Cluster dynamics simulation of Xe diffusion in UO_2 and doped UO_2 for out-of-pile and in-pile conditions

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Diffusion of Xe atoms in UO_2 nuclear fuel is important for release of fission gas to the plenum and it also impacts nuclear fuel performance indirectly through, for example, changes in thermal conductivity and swelling. Fission gas behavior remains one of the fuel performance properties with the highest uncertainty, starting with diffusion in the grain interior, through gas bubble formation and resolution and all the way up to percolation of bubbles on grain faces and edges. This presentation focuses on developing new mechanistic models of Xe diffusion within grains for both out-of-pile and in-pile conditions in standard UO_2 as well as for advanced doped UO_2 fuel. The point defect and Xe-vacancy cluster properties determining the diffusion rate for both out-of-pile and in-pile conditions are calculated using a combination of density functional theory and empirical potential methods. The interaction of point defects under irradiation and the formation of mobile Xe-vacancy clusters are modeled based on a free energy cluster dynamics approach implemented in the MARMOT phase field code. This model accounts for the chemical boundary conditions defined by the oxygen potential and calculates the resulting non-stoichiometry in $UO_{2\pm x}$, which has a strong influence on the diffusion rate through the equilibrium concentration of uranium and oxygen vacancies. Our predictions are first validated against experiments for uranium self-diffusion. Next, the simulations of Xe diffusion show that it is dominated by the Xe_{U2O} cluster (a Xe atom occupying a trap site formed by two uranium and one oxygen vacancy) at high temperature (intrinsic conditions) and the Xe_{U4O3} cluster at intermediate temperatures, followed by atomic mixing due to irradiation damage at the lowest temperatures. The predicted diffusion rate is in rather good agreement with experimental observations. Finally, this model is adapted to Cr-doped UO_2 and used to predict the Xe diffusion rate. Implications of our results for fuel performance and plans for future uncertainty quantification are also discussed.