Characterization of Neutron-Irradiated Zr-1Nb-O Using Scanning Transmission Electron Microscopy

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Modern zirconium alloys show excellent water-side corrosion resistance, low hydrogen pickup, and exceptional irradiation-resistance during light-water reactor (LWR) operation as compared to Zircaloys [1]. Improved operational performance of these alloys boosted activities to increase current burnup limits for nuclear fuel rods from ~60 to ~70 GWd/t for LWRs. Burnup extension faces many design challenges from the licensing viewpoint, primarily related to high-burnup structure (HBS) formation as well as mitigation of pellet/cladding chemical and mechanical interaction during reactor transients. In order to succeed, a detailed description of nuclear fuel is needed. This description needs to be robust and accurate enough that computational models can be developed, modeling from nanoscale to engineering scale. State-of-the-art characterization techniques, such as analytical transmission electron microscopy (TEM), can elucidate evolved microstructure and local chemistry at the pellet/cladding interface (PCI) for the development of further experimental investigations and computational efforts. Thus, this study investigated the microstructural characterization of high-burnup nuclear fuel at the PCI with the presence of HBS using analytical TEM. Several TEM samples were prepared from a high-burnup nuclear fuel using focused ion beam milling. Phases present were identified via electron-diffraction patterns, and the local chemistry was investigated by energy-dispersive x-ray spectroscopy (EDS). Findings will be informative for the development of materials computational models, especially for mesoscale approaches.

Results

Characterization of the PCI region revealed the presence of monoclinic and tetragonal zirconium oxide phases formed at the fuel/cladding interface. Monoclinic phase is formed at the zirconium side of the PCI region, and the tetragonal phase was formed at the fuel side. This suggests monoclinic phase was formed prior to the formation of the tetragonal phase. Elemental maps, as shown in Figure 1, showed that no single-phase uranium-zirconium-oxygen compound was detected in the PCI region, but interlocked separate zirconium oxide and uranium oxide phases were observed. Both zirconium and uranium oxides exhibited nanograin structure. Monoclinic grains were elongated while tetragonal grains were equiaxed. A notable observation, shown in Figure 1, was that zirconium oxide encapsulated fuel into small pockets, suggesting the diffusion of zirconium was the main driving force. Mo-Tc-Ru-Rh-Pd precipitates were found not only in the UO_2 fuel region, but also in the zirconium oxide region. The Mo-Tc-Ru-Rh-Pd precipitates are known as five metal precipitates in irradiated UO_2 fuels [2]. Measurement of a large Mo-Tc-Ru-Rh-Pd precipitate enabled the determination of the composition, such as 37 at% Mo, 8.6 at% Tc, 29 at% Ru, 1.4 at% Rh, and 14 at% Pd. It should be noted that almost all the Mo-Tc-Ru-Rh-Pd precipitates were associated with pores.

Conclusion

This study characterized the PCI region of a high-burnup fuel from an LWR using analytical TEM. Results revealed that the PCI region is a complex structure where zirconium oxide diffuses into oxide fuel. At high burnup, the pellet/cladding gap is completely closed by forming an interlocked diffusion layer (see Figure 1). This implies that thermal energy would be transported throughout PCI layer, and the gap thermal uncertainty would be null. Modeling tools must incorporate this effect by including the thermal properties of zirconium oxide phases. However, the thermal conductivity of zirconium oxide phases must be incorporated to heat-transfer models. This study offers a clear description of the PCI layer that can easily be included in mesoscale computation. In addition to that, the nanograined microstructure of the PCI shows the feasibility of applying micromechanical test methods to investigate the mechanical properties of fuel.

References

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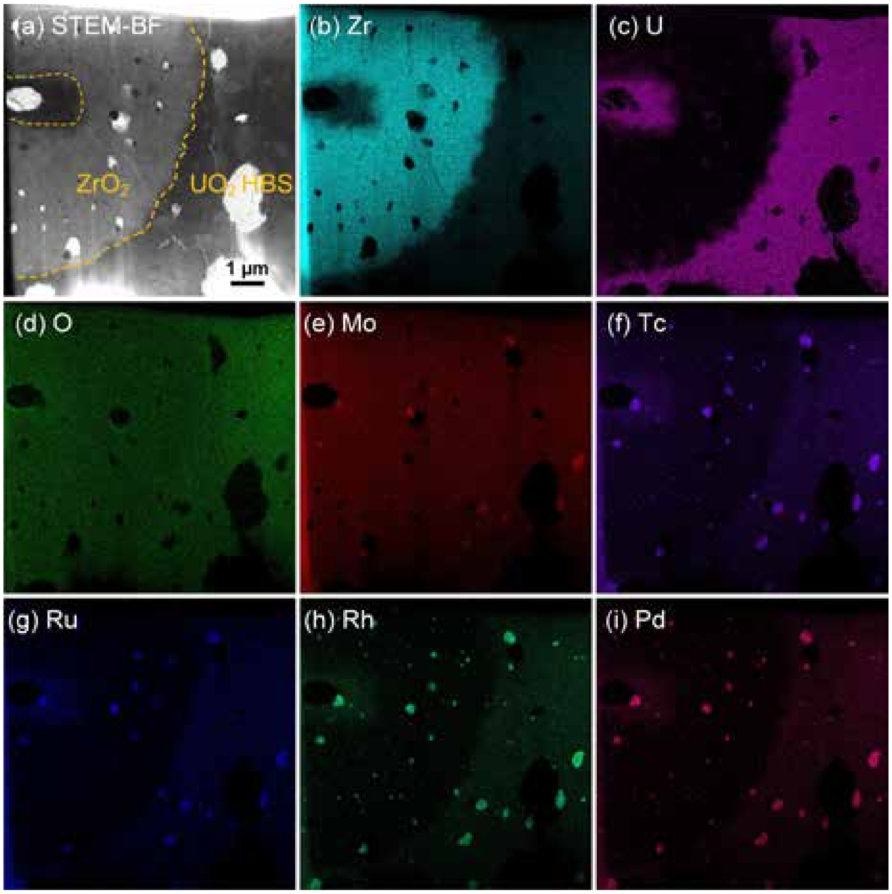

Figure 1. STEM-EDS results of the PCI-UO_2 fuel interface: (a) STEM bright-field image showing the ZrO_2 region and the UO_2 region; both regions contain pores. (b) to (i) Elemental maps of Zr, U, O, Mo, Tc, Ru, Rh, and Pd, respectively.

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