

A Challenge to Multivariate Statistical Analysis: Spent Nuclear Fuel

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Nuclear fission accounts for most of the non-polluting, non-fossil-fuel electrical power in the world. Higher burnup of fuel – that is, using a given fuel bundle for a longer time to produce more power – reduces the uranium resources needed, greatly enhances the economics of nuclear electricity, and reduces the amount of spent fuel for disposal. However, as the fuel burnup progresses, the fission process builds up large atomic fractions of fission products, consisting of most elements in the central region of the periodic table, in the fluorite UO₂ matrix; and, a fuel/clad interaction (FCI) layer forms at the interface between the oxide fuel and the Zircaloy cladding. Providing a scientific basis for understating fuel behavior in the high burnup regime requires detailed characterization of high-burnup uranium fuel. We have used X-ray spectrum imaging (SI) in SEM and STEM to analyze high-burnup (irradiated for 7 eighteen month long cycles to average burnup of 72 GWd/MTU) fuel from the H.B. Robinson pressurized water reactor. Multivariate statistical analysis (MVSA) is irreplaceable for understanding the extremely complex chemistry found.

A polished fuel slice was X-ray mapped using a 150 mm² Oxford SDD on the ORNL FEI Versa DualBeam FIB-SEM [1]. Special work controls were needed to protect the operators during sample loading, because the ~50 μm thick, ~1 cm×1 cm square sample produced ~60 rad/hour β-radiation, and produced ~60% downtime on the SDD with the electron beam blanked. X-ray maps indicated the presence of the UO₂ matrix, Zr clad, FCI layer, sub-surface Xe-bubbles, and precipitates containing refractory metals (Mo, Ru, Pd, etc.). A detailed view of the fuel region is shown in Figure 1. MVSA analysis [2] finds clear differentiation between the regions, and shows the sub-surface Xe bubbles, in particular, very clearly. MVSA also reduced the noise in the refractory-metal K-lines compared to point or regional analysis, making lower-level contributions in the precipitates (i.e., Tc) visible.

A FIB section was prepared using a lead-shielded FEI Quanta 3D DualBeam, and then X-ray SIs were taken in the ORNL LAMDA FEI F200X Talos S/TEM tool [3]. This S/TEM uses the 4-detector SuperX system [4]. Although the FIB liftout (~100 nm×10 μm×10 μm) showed measurable α (~2,000/sec) and β (~20,000/sec) counts and required radiological control technician support for sample loading/unloading, no dark counts were detected on the SDD system. A low-magnification STEM image and X-ray maps (Figure 3) shows fine UO₂-rich grains and large metallic precipitates. Large (~2 μm) pores are prior-Xe bubbles. MVSA (Figure 4) determines that the metallic precipitates are not a single uniform phase, and finds low levels of Tc, which was not found from gross X-ray mapping analysis. High-magnification maps show significant sub-structure in the precipitates. The complex mélange of many different elements, arrayed in several populations of precipitates and bubbles, make high-efficiency X-ray mapping and MVSA data treatment necessary to determine the microstructures encountered. [5]

References:

- [1] C.M. Parish et al., *Microsc. Microanal.*, **V21**(Suppl.3) (2015), P. 1003-1004.
- [2] P.G. Kotula, et al., *Microsc. Microanal.* **V9** (2003), P. 1-17.
- [3] C.M. Parish, ORNL Report MT3FT-15OR0204122, <http://www.osti.gov/scitech/biblio/1234344/>.
- [4] P. Schlossmacher, et al., *Microscopy Today*, July (2010), P. 14-20.
- [5] Supported by Laboratory Directed R&D funds at ORNL. This research was performed, in part, using

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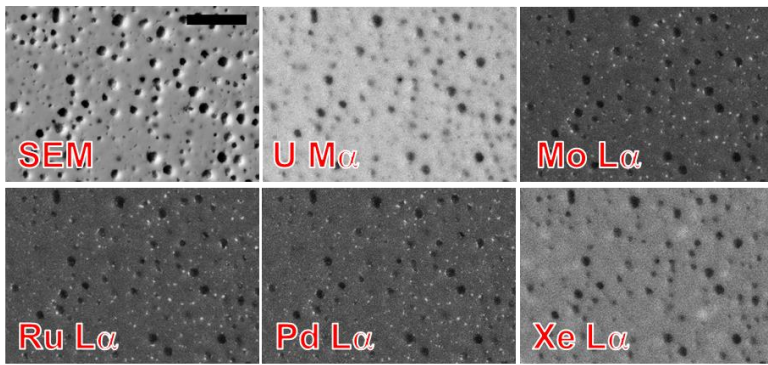


Figure 1. SEM image and X-ray maps of the high-burnup structure. Scale bar in SEM image = 10 μm.

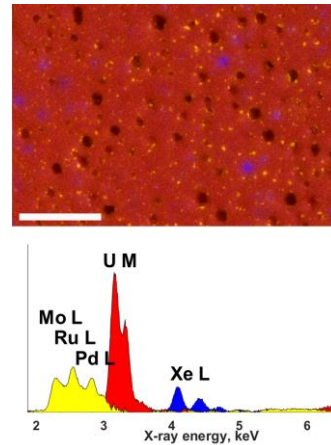


Figure 2. MVSA score images and loading spectra, showing precipitates and sub-surface Xe bubbles. Scale bar=10 μm.

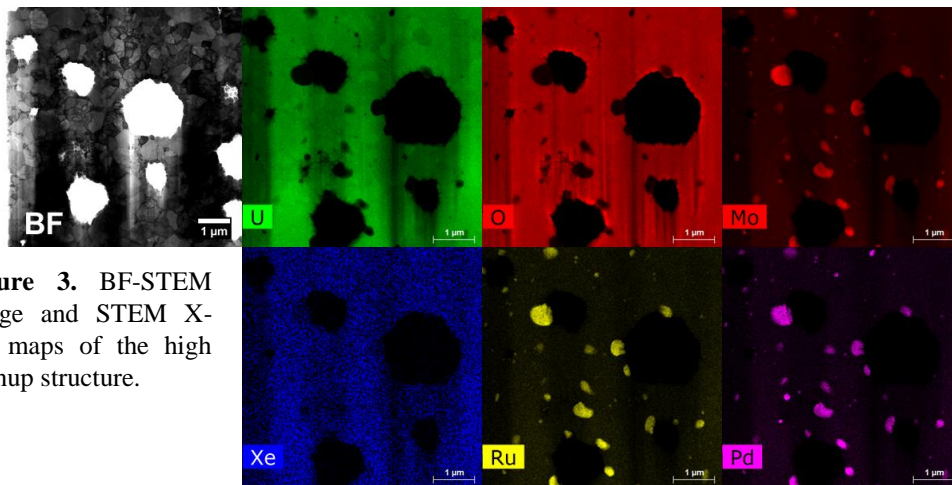


Figure 3. BF-STEM image and STEM X-ray maps of the high burnup structure.

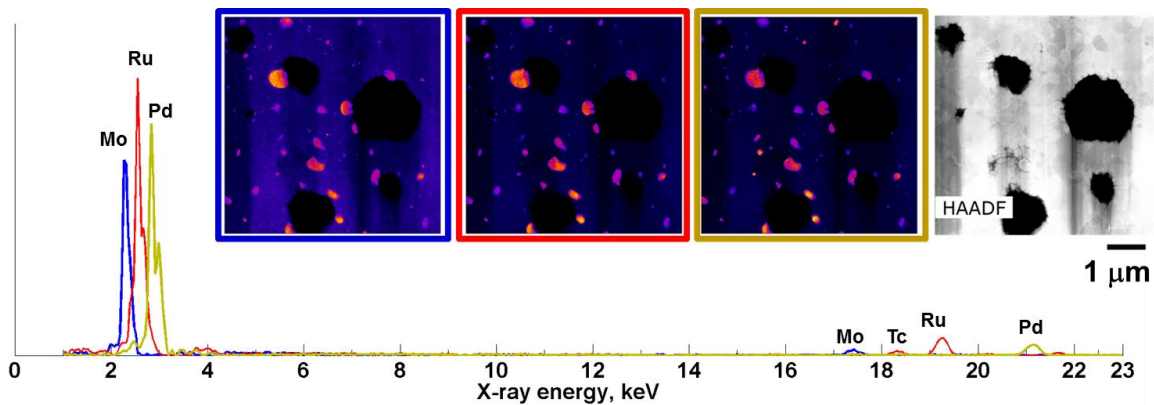


Figure 4. MVSA score images and loading spectra of refractory metal precipitates in the high-burnup fuel. The blue, red, and yellow components show different elements (Blue: Mo, Red: Tc-Ru, Yellow: Pd).