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Electron Probe Microanalysis of Irradiated U-Pu-Zr Fuel with Added Minor Actinides

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Among the barriers to developing a disposal strategy for nuclear fuel is the presence of long-lived minor actinides (Np, Am, and Cm) that result from the irradiation of oxide fuels in a light water reactor. One method for addressing this problem is to separate minor actinides (MA) from the fuel and transmute them into shorter-lived, less toxic nuclides. One way this can be accomplished is by adding the MA to U-Pu-Zr metallic fuels and irradiating them in a fast breeder reactor. U-Pu-Zr fuels are used because of the relatively high solidus temperature and because of the tolerable interaction between the fuel and cladding.

While the behavior of U-Pu-Zr fuels has been well-studied [1, 2], there are far fewer studies involving the addition of MA to such fuels [3, 4]. Concerns about the addition of MA to fuels include the idea that the solidus temperature can be lowered due to the redistribution of elements during irradiation, thus resulting in degradation of the cladding.

As part of the FUTURIX-FTA experiment to test various compositions of transmutation fuel, 34.5U-28.1Pu-3.8Am-2.1Np-31.5Zr (wt. %) fuel was irradiated to 9.5 at% fissions per initial metal atom in the Phénix fast reactor. Electron probe microanalysis (EPMA) was used to analyze the distribution of fuel elements across the diameter of the fuel, as well as to analyze secondary phases formed within the fuel and as a result of fuel-cladding interaction. EPMA is an ideal instrument to use to study irradiated fuel due to its superior peak resolution and peak deconvolution capabilities compared to that of a scanning electron microscope.

Figure 1 (a-b) shows a cross section of the irradiated sample with the diameter traverse and mapping area indicated. As can be seen in Figure 2, following irradiation, the U concentration is depressed in the fuel center and enhanced toward the cladding, while the Zr concentration is reversed. Np appears to follow U behavior. Element redistribution behavior has been observed in U-19Pu-10Zr fuel, though the specific pattern of redistribution depends on whether the temperature of the fuel center exceeds 657^{o}C [5]. Between approximately 1000-2000 μm, there is extensive secondary phase precipitation, where rare earth elements are concentrated, sometimes with Am accompanying (Figure 1 (c-d)). The phase approximates Nd_7Pd_3, with extensive substitution of rare earth elements and actinides for the Nd site and Rh for the Pd site, similar to that found by Brémier et al. [4]. Figure 3 shows phase separation that occurs between 2500 and ~3000 μm. The composition of this phase approximates (U, Pu, Np)Zr_2.

Use of the software package, Probe for EPMA's CalcImage modal analysis feature allows the quantitation of Figure 3 with respect to computing the modal abundance of phases present. A weighted average of the components in the phases suggests that U concentration roughly doubles from the fuel center to the periphery and that Zr concentration falls by approximately 20% between the center of the fuel and the periphery.

These results show that similar to higher temperature U-Pu-Zr fuels, the major elements redistribute during irradiation such that Zr is enriched in the center relative to the as-fabricated composition. This is

fortuitous as Zr enrichment in the center will raise the solidus in the region with the highest temperature. In addition, while some Am does migrate to the cladding, much of it is immobilized as minor phases in the interior of the fuel. The addition of MA to metallic fuel appears to be a promising method to transmute these elements.

References:

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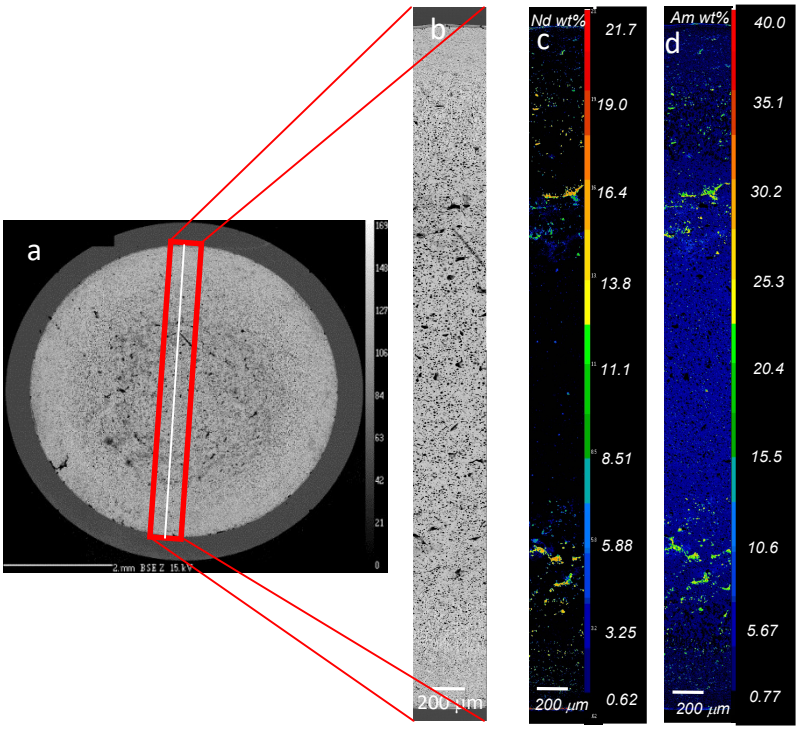

Figure 1. a) Backscattered electron image of fuel sample showing path of quantitative spot measurement (white line) and region of X-ray map collection (red box). b) Backscattered electron image showing X-ray map collection region. c-d) X-ray map showing Nd- and Am-rich precipitates.

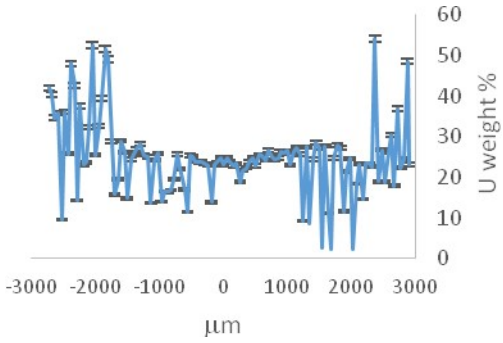

Figure 2. Quantitative U traverse showing decreased U in the fuel center with greater concentrations toward the mid-radius and cladding

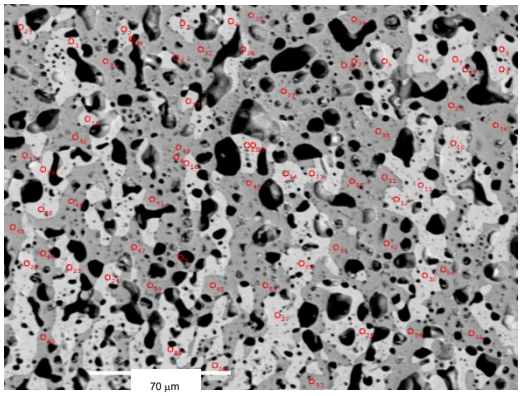

Figure 3. Backscattered electron image showing phase separation. Marked locations indicate locations for quantitative analysis to determine the phase compositions.

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