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Title: 2022 Accomplishments Report Template

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2022 Accomplishments Report Template

1. **Title (from outline):** Fabrication and Thermophysical Properties Evolution in Chromium-doped UO_2
2. **Principal Investigator:** Adrien J. Terricabras
3. **Team Members/ Collaborators:** Joshua T. White, Scarlett Widgeon Paisner, Darrin D. Byler.
4. **Introduction:** Chromium-doped UO_2 fuels is an industrially important Accident Tolerant Fuel (ATF) due to the ability to increase the average grain size relative to standard UO_2 . However, there is a lack of knowledge of the thermophysical properties of these doped fuels, which is necessary for accurately modeling the in-pile performance. The work accomplished this FY focused on developing large grain Cr-doped UO_2 pellets as a function of Cr content and subsequently measuring the thermophysical and mechanical properties. The microstructural variations were also investigated to evaluate chromium formation within the microstructure of the sintered pellets.
5. **Project Description:** Both the Nuclear Regulatory Commission (NRC) and the Nuclear Energy Advanced Modeling and Simulations (NEAMS) program need thermophysical and thermomechanical data of the Cr-doped UO_2 system in order to validate information provided by industry. Because the data provided by industry is controlled as proprietary information, NEAMS is not able to use these data sets to develop codes to study the performance. To make this data widely available, LANL is working on fabricating Cr-doped, large grain UO_2 pellets and has collected measurements on the thermophysical properties. In particular, the thermal conductivity is of interest due to the high importance of this property for the safety of LWR fuels. Although the fabrication of the industry doped fuel concepts is proprietary, LANL has spent significant efforts in past years to develop a fabrication procedure to produce analogous samples. This includes carefully controlling the sintering atmosphere and chromium content to fabricate fuels with large average grain sizes, which has been found to be $> 28 \mu\text{m}$ and is dependent on the Cr concentration. The information from these measurements will be fed into modeling efforts through NEAMS to understand the performance of these fuels under irradiation.

There has been particular interest on the location of the Cr after sintering, which has been hypothesized to either accumulate at the grain boundaries or may be incorporated into the UO_2 lattice. The former has been inferred from XRD patterns, which is seen as a contraction of the lattice parameter. The Cr content, oxidation state, and location have implications on the fission gas release of the doped fuels upon irradiation. The work accomplished here will be unified into fuel performance code simulations of the doped fuels to assist in validating commercial codes provided by industry partners.

Accomplishments: The work supported under this work package has resulted in three milestone reports, M3FT-22LA020201018 (submitted March 2022), M3FT-22LA020201013 (submitted March 2022) and M2FT-22LA020201011 (to be submitted September 2022). The work accomplished here involves sintering of Cr-doped UO_2 pellets with Cr concentrations ranging from 750 to 7800 ppm Cr. Inductively coupled plasma mass spectrometry (ICP-MS) was collected before and after sintering to determine any Cr loss during sintering. The sintered pellets have densities > 94.5 percent theoretical density (%TD), and post sintering X-ray diffraction (XRD) patterns show only peaks from UO_2 . There is a small shift towards lower angles with increasing Cr content, which indicates a contraction of the lattice with higher Cr contents. This indicates that Cr is being dissolved into the UO_2 lattice as opposed to forming a secondary phase. Scanning electron microscopy (SEM) images and energy dispersive X-Ray (EDX) spectroscopy show that at higher Cr contents, Cr_2O_3 does precipitate out as a secondary phase and indicates that Cr is only dissolved into the lattice up until a threshold level, after which the secondary phase is seen. The precipitates are found within the grains as opposed to being located at the grain boundaries.

The average grain size in the doped UO_2 sintered pellets was measured and show increases in the grain sizes with increasing Cr content, as is shown in Figure 1. The 750 and 2500 ppm Cr-doped pellets revealed grain sizes of $28.2 \pm 2.3 \mu\text{m}$ and $41.5 \pm 3.8 \mu\text{m}$, respectively. For comparison, the average grain size of standard, undoped UO_2 is $\sim 10 \mu\text{m}$. The sintered pellets were used to measure heat capacity, thermal expansion, density, and thermal diffusivity of the Cr-doped materials. These measurements were used to calculate the thermal conductivity as a function of temperature and were compared to that of standard UO_2 published in 2008 by Fink. The results show that the thermal conductivity decreases at higher temperature and likely results from the phonon scattering due to Cr dopants in the

UO₂ lattice. The results are shown in the attached Figure 2. Doped and undoped large grain UO₂ were fabricated and shipped to High Flux Isotope Reactor (HFIR) for irradiation tests.

6. **Publications:** Two manuscripts are in preparation that details the fabrication and thermos-physical properties of Cr-doped UO₂ pellets as well as the Cr speciation in Cr-doped accident tolerant fuels using XAS.
7. **Write one sentence on why this project is important.** Cr-doped UO₂ shows an increase in grain size and also shows changes in the thermal conductivity relative to UO₂, which will be used to model in-pile performance.
8. **Photos/ Figures/ Graphics.**

JTWhite_1: Grain morphologies of a) 750 ppm and b) 2500 ppm Cr-doped UO₂ as observed under SEM following thermal etching.

JTWhite_2: Thermal conductivity evolution of 750 ppm and 2500 ppm Cr-doped UO₂ from 50 to 1000 °C