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Structural Characterization of Fission Products in Irradiated TRISO Fuels using Transmission Kikuchi Diffraction, Transmission Electron Microscopy, and Synchrotron X-ray Absorption Spectroscopy

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The design philosophy of tristructural isotropic (TRISO) coated nuclear fuel particles is to retain radionuclides over a wide range of operational parameters (e.g., power, burnup, and temperature) to ensure safe operation of high-temperature gas-cooled nuclear reactors. Previous studies have indicated the release of metallic fission products (Ag, Pd) at temperatures that simulate accident scenarios [1, 2]. The exact mechanism for this release that may involve fission product/SiC interactions remains elusive and requires further study. In this work, we used a multi-faceted approach to analyze the effect of post-irradiation heating on SiC integrity and fission product transport in neutron-irradiated TRISO fuel from the Advanced Gas Reactor Fuel Development and Qualification first irradiation experiment (AGR-1) programs [3]. We combined synchrotron x-ray absorption spectroscopy (XAS), transmission Kikuchi diffraction (tKD), and scanning/transmission electron microscopy (S/TEM) with energy dispersive x-ray spectroscopy (EDS) to gather a comprehensive dataset on the material.

The TRISO particles utilized in this work were fabricated at Oak Ridge National Laboratory [4] and irradiated in the Advanced Test Reactor at Idaho National Laboratory to temperatures reaching 1250°C and a burnup of 19.6% fissions per initial metal atom [5]. After irradiation selected compacts were heated to high temperatures for margin temperature testing to simulate in-reactor accident scenarios [6]. The SiC shells from three selected particles (one as-irradiated particle, one heated at 1700°C for 300 hours, and one heated at 1800°C for 600 hours) were isolated by deconsolidating the fuel compact [7]. This was followed by the breaking of the particles, burning off the carbon layers in air at 750°C, and cleaning any exposed uranium or fission products from the SiC surface with hot nitric acid. XAS was collected on the isolated SiC shells at the Materials Research and Collaborative Access Team (MRCAT) insertion device beam line at the Advanced Photon Source [8]. The Low Activation Materials Development and Analysis (LAMDA) laboratory at ORNL includes a microscopy suite that allows for the examination of low-dose radioactive material, including neutron irradiated fuels [9]. Here, small lamella (liftouts) from the SiC and the IPyC/SiC and OPyC/SiC interfaces of each of the three shells were prepared and studied using S/TEM, EDS, and tKD to study the microstructure, dispersion, and phase and grain structure of the SiC and release products. Specimens were analyzed using an FEI Versa3D FIB-SEM DualBeam with Oxford Nordlys EBSD system for tKD, and an FEI Talos F200X system with “SuperX” 4-detector SDD-EDS system for S/TEM and EDS.

Preliminary findings confirm measurable fissile and fission product inventories within the SiC layers [6, 10]. XAS provided information on the average coordination chemistry of the material. It suggests the formation of uranium carbides, palladium silicides, and metallic silver. From S/TEM, we observed concentrated deposits of actinides and fission products along grain boundaries and at triple points within the SiC, an example shown in Figure 1, suggesting grain boundary diffusion as a dominant diffusion mechanism. Metal fission products were found co-located in the material. Uranium was found in

isolated pockets, co-located with plutonium, and was sometimes co-located with metallic fission products. Silver was only observed in the SiC particle annealed to 1700°C in small quantities. Although the grain size in the SiC increased from IPyC to OPyC interface, it remained fairly consistent between particles. With increasing post-irradiation temperature, uranium and palladium migrated further across the SiC layer of the TRISO, but the size of the precipitates decreased as a function of increasing distance from the IPyC. We use tKD to identify larger precipitates in the material to help draw conclusions about the precipitate formation and to confirm composition determined using XAS. Results from these three techniques will be discussed in greater detail in this presentation including their application to future versions of TRISO nuclear fuel [11].

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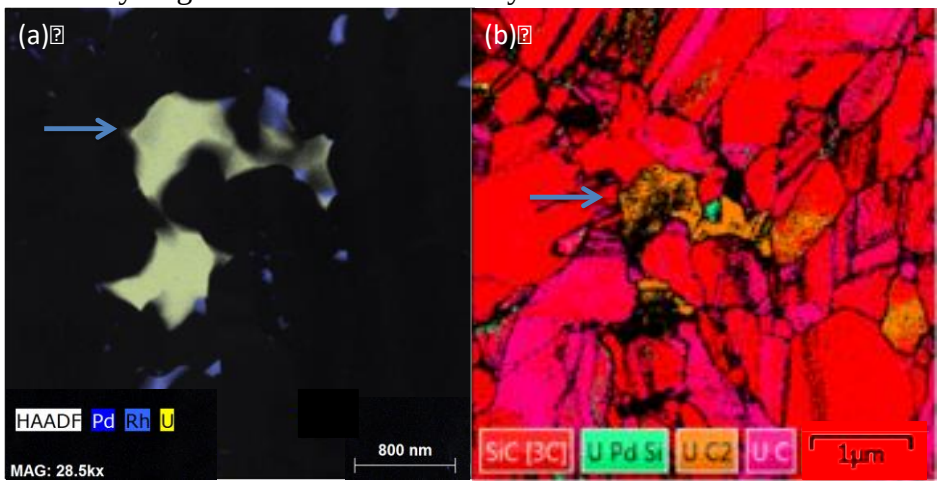

Figure 1. (a) STEM/EDS map of fission product precipitates at the boundaries of SiC grains. In this example, from the as-irradiated particle, uranium is shown both co-located with metallic fission products Pd and Rh, and on its own. (b) Is a tKD Euler micrograph of the same region, as indicated by the blue arrows. Preliminary results indicate UC_2 as the bulk uranium precipitate.