Radiation-Enhanced Diffusion of Ag, Ag-Pd, Eu, and Sr in Neutron Irradiated PyC/SiC Diffusion Couples

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ABSTRACT:

The objective of this work is to provide diffusion kinetics that reflect the observed transport of select fission products across intact tristructural-isotropic (TRISO) fuel silicon carbide (SiC) layers. The determination of accurate diffusion kinetic data will support the development of fuel performance models necessary to ensure safe and efficient reactor operations. The historic diffusion kinetics available for modeling release of fission products have primarily been obtained by fractional release measurements from irradiated TRISO fuel particles. The fractional release measurements, for Ag in particular, have resulted in a large range in diffusion kinetics and are challenged by the indirect nature of the diffusion analysis, limited understanding of boundary conditions and the complex, time-dependent nature of fuel irradiations and fission product yield. This leads to uncertainty in inputs of the diffusion kinetics in fuel performance models. Many recent efforts have been focused on providing diffusion kinetics of fission product elements in SiC through surrogate diffusion systems. These studies have provided valuable insight on the influence of grain boundaries, diffusion source effects, and diffusion in irradiation-damaged substrates. However, the comparison of surrogate diffusion couple results shows inconsistency in diffusion kinetics and transport mechanisms. This leads to multiple knowledge gaps in the current understanding of diffusion of fission products in the TRISO fuel system, including the influence of neutron radiation on diffusion, the pyrocarbon (PyC)/SiC interface, and the SiC microstructure.

The scope of this work includes investigating diffusion of Ag, Ag-Pd, Eu, and Sr in PyC/SiC diffusion couples fabricated using identical coating systems to those used to coat TRISO fuel particles at Oak Ridge National Laboratory (ORNL) for the Advanced Gas Reactor (AGR) Fuel Development and Qualification Program. In lieu of spherical particles, diffusion couples containing a PyC/SiC construction will be developed with a wafer geometry to facilitate depth profiling by Rutherford Backscattering Spectrometry (RBS). Depth profiling will allow for determination of diffusion kinetics for the systems of interest, while studying a system with the same layer properties as TRISO fuel. Both thermal and neutron radiation-enhanced diffusion effects will be studied. The influence of neutron irradiation will be studied by irradiating diffusion couples in the High Flux Isotope Reactor (HFIR) at high-temperature gas-cooled reactor (HTGR) relevant temperatures (~1100°C) to SiC doses of 0.5 and 1 dpa. Parallel diffusion couple samples will be subjected to identical thermal schedules to determine the magnitude of neutron radiation-enhanced diffusion in the Ag, Ag-Pd, Eu, and Sr. The inclusion of the Ag-Pd condition serves to highlight the potential influence of Pd-assisted Ag transport. Diffusion couples will be fabricated with multiple variants to close the knowledge gaps associated with SiC microstructure and the PyC/SiC interfacial effects by varying process variables during PyC/SiC diffusion couple fabrication. High temperature thermal diffusion (>1500°C) will be studied to understand transport behavior at accident and margin temperatures and provide context to the post-irradiation safety testing examination reported for the AGR irradiation campaigns. Select diffusion couples will also be investigated by scanning transmission electron microscopy to identify the nature of the fission product species interactions with the PyC/SiC interface and SiC microstructural features to provide further insight on the active diffusion mechanisms. This work will be completed through access provided by the National Science User Facilities to the world-class facilities of the Michigan Ion Beam Laboratory, HFIR, the Irradiated Materials Examination and Testing hot cell facility, and the Low Activation Materials Development and Analysis laboratory.