

**MATERIALS ENGINEERING
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“The Crystallographic Evolution in the Uranium-Zirconium System”

By

Walter J. Williams

Purdue MSE PhD Dissertation

Advisor: Professor Maria Okuniewski

ABSTRACT

Metallic uranium-zirconium (U-Zr) nuclear fuel is a primary candidate for future fast reactors. The U-Zr system has been studied for decades with thousands of fuel pins being irradiated, yet the phase boundaries and lattice evolution with respect to temperature and composition remain poorly quantified. Historic engineering scale testing has resulted in empirical models for fuel evolution and subsequent fuel performance. However, these historic tests are on a convoluted system, consisting of dynamic temperatures, evolving thermal gradients, varying irradiation damage and damage rates, evolving compositions via fission and redistribution of primary constituents, and morphological evolution. This system proves exceedingly difficult to describe mechanistically due to the coexistence of various intertwined thermodynamic driving forces (e.g., temperature, composition, fluence, and fission rate which all vary concurrently). The driving forces influence the manifestation of the primary life-limiting phenomena present within the U-Zr system, specifically fuel-cladding mechanical interaction, fuel-cladding chemical interaction, fuel swelling, and fuel constituent redistribution. Although the phenomena present in the U-Zr system are known and qualitatively described, they are lacking in fundamental descriptions due to the historic inability to deconvolve the effects of temperature, composition, and fission rate. This study evaluates the current understanding of U-Zr fuel swelling and constituent redistribution in a uniquely quantified manner using Phenomena Identification and Ranking Tables.

In response to these findings, a novel separate effects irradiation test vehicle, housing uniquely fabricated U-Zr alloys, was proposed, developed, and successfully fabricated to provide the community with a means to decouple temperature, composition, initial microstructure, and fission rate. Initial out-of-pile characterization was conducted with scanning electron microscopy, transmission electron microscopy, and neutron diffraction with in-situ heating on various U-Zr alloys (U- 6, 10, 20, and 30 wt.% Zr). This work quantifies the initial microstructure throughout the fabrication process and the thermal response of the material. Results include the phase morphology, phase boundaries, absolute lattice parameters, and lattice specific coefficients of thermal expansion. The phase boundaries identified in this study were then used to develop a new U-Zr phase diagram. The isolation of thermal and compositional dependencies furthers the understanding of the fuel system and can be used to increase fuel longevity.



School of Materials Engineering

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