

Advanced electron microscopy study of fission product distribution in the failed SiC layer of a neutron irradiated TRISO coated particle

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Tristructural isotropic (TRISO) coated particle fuel has been designed for application in high-temperature gas-cooled reactors (HTGR). TRISO particles for the HTGR fuel development effort underway at Idaho National Laboratory (INL) and Oak Ridge National Laboratory (ORNL) consist of a two-phase uranium oxide-uranium carbide (UCO) fuel kernel, a carbon buffer layer, an inner pyrolytic carbon (IPyC) layer, a SiC layer, and an outer PyC (OPyC) layer [1]. The first in a series of irradiation experiments (AGR-1) clearly shows release of certain metallic fission products, e.g., Ag and Pd, through intact TRISO coatings, with Cs generally well retained [1]. No significant chemical interaction was observed between Pd and SiC for UCO TRISO coated particles, which retained Cs [2].

During post-irradiation examination of TRISO particles from AGR-1 Compact 5-2-3, gamma-activity measurements determined that Cs had been released by two specific particles. The fractional Cs and Ag releases from one of the two particles, Particle 523-SP01, were determined to be ~30% and ~85% respectively. Non-destructive three-dimensional X-ray tomography performed on this particle revealed that the SiC failure was localized to one specific region. Figure 1 (a)-(c) shows x-ray, optical, and scanning electron microscopy (SEM) images of the SiC failure in this particle. The failure mechanism was found to be SiC degradation through chemical reaction with Pd and U initiating at a SiC inner surface exposed by IPyC fracture [3].

To further examine the failure mechanism, fission product distribution and transport, scanning transmission electron microscopy (STEM) specimens were prepared using the focused ion beam (FIB) technique. Figure 1(d) shows the locations where STEM lamellae were sectioned by FIB. STEM and energy-dispersive X-ray spectroscopy (EDS) were conducted. In lamella 7, all precipitates close to the crack are rich in Pd and Si, ~63% of which contain a small atomic concentration of U. Approximately 63% of the Pd-Si and Pd-Si-U precipitates contain Cs, with concentrations ~0.20–1.43 at%. No Ag is evident in any of the precipitates. Ce and Cs have very close peaks in the EDS spectra; because the TRISO particle had no detectable Ce release and significant Cs release, all of the overlapping Cs and Ce peaks are considered Cs peaks. Further examination using EDS with better energy resolution is underway to obtain more accurate Cs concentrations. In lamella 8, most precipitates are composed of Pd and Si, with ~50% also containing a small atomic concentration of U. Approximately 36% of the Pd-Si and Pd-Si-U precipitates contain Cs, with concentrations ~0.20–1.99 at%. Only one precipitate (out of 36) rich in Pd and Ag was found, with 5.34 at% Pd and 2.02 at% Ag.

Figure 2 shows representative STEM images of lamella 10. In Figure 2 (a)–(d), precipitates numbered and arrowed were subjected to EDS investigation. Precipitates in the SiC layer are Pd-Si-U, Pd-Si, Pd-U or Pd. Approximately 30% of these precipitates contain Cs, with concentrations usually in the range ~0.40–0.80 at%, highest concentration being 2.20 at%. No precipitates containing only Cs were

observed. Ag and/or Cd containing precipitates were identified by EDS point scans 1–8 and 15–16 at grain boundaries or triple junctions. The Ag concentration in these precipitates is 0.22–7.95 at%, and the Cd concentration is 0.20–1.31 at.%. Ag and/or Cd may exist by themselves or coexist with Pd. The number of Cs containing precipitates is significantly higher than that of Ag or Cd containing precipitates; however, Cs concentration in the precipitates is usually significantly lower than Ag or Cd concentration. Dark areas in the SiC layer (Figure 2(d)) were identified by EDS to be pure carbon. Bright precipitates in the pure carbon areas (17, 18 and 19 in (d)) were identified to be Pd₂Si or PdSi. These results confirm chemical reactions between Pd and SiC, forming Pd₂Si or PdSi and carbon. Note that Cs containing precipitates were seldom found and no Ag or Cd containing precipitates were found in other lamellae sectioned from other locations in the SiC layer of this TRISO particle.

In summary, a TRISO coated particle with ~30% Cs and ~85% Ag release had a major crack across the buffer layer and the IPyC layer. Areas in the SiC layer close to the crack tip were corroded by Pd, forming pure carbon areas and Pd₂Si or PdSi; such corroded areas may provide pathways for Cs migration. Ag and/or Cd containing precipitates were identified in the SiC layer only in locations close to the crack.

References:

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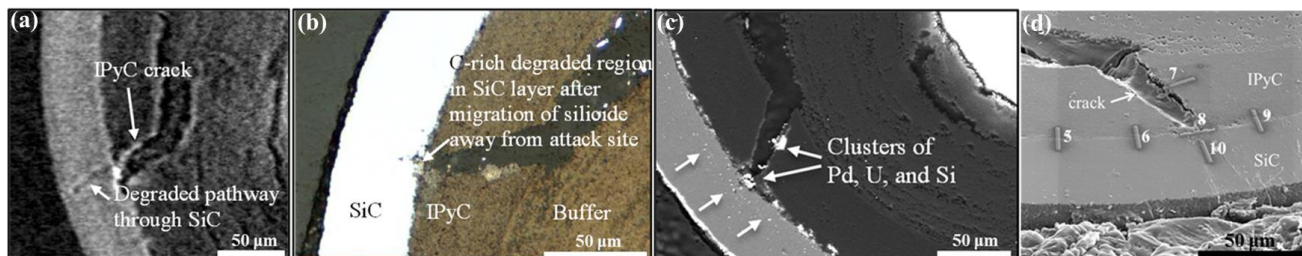


Figure 1. X-ray tomograph (a), optical micrograph (b), and backscattered electron composite image (c) showing localized degradation from Pd and U attack of the SiC layer where it was exposed by an IPyC crack; (d) SEM image showing locations where STEM lamellae were sectioned.

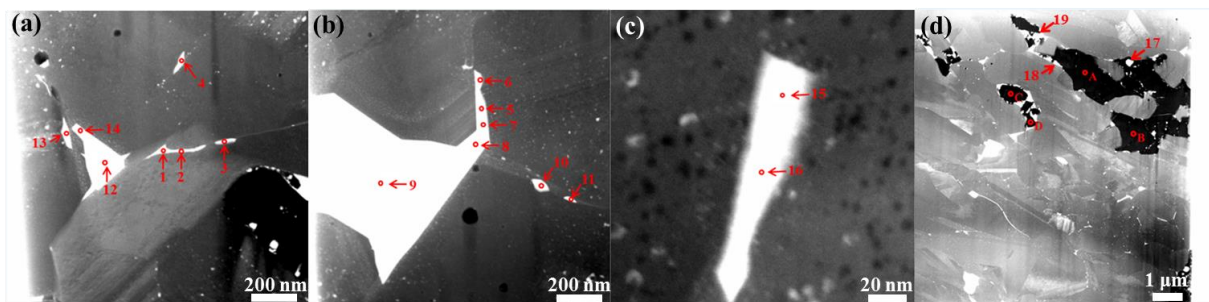


Figure 2. STEM images of STEM lamella 10 shown in Figure 1(d). Precipitates numbered and arrowed in (a)-(d) were subjected to EDS analysis. Dark areas in (d) are carbon, and letters A-D indicate locations for EDS analysis. Ag and/or Cd are detected in EDS point scans 1-8 and 15-16.