

Simulating Xe redistribution in UO_{2+x}

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From an engineering point of view, the formation and redistribution of fission gases are critical determinants of nuclear fuel performance and in particular limit the extent of burnup. Most fission gases have low solubility in the UO_{2+x} fuel matrix and as a result there is a significant driving force for segregation of gas atoms to heterogeneities such as grain boundaries and subsequently for nucleation of gas bubbles. These effects are most pronounced for large fission gas atoms, which specifically include Xe. Segregation to grain boundaries is often assumed to be followed by more rapid release to the fuel plenum, either via fast diffusion of individual gas atoms or via cooperative transport mechanisms involving nucleated gas bubbles. This implies that the first controlling step for fission gas release is diffusion of individual gas atoms through the fuel matrix to existing bubbles or grain boundaries (sinks), a process that is governed by the activation energy for bulk diffusion of gas atoms, the driving force for segregation to existing sinks (bubbles or grain boundaries) and their saturation limit. First we have studied the bulk diffusion mechanisms of Xe by calculating the corresponding activation energies as function of the UO_{2+x} stoichiometry using density functional theory (DFT) methods.

Estimating the activation energy involves determining Xe migration barriers as well as thermodynamics of Xe trap sites in UO_{2+x} and their interactions with Uranium vacancies that enable Xe in trap sites to move. In separate studies we used atomistic simulations to determine the sink strengths for segregation of Xe to different types of grain boundaries as well as the sink strength variation as function of the Xe loading. Additionally, these studies have established the spatial range of the Xe interaction field with grain boundaries. In summary these studies suggest that in order to properly model Xe redistribution we need to account for, not only the position of grain boundaries, but also the distribution of various types of grain boundaries.

Using the atomistic information on Xe bulk diffusion and segregation we have developed a thermodynamic description of Xe in micro-structurally heterogeneous UO_{2+x} fuels, i.e. a model that accounts for the existence of various types of grain boundaries, each exhibiting unique properties, as well as the local concentration of Xe atoms. This thermodynamic model is then applied in conjunction with calculated Xe mobilities to derive a transport model that explicitly accounts for the Xe interaction field with grain boundaries. Finally this model is solved for a number of grain boundary distributions having different character and grain sizes by using a sharp interface model. We conclude by discussing generalizations of the sharp interface model to a phase field model capable of describing a wider range of phenomena.