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Fission Products in Nuclear Fuel: Comparison of Simulated Distribution with Correlative Characterization Techniques

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During the fission process in a nuclear reactor, uranium dioxide (UO_2) fuel material is irradiated, forming fission products (FPs). The addition of FPs alters the path phonons travel in UO_2, detrimentally altering the thermal conductivity of the fuel. [1] To improve fuel performance, a fundamental understanding of the role of insoluble FPs, such as Xenon (Xe), during microstructural evolution is critical. Correlative characterization techniques where atom probe tomography (APT) is paired with transmission electron microscopy (TEM) can provide unique insights into the segregation behavior of FPs. Coupling these techniques with computer simulations of fission product distribution provide deeper understanding of FP migration during service. Although there are limitations with each of these techniques in isolation, significant insight into material behavior can be gained with the concurrent and synergistic pairing of multiple experimental and computational techniques.

In this investigation, cerium dioxide (CeO_2) was used as a surrogate for UO_2. CeO_2 is a model material with similar properties to UO_2 such as lattice parameter, fluorite crystal structure, thermal conductivity and irradiation behavior. [2] In this investigation, polycrystalline CeO_2 samples were ion irradiated with 400 keV Xe^+ ions at 30°C to a fluence of $1x10^{16}$ Xe ions/cm^2. Post irradiation, Xe-irradiated samples were annealed at 1200°C for 1 hour to induce Xe clustering. For atom probe analysis, a UV-laser assisted CAMECA LEAP 4000X HR was used and the atom probe data was reconstructed using IVAS 3.6.4.

The expected distribution of Xe in CeO_2 was predicted using the Transport of Ions in Matter (TRIM) program in the Stopping and Range of Ion Matter (SRIM) software. [3] TRIM is a kinetic monte carlo simulation program that utilizes potentials to replicate radiation damage. The Xe distribution predicted by the TRIM calculations shown in Figure 1(a) and shows good agreement with the TEM-Energy Dispersive Spectroscopy on as-irradiated CeO_2 is shown Figure 1(b). APT was used to measure Xe in post-irradiation annealed CeO_2 and the results are presented in Figure 1(c). Xe distribution measured by APT and TEM matches well with expected TRIM calculations in distribution. An atom probe reconstruction of the Xe irradiated CeO_2 sample is shown in Figure 2. Distinct differences, however, can be observed in the specific concentrations detected. It can be seen that the concentration of Xe using APT is significantly lower than the TRIM calculation and TEM-EDS analysis. The decrease in amount of Xe detected with APT can be attributed to the clustering of Xe during heat treatment and subsequent release during evaporation in the atom probe. It can be seen that the heat treatment causes Xe to cluster and form gas bubbles (see Figure 3). As the sample is evaporated during analysis, the Xe gas is released within the ultra-high vacuum chamber, leading to low observed concentrations. However, the Xe signal produced in the mass-to-charge spectrum could be attributed to the minute solubility of Xe in CeO_2 caused by the large number of defects formed during implantation. [4] Lastly, it is noted that these

results show the validity of TRIM calculations to accurately predict the distribution of Xe, this is in light of the fact that TRIM simulations do not take into account irradiation temperature, time, or crystal structure of the material under investigation. This works shows the importance of utilizing correlative experimental and computational techniques to develop a framework for the understanding of FP behavior in nuclear fuel. This material is based upon work supported as part of the Center for the Materials Science of Nuclear Fuels, an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Award Number DE-AC07-05ID14517.

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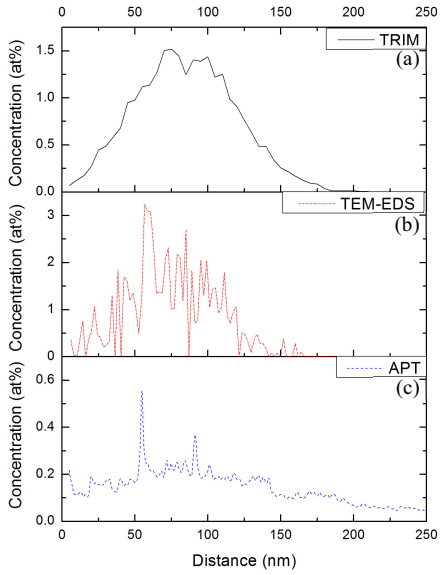

Figure 1: Xe distribution in CeO_2 (a) simulated using TRIM, (b) as-irradiated analyzed using TEM and (c) post irradiation annealed analyzed using APT.

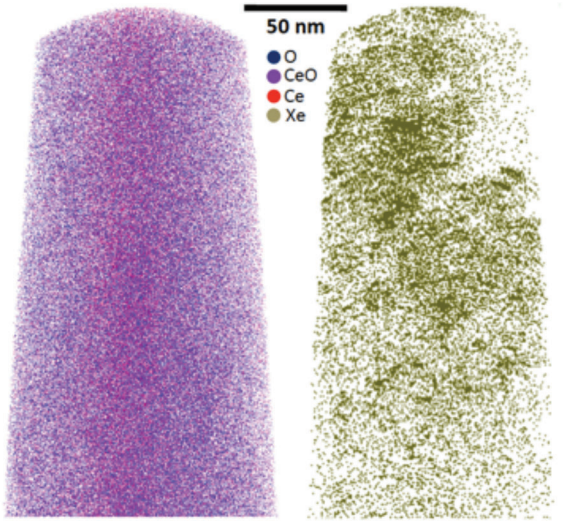

Figure 2: Atom probe reconstruction of post irradiation annealed Xe implanted CeO_2.

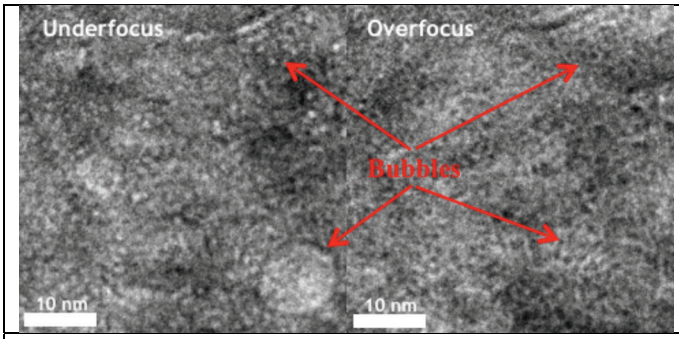

Figure 3: TEM micrograph depicting Xe bubbles in as-irradiated CeO_2.

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