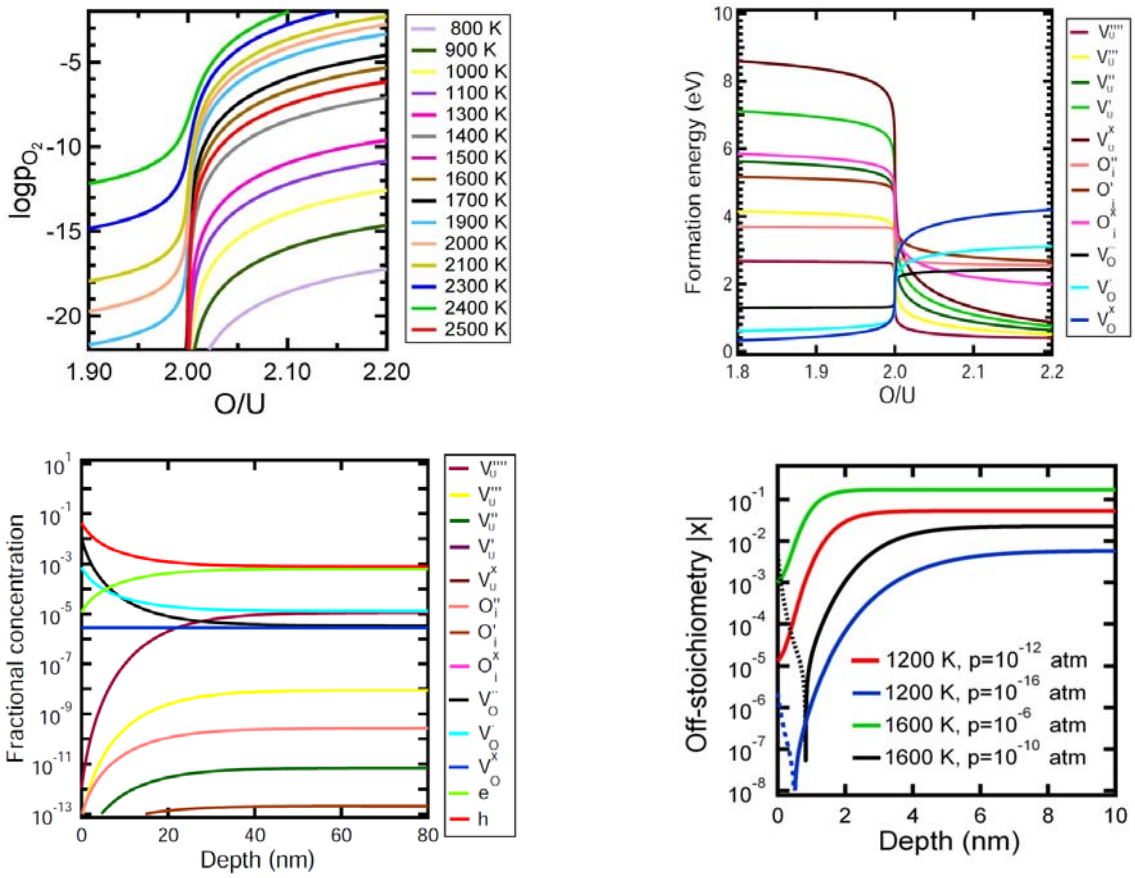
EFRC Center for Materials Science of Nuclear Fuel
Purdue University

Highlight: Thermodynamics and Kinetics of Defect Disorder in UO_2



EFRC researchers Abdel-Rahman Hassan (Ph.D. student) and Anter El-Azab (PI) at Purdue University have developed a model to understand the off-stoichiometry of UO_2 in terms of atomic defects and electronic carriers. The model links the disorder state to the thermodynamic state variables, i.e. temperature and oxygen partial pressure. Its energetic parameters are drawn from DFT literature. A local off-stoichiometry model is devised by including mechanical and electrostatic fields active at the mesoscale level. The result is the spatially resolved off-stoichiometric profile of the oxide near free surfaces or voids. **Top left:** Partial equilibrium oxygen pressure-temperature loci of off-stoichiometric UO_2 for a temperature range of (800-2500 K) as obtained from the point defect model. **Top right:** Free energies of formation for atomic defects in UO_2 as a function of off-stoichiometry at 1600 K. **Bottom left:** Spatial variation of atomic defects and electronic carriers near the surface under the effect of oxygen atmosphere at 1600 K and $p=10^{-20}$ atm. **Bottom right:** Off-stoichiometry variation as a function of depth obtained by the local off-stoichiometry model for several states of UO_{2+x}. Solid lines represent (x>0) or hyper-stoichiometry and dashed lines represent (x<0) or hypo-stoichiometry. *This work has been presented at the 2011 MRS Fall Meeting in Boston, MA and also at the 2012 MRS Spring Meeting in San Francisco, CA. A manuscript has been submitted to Solid State Ionics.*