DRAFT

New Results from the Scanning Electron Microscopy Characterization of Fuel Plates Irradiated in the EMPIrE Irradiation Experiment

Dennis Keiser Jr., Daniele Salvato, Charlyne Smith, Tammy Trowbridge, William Hanson, Adam Robinson, and Irina Glagolenko
Idaho National Laboratory, P. O. Box 1625, Idaho Falls, ID 83415-6188 U.S.A.

Bei Ye, Laura Jamison, and Gerard Hofman
Argonne National Laboratory, 9700 S. Cass Ave., Lemont, IL 60439, USA

ABSTRACT

Irradiation of fuel plates in the European Mini-Plate Irradiation Experiment (EMPIrE) has been completed. The mini-plates (2.54 cm in width by10.16 cm in length) were irradiated in the Advanced Test Reactor (ATR) located at the Idaho National Laboratory (INL). The dispersion fuel plates were comprised of zirconium nitride (ZrN)-coated U-Mo fuel particles in an aluminum (Al) matrix, clad in Al-alloy. Several fabrication parameters were varied as a part of the experiment design to evaluate their influence on the fuel performance, including fuel powder heat treatment, coating methodology, coating thickness, fuel particle size distribution, Mo-alloying content, and fuel powder source. Of interest is the swelling behavior of the fuel plate, the effectiveness of the ZrN coating in mitigating fuel/matrix interaction during irradiation, and other microstructural developments in the fuel system. Samples from a selection of EMPIRE fuel plates have been microstructurally characterized along transverse cross-sections taken near the midplane using scanning electron microscopy (SEM). The results of the SEM characterization are presented here.

1. Introduction

The Material Management and Minimization (M3) Fuel Development program (earlier known as the Reduced Enrichment for Research and Test Reactors program) has been working with international collaborators over the years to develop low-enriched uranium (LEU) fuel to reduce the demand of high-enriched uranium (HEU) fuels currently used in research and test reactors throughout the world [1]. One fuel type being evaluated for use is a U-7Mo dispersion fuel with a barrier coating deposited on the U-7Mo particles that are encased in Al matrix to comprise the fuel meat which is then contained in Al alloy cladding [2]. Recently, the European Mini-Plate Irradiation Experiment (EMPIrE) experiment was completed in the Advanced Test Reactor (ATR) to test the performance of this fuel where the barrier coating is ZrN [3]. Microstructural characterization of the different fuel plates is performed using scanning electron microscopy (SEM). The SEM is used to produce secondary electron (SE) and backscattered electron (BSE) images, and composition analysis is conducted using energy dispersive spectroscopy (EDS) and wavelength dispersive spectroscopy (WDS). Recently, three new cross-sections from irradiated fuel plates (EMPI 0702, 0712, and 0905) have been microstructurally characterized by producing SEM/BSE images. In this paper, comparisons of these new images for some fuel plates will be made with those reported for three other EMPIrE fuel plates (EMPI 0818, 0820, and 0821) [4].

2. Experimental

During irradiation in the Advanced Test Reactor the EMPIrE fuel plates were oriented face-on to the core resulting in fairly, uniform flux profiles across the width of the fuel plates. Transverse cross-sections were produced near the midplane of the fuel plates at the Hot Fuels Examination Facility (HFEF) at the Idaho National Laboratory, and some of these cross-sections have been shipped to the Irradiated Materials Characterization Laboratory (IMCL) for SEM characterization. The cross-sections were mounted and polished for SEM analysis. Most recently, samples from three fuel plates have been characterized (EMPI 0702, 0712, and 0905). Details of the calculated fission density values for the SEM cross-sections and for the calculated surface heat fluxes for the complete fuel plates are presented in Table 1. Two of the fuel plates (EMPI 0702, 712) have ZrN coating on as-atomized U-7Mo produced by the Korean Atomic Energy Research Institute (KAERI). The ZrN coating was applied to the as-received powder using physical vapor deposition (PVD). All the fuel particles were between 44 and 125 µm in size. For EMPI 0905, the powder was produced by Framatome-CERCA and then heat-treated at 1,000˚C for 1 hour, and a ZrN coating was then applied using PVD. All the fuel particles were between 44 and 125 µm in size. For the microstructural characterization performed, both secondary electron (SE) and backscattered electron (BSE) images were produced from the polished samples.

Table 1: Calculated irradiation parameters for EMPI 0702, 0712, and 0905.

Sample	SEM Cross-Section Fission Density (FD) Minimum (fissions/cm^3 UMo)	SEM Cross-Section Fission Density (FD) Average (fissions/cm^3 UMo)	SEM Cross-Section Fission Density (FD) Maximum (fissions/cm^3 UMo)	Fuel Plate BOL Average Surface Heat Flux (W/cm^2)	Fuel Plate BOL Peak Surface Heat Flux (W/cm^2)
EMPI 0702	5.6 x10^{21}	5.7 x10^{21}	5.8 x10^{21}	448.9	513.4
EMPI 0712	2.9 x10^{21}	3.0 x10^{21}	3.2 x10^{21}	446.8	520.9
EMPI 0905	6.0 x10^{21}	6.1 x10^{21}	6.2 x10^{21}	474.0	544.1

3. 1 SEM Images

3.1.1 EMPI 0702

Figure 1 shows the microstructure observed for EMPI 0702. Some fuel/matrix interaction was identified, and intact ZrN coating could be found. ZrN particles were found in the matrix, and fission gas bubbles could be resolved within the fuel particles. As shown in (c), regions could be found where Al penetrated the ZrN.

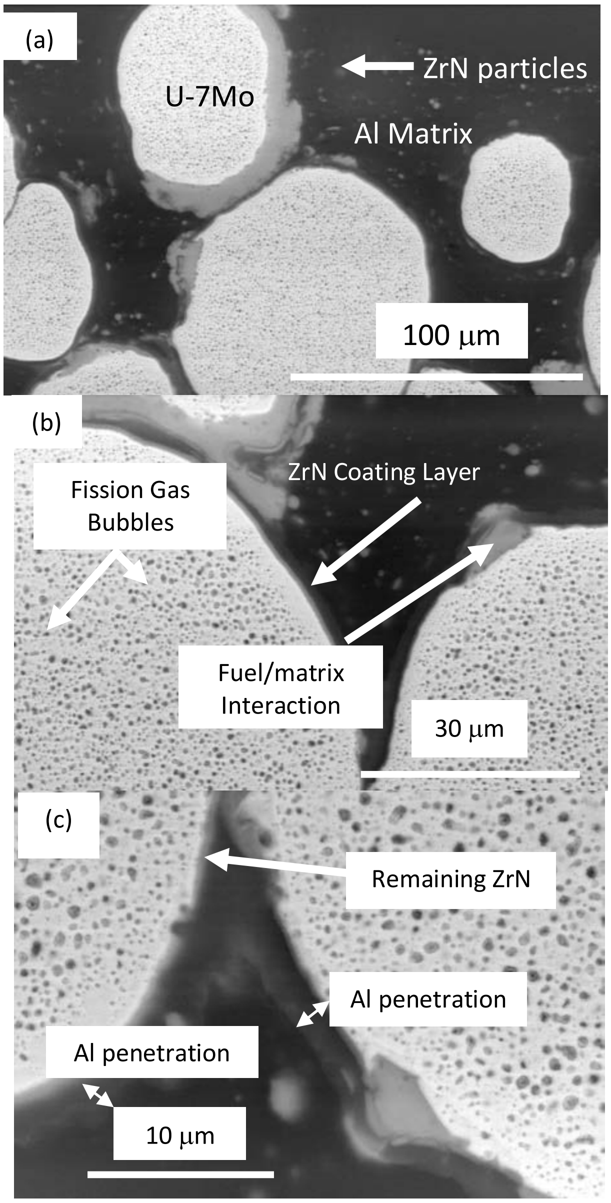


Figure 1. BSE images (a-c) of the polished surface of a cross-section from EMPI 0702. In (a), the microstructure exhibits very small amounts of interaction layer. In (b), is a higher magnification image showing fission gas bubbles, the coating, and interaction layer. In (c), regions can be seen where Al has penetrated into the ZrN coating.

3.1.2 EMPI 0712

Figure 2 shows BSE images of the EMPI 0712 microstructure, where some interaction layer could be found along with intact ZrN coating, and fission gas bubbles could be found in the U-7Mo fuel particles.

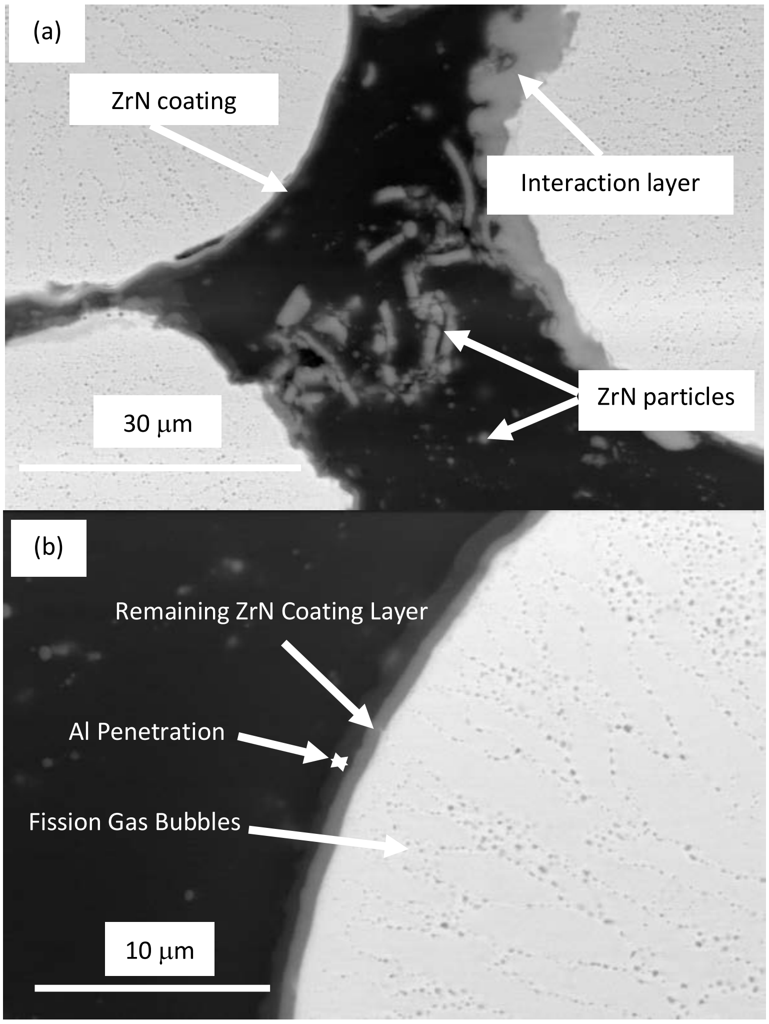
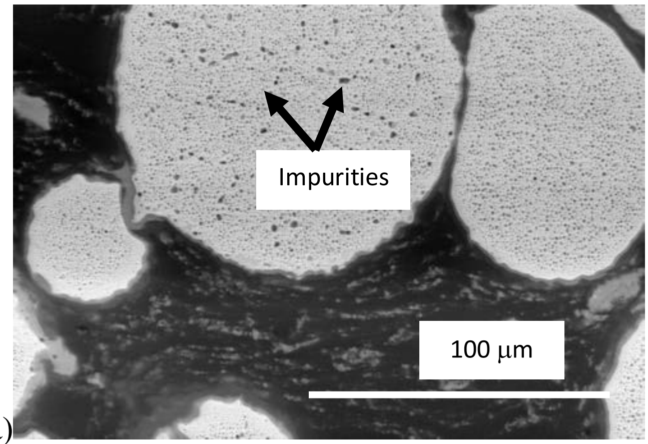


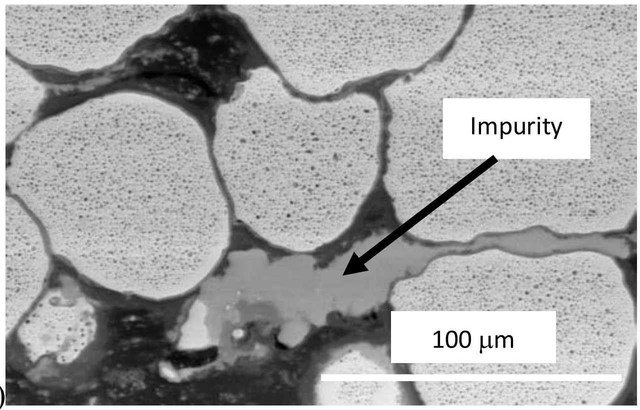
Figure 2. BSE images (a-c) of the polished surface of a cross-section from EMPI 0712. In (a), the microstructure exhibits in ZrN coating and very small amounts of interaction layer. In (b) is a higher magnification image showing fission gas bubbles, the ZrN coating, and penetration of Al into the ZrN coating.

3.1.3 EMPI 0905

Figure 3 shows the microstructure for EMPI 0905. Like was the case for the other two fuel plates, only limited fuel/matrix interaction was observed, along with intact ZrN coating. The U-7Mo fuel particles displayed uniformly distributed small fission gas bubbles. However, impurity phases were observed in many of the fuel particles, and relatively large impurity phases could be observed in the fuel meat.



(a)



(b)

Figure 3. BSE images (a,b) of the polished surface of a EMPI 0905. In (a), the U-7Mo fuel particles exhibited uniform, fine fission gas bubbles with limited fuel/matrix interaction layer formation. Some fuel particles contained impurity phases. In the image in (b) is an example of an impurity phase found in the fuel meat.

3.1.4 Comparison of Type 7A and Type 8A Microstructures

Different types of fuel plates were irradiated in the EMPIrE experiment, where the type 7A plates used as-atomized U-7Mo particles, and the Type 8A fuel plates employed atomized fuel particles that had been heat treated for 1 hour at 1,000˚C [3]. Earlier reported irradiation condition data that was projected for the EMPIRE fuel plates 0818, 0820, and 0821 [4] has been updated using the actual neutronics as-run reactor data for the EMPIRE irradiation experiment (see Table 2).

Table 2: Calculated irradiation parameters for EMPI 0818, 0820, and 0821.

Sample	SEM Cross-Section Fission Density (FD) Minimum (fissions/cm^3 UMo)	SEM Cross-Section Fission Density (FD) Average (fissions/cm^3 UMo)	SEM Cross-Section Fission Density (FD) Maximum (fissions/cm^3 UMo)	Fuel Plate BOL Average Surface Heat Flux (W/cm^2)	Fuel Plate BOL Peak Surface Heat Flux (W/cm^2)
EMPI 0818	3.3 x10^{21}	3.3 x10^{21}	3.5 x10^{21}	451.0	521.6
EMPI 0820	5.0 x10^{21}	5.1 x10^{21}	5.2 x10^{21}	310.6	383.7
EMPI 0821	6.2 x10^{21}	6.3 x10^{21}	6.4 x10^{21}	444.8	519.9

This irradiation condition information can be employed to compare the microstructures of selected Type 7A and Type 8A fuel plates that were exposed to similar irradiation conditions.

To determine if there is a difference in irradiation behavior under similar conditions for U-7Mo powders that are heat treated vis-à-vis as-atomized U-7Mo powders, fuel plates EMPI 0712 and EMPI 0818 can be compared. The EMPI 0712 fuel plate was irradiated at a peak heat flux of 520.9 W/cm^2, and the average FD of the SEM cross-section examined was 3.0 x 10^{21} fissions/cm^3. For EMPI 0818, it was 521.6 W/cm^2 and 3.3 x 10^{21} fissions/cm^3, respectively. Images for the fuel plates are presented in Figure 4. Both types of powders exhibit similar grain boundaries with precipitated fission gas bubbles.

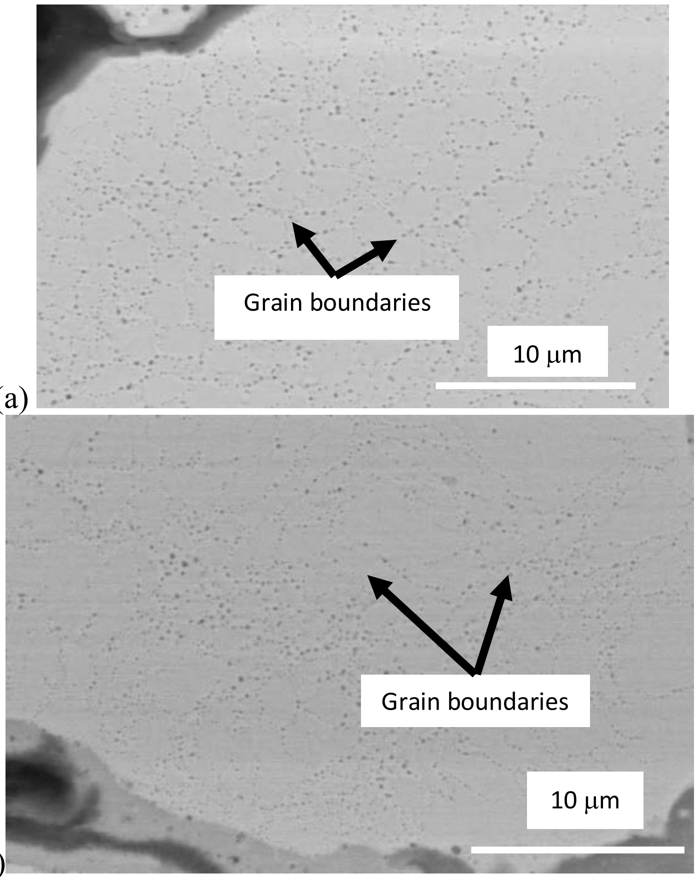
(a)
(b)


Figure 4. High magnification BSE images of high-power fuel plates (a) EMPI 0712 and (b) EMPI 0818. Multiple boundaries in the microstructure for both fuel plates contain fission gas bubbles.

The U-7Mo microstructures for fuel plates EMPI 0702 and 0821 are presented in Figure 5. Fuel plates EMPI 0702 and EMPI 0821 were irradiated at a peak heat flux of around 513 W/cm^2 and 520 W/cm^2, respectively. The average FD values for the SEM cross-sections examined for EMPI 0702 and 0821 were 5.7 x10^{21} fissions/cm^3 and 6.3 x10^{21} fissions/cm^3, respectively. Both types of powders exhibit similar microstructures comprised of uniform fission gas bubbles with diameters less than 5 microns.

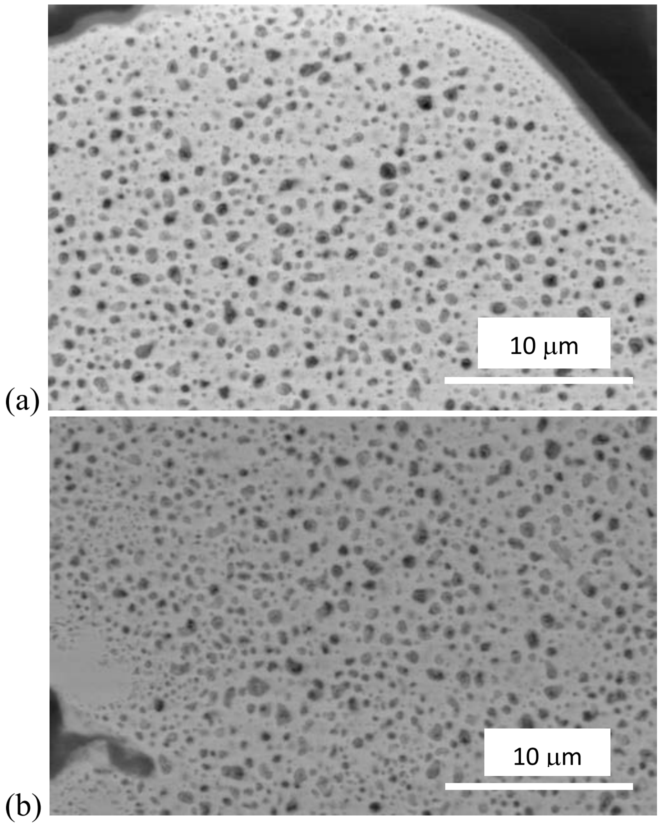
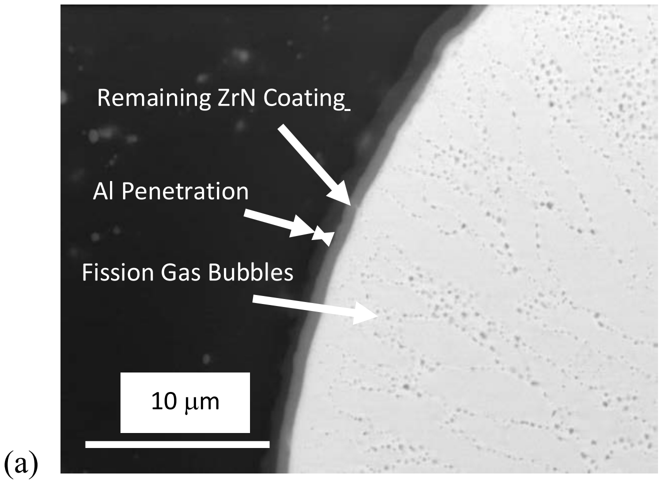
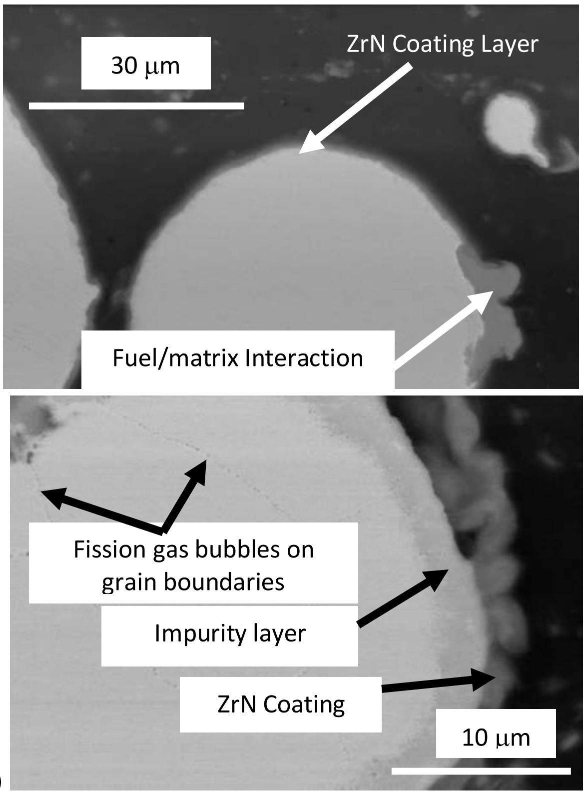


Figure 5. High magnification BSE images of fuel plates (a) EMPI 0702 and (b) EMPI 0821. Both microstructures show uniformly distributed fission gas bubbles a few microns in diameter.

Different batches of ZrN-coated U-7Mo powder were employed to fabricate Type 7A and Type 8A fuel plates. It is of interest to determine if noticeably different behaviors could be discerned for the different coating batches. Figure 6 shows example SEM images for fuel plate EMPI 0712 and EMPI 0818, which were irradiated at similar heat fluxes to similar burnups. Similar behaviors are observed for the ZrN coating in terms of how the amount of fuel/matrix interaction is reduced. Some penetration of Al into the ZrN is observed, and an impurity layer, which is formed on the U-7Mo surface during the heat treatment, can be observed between the U-7Mo and ZrN coating for the EMPI 0818 plate that uses heat-treated U-7Mo, but this layer doesn't seem to adversely affect the performance of the ZrN coating.



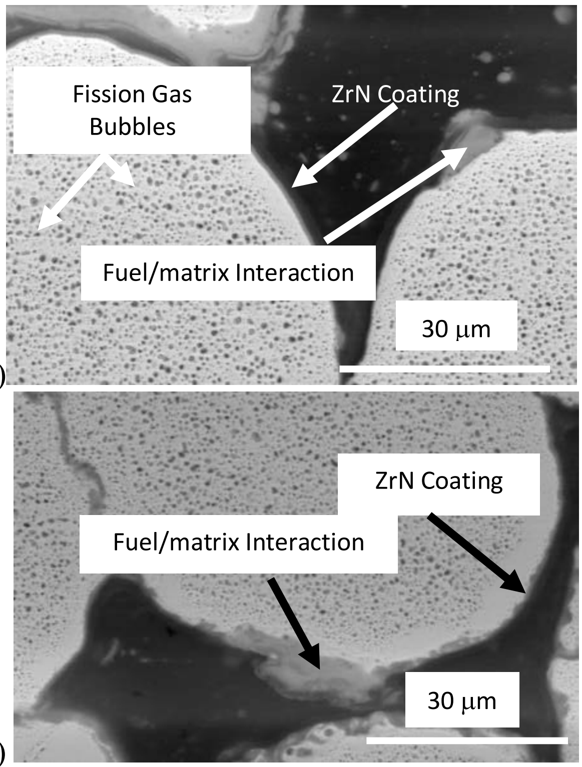


(b)

(c)

Figure 6. SEM images showing the ZrN coating (batch LA3 zero) for EMPI 0712 (a) and the ZrN coating (batch LA4 zero) for EMPI 0818 (b,c), which were both irradiated at high power to medium FD.

Figure 7 shows SEM images for fuel plates EMPI 0702 and EMPI 0821, which were irradiated to relatively high FD. Like was the case for EMPI 0712 and EMPI 0818, intact ZrN coating and some fuel/matrix interaction could be observed. Overall, the irradiation behavior of the different batches of ZrN coating was similar for the two fuel plates.



(a)

(b)

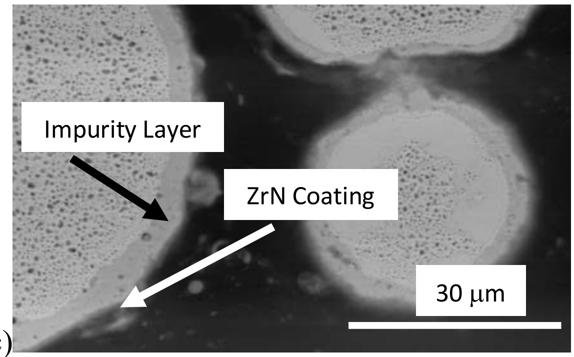
(c)


Figure 7. SEM images showing the ZrN coating for (a) EMPI 0702 and (b,c) EMPI 0821.

3. 2 Discussion

To identify the effects of irradiation on the microstructure of a nuclear fuel during a reactor experiment, it is important that the nuclear fuel's starting microstructure be well understood. Results of microstructural characterization of the U-7Mo powders and as-fabricated fuel plates employed in the EMPIRE experiment have been reported [5-7]. The as-atomized powders contained in the Type 7A fuel plates were made using a centrifugal atomization process [7]. For the Type 8A fuel plates, the centrifugally atomized powders were heat treated in a static furnace under vacuum at 1,000˚C for 1 hour and air quenched [7]. Characterization of the as-atomized powders showed that the grains in the material were a few microns in size. For the heat-treated powders the grains were tens of microns in size and a lamellar microstructure could be observed due to the fast cooling during the quenching process [5,6]. It was reported that the grains could be difficult to identify due to the presence of dark-contrast lamella in the material. Martensitic transformations can occur in U-Mo alloys when they are rapidly cooled, where the result can be the development of a banded microstructure that is comprised of long, parallel martensite plates containing fine arrays of transformation twins. Deformation can also induce a martensite reaction in some U-Mo alloys [8]. More characterization is needed to determine if the fine structure with fission gas bubbles on grain boundaries observed in the fuel plate with heat treated powders that was irradiated to an average FD of 3.3 $x10^{21}$ fissions/cm^3 (EMPI 0818) can be linked to the starting as-fabricated banded microstructure, or if it was just the microstructure that develops for heat treated powders during irradiation at certain reactor conditions. Since after irradiation to an average FD of 3.3 x 10^{21} fissions/cm^3 the atomized and heat-treated powders exhibited similar microstructures, it is not surprising that the atomized and heat treat powders also exhibited similar microstructures after an average FD of around 6 x 10^{21} fissions/cm^3.

The different batches of ZrN coating appeared to behave in a similar manner at different irradiation conditions for both as-atomized and heat-treated U-7Mo powders. For the powders heat treated at 1,000˚C before applying the PVD ZrN coating, more oxidation layers were observed adjacent to the U-7Mo after fabrication. The presence of such a layer did not appear to adversely affect the bonding of the ZrN coating. In the future, image analysis will be employed to quantify the changes in microstructure that occur during irradiation of each type of fuel plate.

4. Conclusions

Based on the microstructural characterization of Type 7A EMPIrE fuel plates with PVD-deposited ZrN coating on the U-7Mo fuel particles that were irradiated up to an average FD of 6.1 x 10^{21} fissions/cm^3, stable swelling behavior was observed in the as-atomized fuel particles employed in the fuel plates, and only limited fuel/matrix interaction occurred during irradiation. The original ZrN coating was still present around the fuel particles, and some Al diffusion from the matrix into the ZrN was observed. Comparison of the Type 7A SEM images with those available for Type 8A fuel plates, which utilized powders annealed at 1,000˚C for 1 hour, showed that Type 7A and Type 8A fuel particles exhibited similar microstructural evolution during irradiation. Additionally, the different ZrN batches used for the different types of fuel plates seemed to behave in a similar manner during irradiation.

Acknowledgments
This work is supported by the U.S. Department of Energy, under DOE Idaho Operations Office Contract DE-AC07-05ID14517. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

References
[1]. J. Snelgrove et al., Nucl. Engr. Design 178 (1997) 119-126.
[2]. M. K. Meyer, J. Gan, J. F. Jue, D. D. Keiser, E. Perez, A. Robinson, D. M. Wachs, N. Woolstenhulme, G. L. Hofman, and Y. S. Kim, "Irradiation Performance of U-Mo Monolithic Fuel," Nucl. Energy and Tech. 46 (2) (2014) 169-182.
[3]. William A. Hanson, Adam B. Robinson, Nancy J. Lybeck, Joseph W. Nielsen, Bei Ye, Zhi-Gang Mei, Dennis D. Keiser Jr, Laura M. Jamison, Gerard L. Hofman, Abdellatif M. Yacout, Ann Leenaers, Bertrand Stepnik, Irina Y. Glagolenko," Non-destructive analysis of swelling in the EMPiRE fuel test," J. Nucl. Mater. 564 (2022), published on-line. https://doi.org/10.1016/j.jnucmat.2022.153683
[4] Dennis Keiser, Jr. et al., "Recent Results of Scanning Electron Microscopy Characterization of Fuel Plates Irradiated in the EMPIrE Irradiation Experiment," Proc. Of the Research Reactor Fuel Management Conference. Budapest, Hungary. June 6-10, 2022.
[5] X. Iltis, H. Palancher, F. Vanni, J. Allenou, B. Stepnik, et al.. Characterization of fresh EMPIrE and SEMPER FIDELIS plates made with PVD-coated U(Mo) particles. RRFM 2018, Mar 2018, Munich, Germany.
[6] Xavière Iltis, Hervé Palancher, Jérôme Allenou, Florence Vanni, Bertrand Stepnik, Ann Leenaers, Sven Van Den Berghe, Dennis D. Keiser, Irina Glagolenko, Characterization of fresh EMPIrE and SEMPER FIDELIS U(Mo)/Al fuel plates made with PVD-coated U(Mo) particles, EPJ Nuclear Sci. Technol. 4, 49 (2018)
[7] X. Iltis, I. Zacharie-Aubrun, H.J. Ryu, J.M. Park, A. Leenaers, A.M. Yacout, D.D. Keiser, F. Vanni, B. Stepnik, T. Blay, N. Tarisien, C. Tanguy, H. Palancher, Microstructure of as atomized and annealed U-Mo7 particles: A SEM/EBSD study of grain growth, J. Nucl. Mater. 495 (2017) 249-266.
[8] J. G. Speer and D. V. Edmonds, "An Investigation of the γ-α Martensitic Transformation in Uranium Alloys," Acta Metallurgica 36(4) (1988) 1013-1033.