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ANALYSIS OF PRELIMINARY PIE RESULTS FOR PVD COATED U-7MO DISPERSION FUEL PLATES IRRADIATED IN THE EMPIrE EXPERIMENT

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ABSTRACT

This paper offers a preliminary evaluation of the recent post-irradiation-examination (PIE) results collected for the low enriched uranium (LEU)-molybdenum (U-Mo) dispersion fuel test, EMPIrE, with emphasis on PVD coated U-7Mo particles. The fuel swelling up to a burnup of ~80% U-235 was stable and in the expected range of values, as compared to other irradiation experiments to similar burnups. Plate-averaged non-destructive examination results did not reveal a difference in swelling performance between the 700 series plates (non-heat treated particles) and the 800 series plates (heat treated particles). Preliminary destructive examination results found that the addition of a ZrN coating on the fuel particles resulted in a reduction in the fuel-Al interaction at high burnup, as determined by comparison of a limited number of cross-section images from EMPIrE to dispersion fuel plates without coating from previous tests. The preliminary destructive examination results indicate that the condition (thickness, level of coverage, etc) of the coating may impact the level of interaction layer thickness formation in the fuel.

1. Experimental Details

The EMPIrE (European Mini-Plate Irradiation Experiment) experiment was irradiated at the Advanced Test Reactor at Idaho National Laboratory, and consisted of 48 LEU miniplates (44 dispersion, 4 monolithic-type), the details of which have been presented elsewhere [1,2]. This report will focus on plate types 700(A and B) and 800 (A and B), as there are preliminary destructive examination (DE) results available to correlate to the non-destructive examination (NDE) swelling results. These plate types were all fabricated with LEU-7Mo powder from KAERI coated with ZrN using the physical vapor deposition (PVD) method. The nominal fabrication parameters for the 700 and 800 series plate types are detailed in Table 1.

Plate Series	Heat Treatment	Powder Size Distribution	Coating Batch	Coating Thickness	Examined Plates	
					As-Fabricated	Irradiated
700(A)	As-solidified	Zero Fines	LA1A	1.19±0.22[3] 0.86±0.16 [4]*	711 [3]	702, 712
700(B)	As-solidified	Modified	LA 8A	1.37±0.26[3]	717 [3]	
800(A)	Annealed	Zero Fines	LA4A	1.98±0.23[3] 1.59±0.29[4]*	819 [3]	818, 820, 821
800(B)	Annealed	Modified	LA3A	1.53±0.29[3] 1.32±0.29[4]*	803 [3]	

Table 1: Fabrication parameters. *coating thickness determined from as-fabricated powder

Fuel powder fines are defined as particles less than 44μm in diameter. The modified particle size distribution contains 10% fines by weight. Note that additional powder batches than that listed were used to fabricate 800(A) plate type, but the data included here is limited to the powder batch used to fabricate the plates that have DE results available.

2. Results

The preliminary results from both NDE (profilometry) and DE (optical microscopy) for the 700 and 800 series plates will be discussed in the following sections.

2.1 Non-Destructive Examination

Profilometry measurements were conducted on all EMPIrE plates, where ~750 thickness measurements per plate were collected. The calculated fission densities at each of these thickness measurements were corrected for fuel loading (U-Mo volume fraction) based on as-fabricated powder mass. The plate-averaged fission density and fuel particle swelling fraction was calculated for each plate, and are compared in Figure 1, where the error bars indicate the standard deviation of the data.

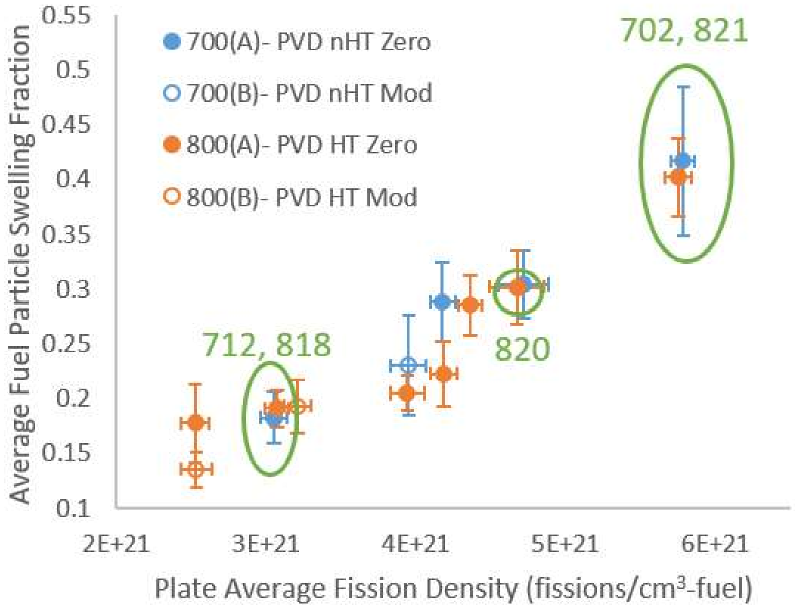

Fig. 1. Plate-average swelling fraction vs fission density for the 700 and 800 series plates irradiated in EMPIrE. The circled and numbered plates are those with available DE.

In order to calculate the fuel particle swelling, it is presumed that the change in plate thickness from the pre-irradiation values is due entirely to fuel swelling and that all pre-irradiation plate porosity is consumed prior to any plate thickness increases, after subtraction of the thickness due to oxide growth during irradiation. The local fuel loading is determined from calibrated radiography, in which Zr and Al foils were used as surrogates on the calibration plate for the ZrN coating on the fuel particles and Al matrix and cladding, respectively. The thickness of the former was selected based on a 2.5 Zr-wt% addition to the fuel powder mass, as measured previously by chemical analysis of ZrN PVD coated U-7Mo powder samples. The thickness of the latter was based on the nominal cladding and matrix thicknesses. It is planned to refine the analysis of the fuel swelling once the coating thickness for each powder batch in the experiment is known. In order to reduce the scatter in the data, thickness measurements with less than 80% of the nominal fuel loading were eliminated from the data set prior to determining the average value.

From comparison of the plate average fuel swelling values of the 700 series plates (non-heat treated, nHT) and 800 series plates (heat treated, HT) in Figure 1, there does not appear to be an impact of heat treatment on swelling behavior from the averaged data points. That said, analysis of higher-resolution data is ongoing, which will allow for greater clarification of the behavior of the 700 and 800 series plates. SEM analysis (currently unavailable) is required to examine the microstructure of the fuel particles in order to determine the impact of the heat treatment on fuel behavior. HT was added as a fabrication parameter in order to determine if grain refinement could be delayed by reducing the grain boundary density in the fuel, as grain boundaries serve as initiation sites for grain refinement.

2.2 Destructive Examination

A number of plates from the 700 and 800 series (all with zero fines) were selected for DE. Transverse samples were taken at the axial middle of the plate. A selection of optical micrographs is shown in Figure 2. Multiple images from each plate were analyzed to determine the interaction layer (IL) content.

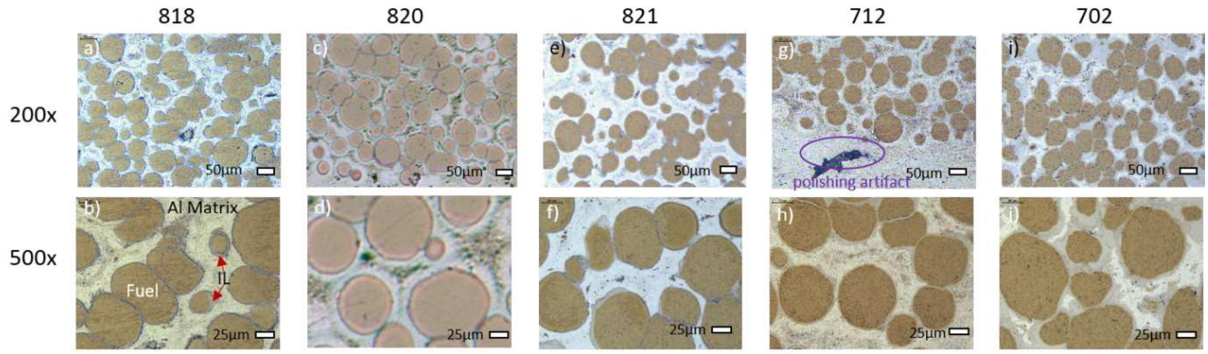

Fig. 2. Selection of micrographs from plates 818 (a&b), 820 (c&d), 821 (e&f), 712 (g&h), and 702 (i&j) at 200x and 500x magnifications. U-7Mo fuel is the dark phase, the Al matrix is the brightest phase, and the interaction layer is medium grey, as exemplified in image b.

Of particular note is that large fission gas bubbles and pores are not observed in any of the plates, even those irradiated to ~6E21 fissions/cm^3-fuel particle (821 and 702). This is indicative of excellent fuel performance. Additionally, there is minimal IL formation, which has been linked to poor fuel performance in the past. An exception to this is plate 702, which has extensive IL formation, although no impact on plate average fuel swelling was observed as a result, from comparison to the behavior of plate 821. IL fraction in each image was determined by the grid-counting method, where a fine grid is overlaid on the image, and an assessment of the phase present at each intersection of the grid is determined. Automated phase analysis of the images in Fig.2 was investigated, but due to the limited contrast of the images, results proved to be inconsistent. Automated phase analysis will be performed on the SEM images of the same samples when they become available to confirm the measured constituent volume fractions. The amount of IL is less than 5% up to 80% burnup (5.6×10^{21} fissions/cm^3), behavior similar to all but the very high burnup of the SELENIUM plate [5,6] , as shown in Figure 3. Plate 702 (~20-25% IL) shows a very different behavior at 80% burnup compared to the rest of the plates (~2-5%IL), much like that of the high burnup part of the SELENIUM plate.

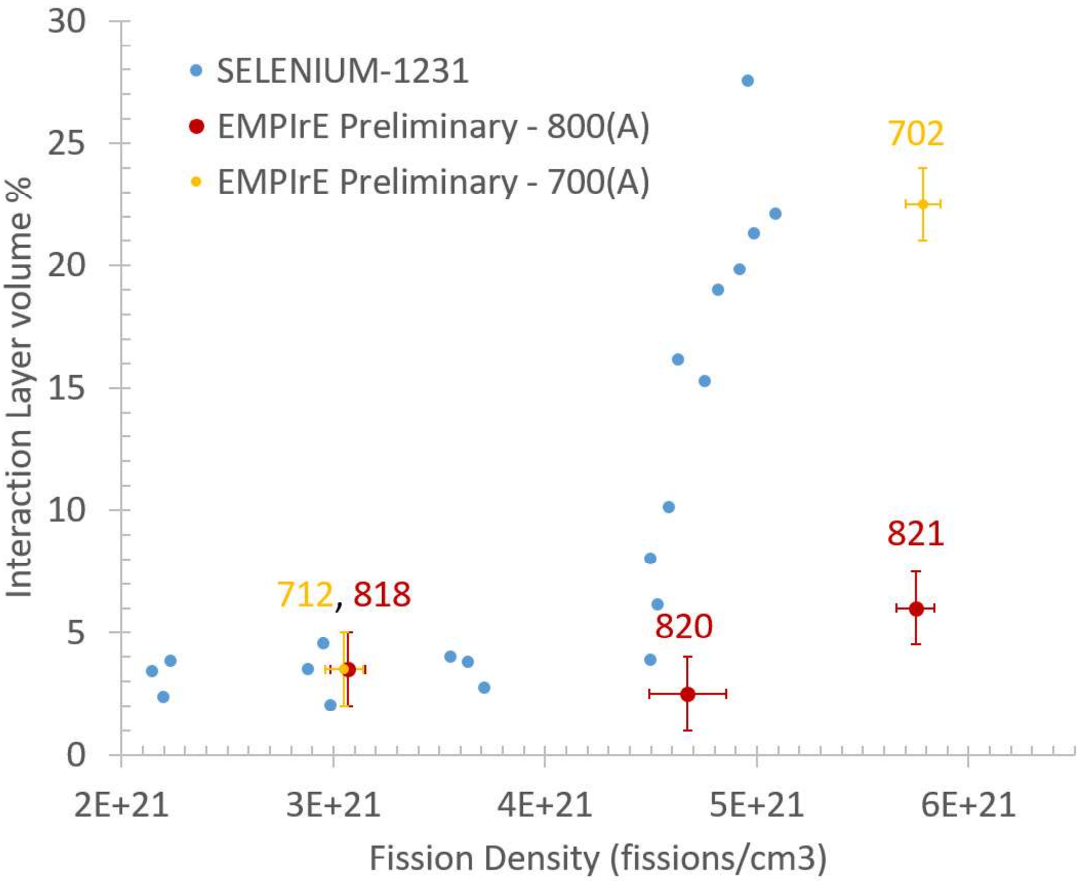

Fig. 3. Preliminary EMPIrE IL volume % compared to SELENIUM-1231. The y-axis error bars in the figure indicate the data variance.

3. Discussion

Of the several experimental variables included in the 700 and 800 series plates, the present analysis does not appear to reveal a significant effect on the average fuel particle swelling behavior. The fuel particle heat treatment and resulting grain growth was expected to delay the start of grain refinement and thus, the increase in gas bubble nucleation and growth. The delay in the grain refinement (and gas bubble nucleation) would be observed as a reduction in fuel plate swelling fraction. As the nHT and HT plates had similar plate average fuel swelling fractions, it is not apparent from this analysis that HT had a significant impact. Any evidence of a delay in grain refinement will need to be determined from SEM images, as the effect was too small to be apparent in plate average fuel swelling behavior. However, as mentioned previously, a higher resolution analysis is ongoing, which may reveal influences from the fabrication variables that are not readily apparent at the plate average scale.

At this point in the evaluation, based on preliminary results of a limited number of plates, there is an indication that coating thickness influences the amount of IL that forms. Coating thickness will be an important fabrication parameter to assess during ongoing analysis.This is based solely on the available optical microscopy, so this conclusion may be revised upon collection and examination of SEM images. Detailed examinations of samples taken from ZrN coated

UMo particles irradiated in experiment SELENIUM [5,6], employing STEM (scanning transmission electron microscopy), EELS (electron energy loss spectroscopy) and APT (atom probe tomography) [7,8] have established that the ZrN coating evolves into a bi-layer consisting of ZrN and ZrN-Al through the penetration of matrix Al into the ZrN coating, presumably by an ion mixing mechanism. This mechanism is dose dependent and therefore the extent of Al penetration at high burnup, whether it would reach the UMo core of the fuel particles and start the formation of UMo-Al interaction depends thus on the as-fabricated thickness of the ZrN layer. As shown in Table 1, the average layer thickness varies between fuel powder batches used in the EMPIRE experiment as well within each batch.

The amount of fuel particle surface area that was covered by the coating is an issue that needs more characterization, as that could also have led to the increased IL formation in plate 702 (if the powder had less coating coverage initially). As long as the IL does not form large fission gas bubbles (which are typically found at high fission rate) it does not affect the overall plate average fuel swelling. From this metric, all plates examined thus far had acceptable performance, as no large fission gas bubbles were visible. However, it is recommended to limit the IL formation to as small as possible, as it is difficult to determine how close plates are to forming large fission gas bubbles.

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