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Mechanisms of Retention and Transport of Fission Products in Virgin and Irradiated Nuclear Graphite

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ABSTRACT:

To fulfill the design and licensing requirements for advanced reactors, the U.S. Department of Energy (DOE) is specifically interested in targeted research to quantify the mechanisms of fission product (FP) transport and retention in graphitic grades that will likely be used in next-generation very/high temperature reactors (V/HTRs), fluoride salt-cooled high temperature reactors (FHRs) and molten salt reactors (MSRs). In this project, we propose an integrated experimental-computational approach to measure the diffusivities of seven fission products – Iodine (I), Cesium (Cs), Krypton (Kr), Strontium (Sr), Ruthenium (Ru), Silver (Ag), and Europium (Eu) in five nuclear (virgin/irradiated) graphite grades – HOPG, POCO (ZXF-5Q/AXF-5Q), NBG-18, PCEA, and IG-110, as well as in a representative matrix graphite grade. Using a suite of experimental analysis techniques and multiscale simulations, we will elucidate the pertinent transport/retention mechanisms of the FPs in the selected graphite grades.

The proposed project entails three tasks; in the first task, non-radioactive species will be implanted into virgin/irradiated graphite samples for simulating realistic FP trajectories. The penetration-concentration profiles of the FPs will be measured using secondary ion mass spectroscopy (SIMS) capable of better than ppm sensitivity. Independent corroboration with the simulated profiles from Rutherford Backscattering (RBS) will strengthen the fidelity of the results obtained using SIMS. Careful analysis of the concentration gradient of the FP species at dilute concentrations will provide the tracer diffusion coefficients for temperatures ranging from 500 to 1800°C. In the second task, the microstructure and the chemical bonds of the samples will be evaluated using a host of electron microscopy, diffractometry, porosimetry, spectroscopy, and tomography techniques. Retention, location, and transport of FPs in irradiated HTR/MSR graphite grades will be examined using 3D/4D X-ray tomography (including synchrotron), Brunauer, Emmett and Teller (BET) analysis, and X-ray photoelectron spectroscopy (XPS) to gain a critical understanding of pore connectivity, pore size, and the distribution of sub-micron sized pores during FP release. High-resolution transmission electron microscopy (TEM) and tomography, electron energy loss spectroscopy (EELS), and energy dispersive spectroscopy (EDS) will be used for radioisotope and elemental mapping at the atomic level. In the third task, electron and x-ray tomography images will be used to generate realistic defect structures of the graphitic samples using inverse Monte Carlo techniques. Subsequent electronic-structure/atomistic and phase field simulations will provide a science-based predictive capability for elucidating the mechanisms of transport/retention of FPs on realistic nuclear graphitic structures.