INL/JOU-17-41347-Revision-0

Observed Changes in As-Fabricated U-10Mo Monolithic Fuel Microstructures After Irradiation in the Advanced Test Reactor

Dennis Keiser, Jr., Jan-Fong Jue, Brandon Miller, Jian Gan, Adam Robinson, James Madden

August 2017

INL/JOU-17-41347-Revision-0

Observed Changes in As-Fabricated U-10Mo Monolithic Fuel Microstructures After Irradiation in the Advanced Test Reactor

Dennis Keiser, Jr., Jan-Fong Jue, Brandon Miller, Jian Gan, Adam Robinson, James Madden

August 2017

Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

Prepared for the
U.S. Department of Energy

Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

Observed Changes in As-Fabricated U-10Mo Monolithic Fuel Microstructures After Irradiation in the Advanced Test Reactor

DENNIS KEISER JR. ,[1,2] JAN-FONG JUE,[1] BRANDON MILLER,[1] JIAN GAN,[1] ADAM ROBINSON,[1] and JAMES MADDEN[1]

1.—Idaho National Laboratory, P. O. Box 1625, Idaho Falls, ID 83403-6188, USA. 2.—e-mail: dennis.keiser@inl.gov

A low-enriched uranium U-10Mo monolithic nuclear fuel is being developed by the Material Management and Minimization Program, earlier known as the Reduced Enrichment for Research and Test Reactors Program, for utilization in research and test reactors around the world that currently use high-en-riched uranium fuels. As part of this program, reactor experiments are being performed in the Advanced Test Reactor. It must be demonstrated that this fuel type exhibits mechanical integrity, geometric stability, and pre-dictable behavior to high powers and high fission densities in order for it to be a viable fuel for qualification. This paper provides an overview of the microstructures observed at different regions of interest in fuel plates before and after irradiation for fuel samples that have been tested. These fuel plates were fabricated using laboratory-scale fabrication methods. Observations regarding how microstructural changes during irradiation may impact fuel performance are discussed.

INTRODUCTION

To support nuclear nonproliferation efforts, the Material Management and Minimization (M3) pro-gram (formerly known as the Reduced Enrichment for Research Reactor program) is developing low-enriched uranium (LEU) fuel to reduce the demand for highly enriched uranium (HEU) fuels currently used in research and test reactors throughout the world.[1] A U-10Mo monolithic fuel is one type of fuel that is being developed.[2] This plate-type fuel is composed of a U-10Mo fuel foil with a Zr diffusion barrier that is clad with AA6061. The U-10Mo foil is bonded to the Zr diffusion barrier using a hot co-rolling process,[3] and the cladding is subsequently bonded to the fuel foil by hot isostatic pressing (HIP).[4] The M3 program is in the process of conducting the necessary irradiating testing so that this fuel can be qualified for use. To support this qualification, it must be demonstrated that this fuel type exhibits mechanical integrity, geometric sta-bility, and stable and predictable irradiation behav-ior to high powers and high fission densities.

The monolithic fuel plate microstructure that will be present in a fuel plate going into a reactor is intimately related to the fabrication processes employed to make it.[5,6] The characteristics of this microstructure must be well understood, since the ultimate fuel plate performance during irradiation will depend on the performance of the phases that compose this microstructure. Microstructural fea-tures are correlated to areas of interest within an as-fabricated fuel plate; the U-10Mo fuel foil, the Zr diffusion barrier, and the Al-6061 cladding are of interest, along with features at the U-10Mo/Zr, Zr/Al-6061 cladding, U-10Mo/ Al-6061 cladding, and Al-6061 cladding/Al-6061 cladding interfaces. When describing the general microstructure of the U-10Mo fuel phase, it is necessary to discuss the phases present, the grain sizes observed, and com-positions observed locally. In the Zr diffusion bar-rier, the primary characteristics of interest are the grain size and orientation, and the presence of impurity phases. For the Al-6061 cladding, the phases present and the grain size observed are of interest. At interfaces, it is common to observe reaction layers that develop due to solid-state interdiffusion during fabrication. As a result, at the U-10Mo/Zr interface, interdiffusion of U, Mo, and Zr during fabrication can result in development of phases containing U, Mo, and/or Zr.[7] At the Zr/Al-6061 interface, phases can develop that contain Zr,

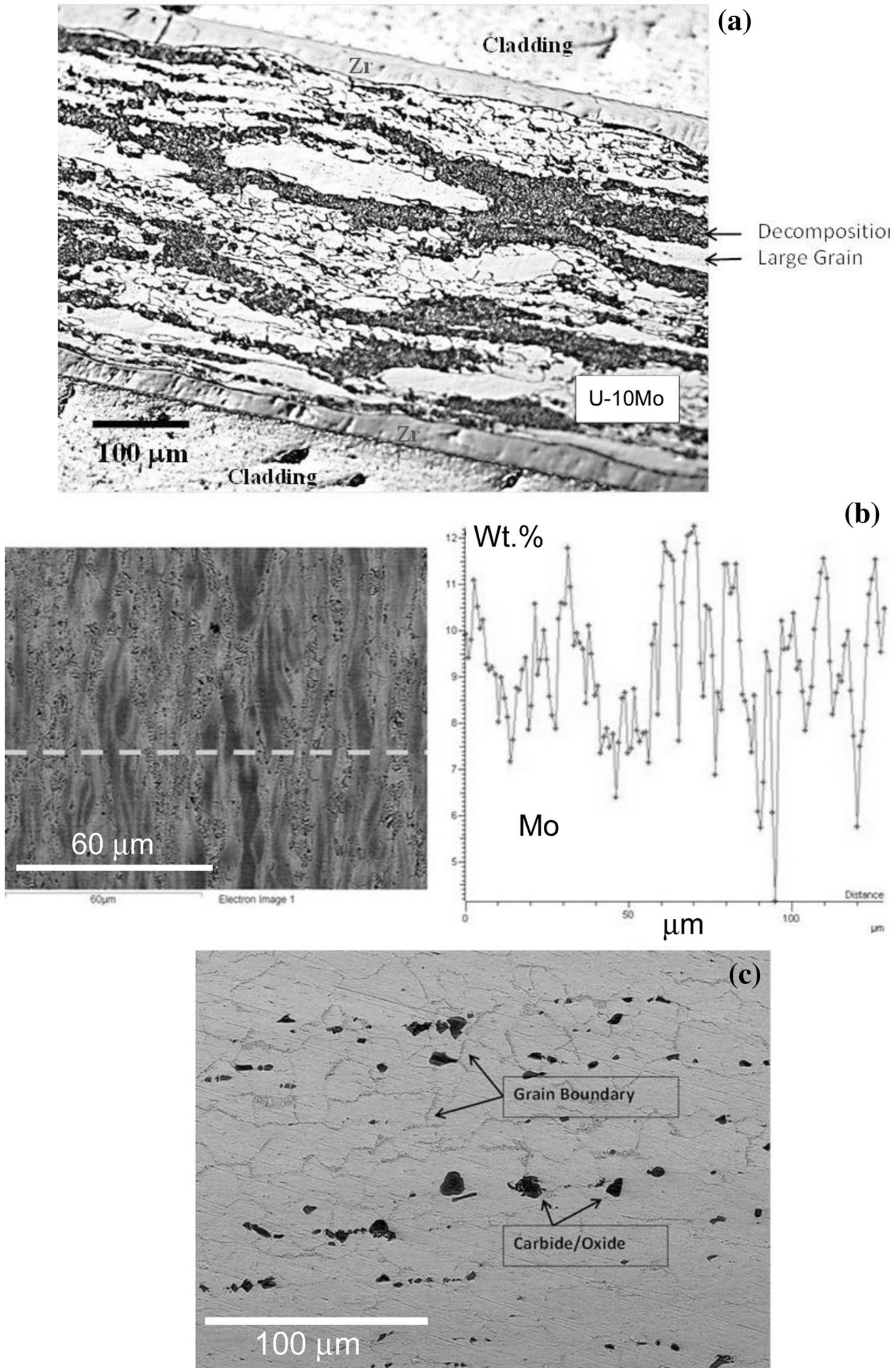

Fig. 1. (a) OM image showing an as-fabricated fuel plate microstructure composed of elongated U-10Mo grains and lamellar regions where single phase γ-(U,Mo) has decomposed, most likely into α-U and γ-(U,Mo). Some regions can transform all the way to α-U and γ'-(U,Mo). (b) BSE image of the U-10Mo fuel in an as-fabricated fuel plate, along with the results of a Mo linescan (dashed). (c) OM micrograph showing where impurity phases are present in the U-10Mo fuel for an as-fabricated fuel plate.

Al, Mg, and/or Si.[8] At the U-10Mo/Al-6061 interface, phases can be present that contain U, Mo, Al, Mg, and/or Si.[9] Zr can also be present due to the presence of the diffusion barrier layer.[5]

This paper describes the microstructural changes at some regions of interest during irradiation within samples characterized to date, produced using lab-oratory-scale fabrication methods that were not

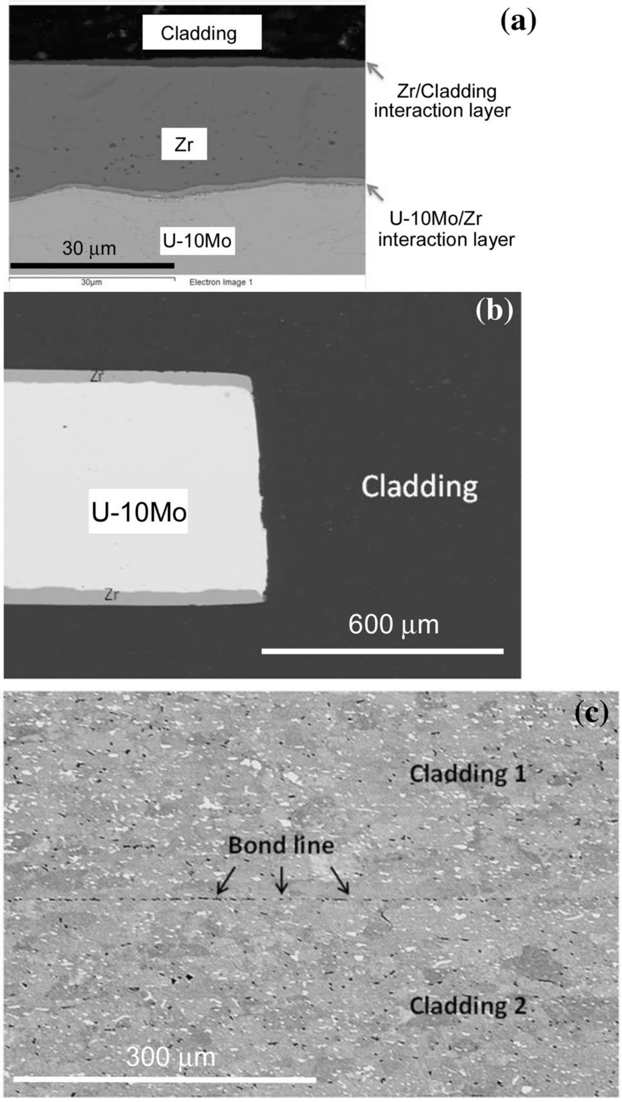

Fig. 2. BSE images showing (a) the solid–state interaction layers that form at the U-10MoZr and Zr/AA6061 cladding interfaces, (b) the microstructure at the U-10Mo/AA6061 interface, which is located at the edge of the fuel zone, and (c) the microstructure at the interface between the AA6061 cladding pieces that have been HIP-bonded to encase the U-10Mo fuel.

optimized. Microstructural characterization data have been obatined from mechanically polished samples, along with samples prepared using focused ion beam (FIB) methods. These samples were analyzed using optical metallography (OM), scanning electron microscopy with energy-dispersive-spectroscopy and wavelength-dispersive spectroscopy (SEM/EDS/WDS), and transmission electron microscopy (TEM). Both backscattered electron (BSE) and secondary electron (SE) images were obtained using SEM. This paper discusses how observed microstructural changes during irradiation may impact fuel performance characteristics such as swelling, heat transfer, and the overall stability of the fuel system.

AS-FABRICATED MICROSTRUCTURES

U-10Mo Alloy

Depending on how a fuel plate is fabricated, it is possible for the U-10Mo alloy fuel microstructure to consist of elongated fuel grains in the rolling direction, areas of transformed γ-(U,Mo) phase, areas of chemical banding, and impurity phases in different areas of the microstructure.[3,10–12] Figure 1 shows examples of these microstructural features. The elongated grains results from the rolling process. The decomposed areas are a result of the metastable γ-(U,Mo) phase transforming to α-U and γ-(U,Mo), and possibly γ'-(U,Mo) if the holding time is long enough, during fabrication at higher temperatures. Repas et al.[13] reported a time–temperature–transformation diagram showing that, if U-10 wt.% Mo is kept at relatively high temperatures (around 500°C) for long enough, transformations to α-U and γ'-(U,Mo) will occur. Yet, even if this transformation does occur during fabrication, literature suggests that these regions will transform back to bcc γ-(U,Mo) phase during irradiation,[14] which is the preferred phase, primarily due to the fact that bcc γ-(U,Mo) phase has isotropic properties and exhibits good irradiation performance.[15] The chemical banding results from rolling a starting as-cast U-10Mo alloy coupon (that exhibited coring during casting) into a foil. The impurity phases are typically uranium carbides that are present because varying amounts of carbon are present in the original starting material.

Interfaces

At the three interfaces in the fuel plates (i.e., U-10Mo/AA6061, U-10Mo/Zr, and Zr/AA6061), interdiffusion zones can be present, giving evidence of a metallurgical bond during the fuel fabrication processes. Figure 2 shows examples of interdiffusion zones observed at these interfaces. A final interface is present between the AA6061/AA6061 cladding, which is employed to encase the fuel (Fig. 2). The presence of impurity phases at the interfaces can impact on the bonding quality; Supplementary Fig. S1 shows a uranium carbide phase present at the U-10Mo/Zr interface.

IRRADIATION TESTING AND CHARACTERIZATION OF IRRADIATED SAMPLES

Irradiation testing was performed in the Advanced Test Reactor (ATR) using small fuel plates and large fuel plates. Testing smaller fuel plates provides an efficient way to test different fabrication methods and to generate irradiation performance data for a variety of conditions. Once the needed data are generated and a final fuel type and fabrication method identified, larger-scale testing can commence. Since smaller fuel plates can be exposed to different neutron flux gradients

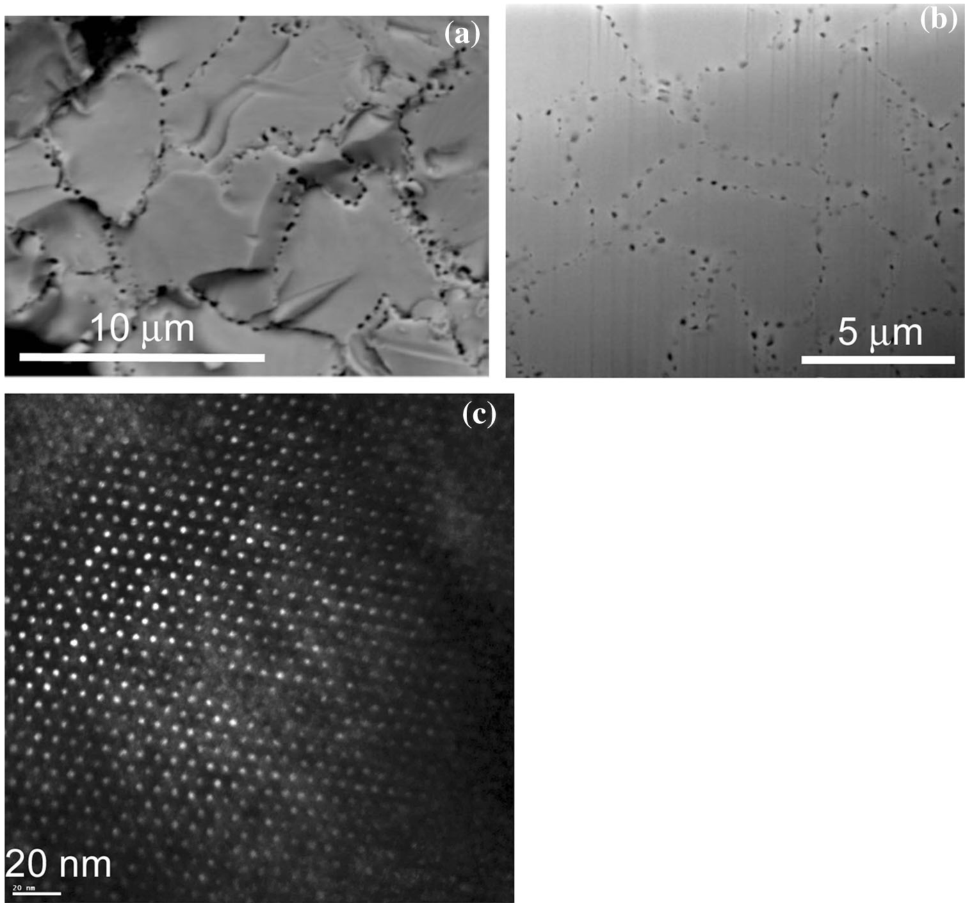

Fig. 3. BSE images taken from a low-burnup fuel (sample L1F040 achieving fission density of $\sim 3.2 \times 10^{21}$ fissions/cm^3), showing (a) a fracture surface present in irradiated fuel that revealed the fission gas bubbles present on the fuel grain boundaries and (b) a FIB sample showing the microstructure for the same fuel plate depicted in (a) where fission gas bubbles are on grain boundaries. (c) Dark-field TEM micrograph of fission gas superlattice observed in U-Mo monolithic fuel irradiated to low burnup (sample MZ50 achieving fission density of $\sim 2 \times 10^{21}$ fissions/cm^3).

and mechanical stresses, care should be taken when comparing results with those from irradiation experiments using larger fuel plates. The typical dimensions a smaller fuel plate are 2.5 cm by 10.2 cm by 0.14 cm, and the fuel zone dimensions are 8.3 cm by 1.9 cm by 0.03 cm. For the larger fuel plates, the fuel plate dimensions are 5.7 cm by 114.3 cm by 0.13 cm and the fuel zone dimensions are 3.5 cm by 57.2 cm by 0.04 cm.[2] To study the microstructural evolution in fuel plates of different sizes, microstructural characterization was performed on relatively small samples taken from the different irradiated fuel plates. Sample cross-sections were produced in the Hot Fuel Examination Facility (HFEF) at the Materials and Fuels Complex at the Idaho National Laboratory for OM analysis. Other smaller samples were produced from the fuel plates in HFEF then sent to a separate facility called the Electron Microscopy Laboratory (EML) for analysis using SEM/EDS/WDS and TEM.

AS-IRRADIATED MICROSTRUCTURES

U-10Mo Alloy

With respect to the elongated fuel grains in the rolling direction, areas of transformed γ-(U,Mo) phase, areas of chemical banding, and impurity phases in different areas of the microstructure that can be found in the starting U-10Mo microstructure,[5] irradiation affects some of these features more than others. Figure 3 shows the microstructure of the U-Mo fuel zone for a fuel plate after irradiation to relatively low burnup (or fission density), where observable fission gas bubbles are present on the grain boundaries, comparing the microstructure for a fracture surface with that for a sample produced by FIB. TEM analysis demonstrated that smaller fission gas bubbles around 2 nm in size were present inside the grains of monolithic fuel U-Mo alloy irradiated to relatively low burnup. These bubbles were organized on a fcc superlattice in the bcc γ-(U,Mo) fuel. This superlat-

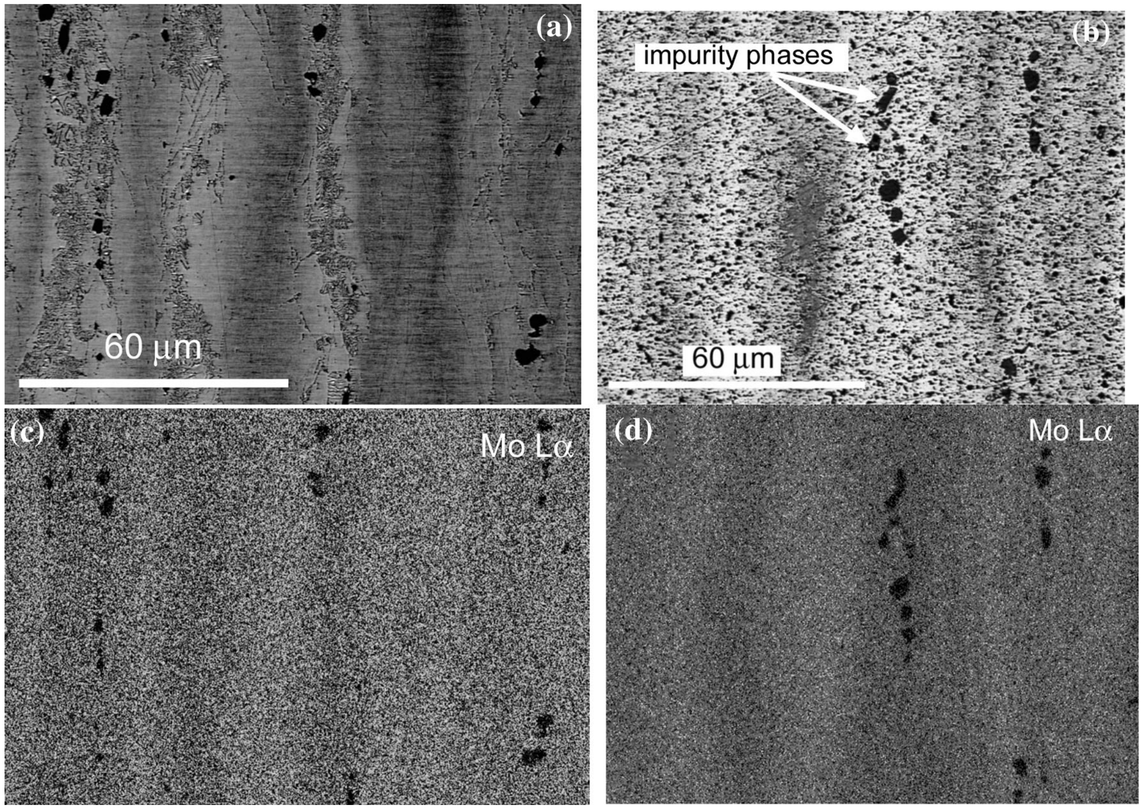

Fig. 4. BSE images showing the microstructure of (a) an as-fabricated fuel plate and (b) a fuel plate after irradiation to relatively high burnup ($\sim 5 \times 10^{21}$ fissions/cm^3). The small black spots in (b) are fission gas bubbles. The x-ray maps in (c) and (d) show the Mo distribution in the archive and irradiated fuel plates, respectively.

tice has been routinely observed in U-Mo dispersion fuels,[16,17] and its presence is an important reason why U-10Mo monolithic fuel exhibits relatively stable swelling behavior during irradiation, since very small bubbles can accommodate a large amount of fission gas without causing significant swelling of the fuel.

At higher burnup, relatively large fission gas bubbles were observed in the U-10Mo fuel (Fig. 4). At this stage of burnup, the fuel has gone through fission-induced recrystallization (grain refinement), which has also been observed for U-Mo dispersion fuel,[17,18] and this process changes the microstructure such that relatively large fission gas bubbles develop through the microstructure. With respect to the chemical banding that can be found in as-fabricated fuel plates, the heterogeneous distribution of Mo remains in the U-10Mo fuel, even after irradiation to high burnup. This is demonstrated by the Mo x-ray maps shown in Fig. 4. A uniform distribution of fission gas bubbles composes the microstructure of the irradiated fuel, and larger phases that appear to be uranium carbide impurity phases can also be observed (Fig. 4).

Interfaces

As mentioned above, interdiffusion zones develop at the U-10Mo/Zr and Zr/AA6061 interfaces during fuel fabrication, and the irradiation behavior of the phases in these zones is of interest. Since fission fragments can recoil from the surface of uranium-bearing fuel, there is a recoil range of fission fragments that extends less than 10 microns into the Zr diffusion barrier, which is nominally 25 microns thick.[19] This means that, at the U-10Mo/Zr interface, it must be determined that this fission fragment recoil does not overly degrade the bonding at the interface and that the phases that are present remain stable. At the Zr/AA6061 interface, which is typically outside the range of the recoil zone, good integrity of the potentially brittle phases present in this region of the fuel plate must also be demonstrated at different levels of burnup. Figure 5 shows BSE images of the observed microstructures for polished samples at these interfaces after fuel fabrication and then after irradiation. Cracks or signficant porosity was not observed at either of the two interfaces, suggesting that good bonding was maintained. Due to the particular interest in the

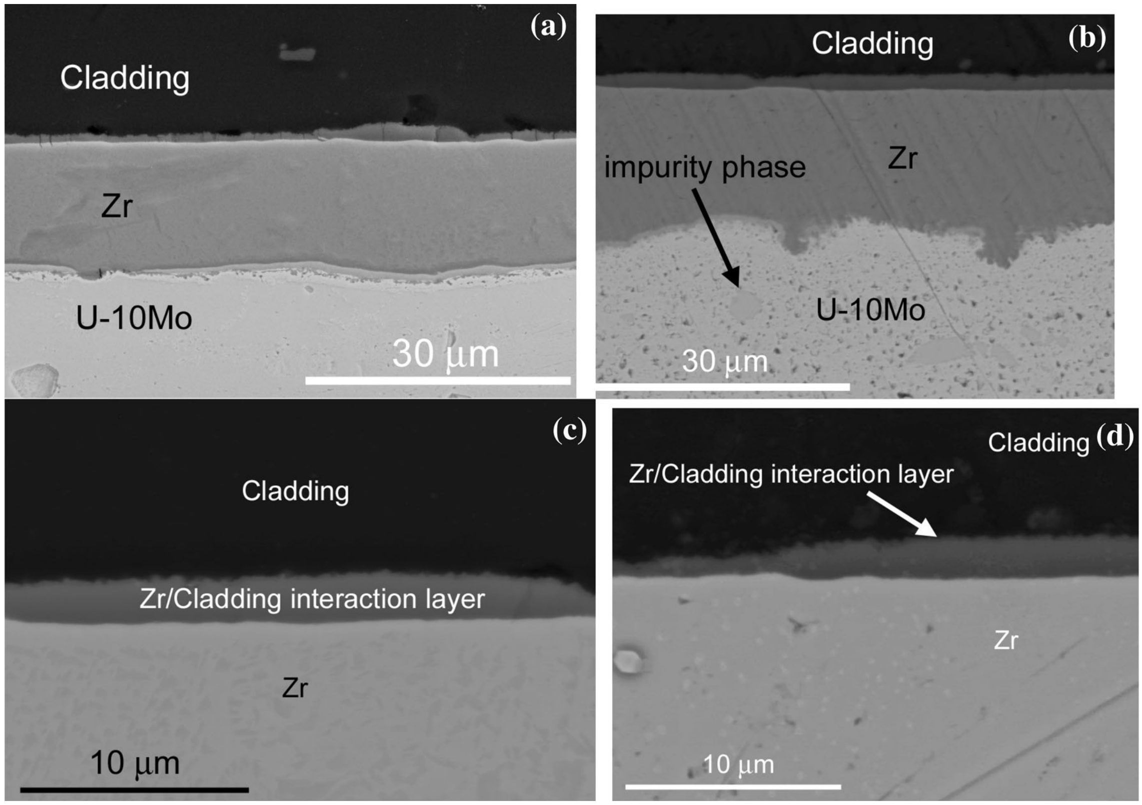

Fig. 5. BSE images of (a) the U-10Mo/Zr interface for an archive fuel plate before irradiation and (b) the same interface after irradiation (~5 × 10^{21} fissions/cm^3). (c) BSE image at Zr/AA6061 interface for an archive fuel plate before irradiation and (d) the same interface after irradiation. What appear to be uranium carbide phases can be found in (b).

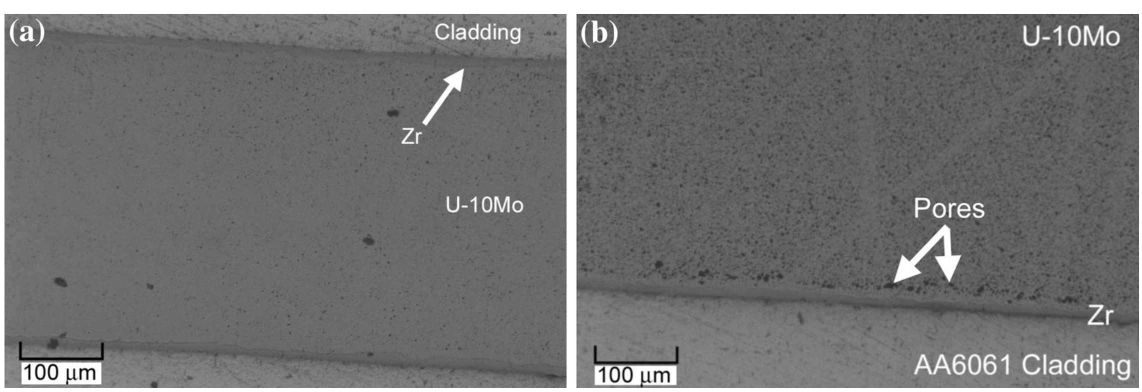

Fig. 6. OM micrographs of observed microstructures for U-10Mo monolithic fuel plates irradiated to (a) intermediate burnup (~2–3 × 10^{21} fissions/cm^3) and (b) high burnup (~5 × 10^{21} fissions/cm^3). In (b), relatively large pores are present near the U-10Mo/Zr interface.

microstructural development at the U-10Mo/Zr interface, FIB samples were taken from different irradiated fuel plates. Supplementary Fig. S2 shows BSE images for FIB samples taken from two different irradiated fuel plates at this interface. Much of the Zr does not exhibit porosity, and there is no evidence of cracks along the interface. Fission gas bubbles can be observed in the U-10Mo fuel all the way to the U-10Mo/Zr interface. In the OM images in Fig. 6, relatively large porosity is observed near the U-10Mo/Zr interface in some fuel samples irradiated to relatively high burnup. This appears to occur at a location in the as-fabricated fuel plate U-10Mo/Zr interdiffuson zone near the U-10Mo/Zr interface where lower Mo contents have been measured. If the Mo content is too low in the fuel after fuel fabrication, α-U could be present in this region of the fuel, and this phase has the potential to exhibit poor irradiation performance.[20] The cause of the development of larger pores in this fuel region at high burnup is currently under investigation.

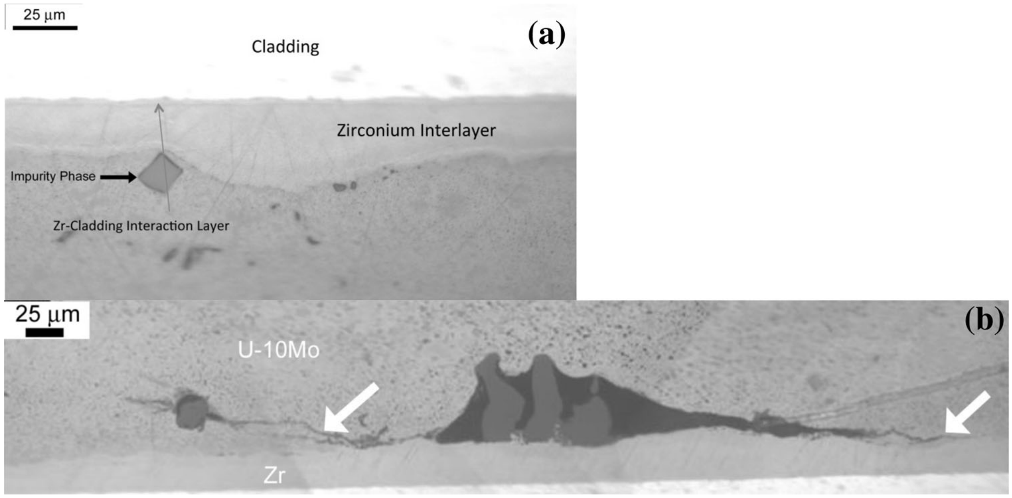

Fig. 7. OM images showing (a, b) impurity phases at the U-10Mo/Zr interface for samples taken from two different locations of an irradiated U-10Mo monolithic fuel plate (AFIP-6 MKII fuel plate irradiated to fission density of $\sim 5 \times 10^{21}$ fissions/cm^3). The arrows in (b) show cracks, and it is not clear if the cracks developed during sample preparation.

Since impurity phases can be present at the U-10Mo/Zr interface after fuel fabrication, it is important to determine how these phases may affect bonding. Optical micrographs showing where these phase have been observed in two different regions of an irradiated fuel plate are presented in Fig. 7. For the samples characterized to date, good bonding seems to be retained in areas with impurity phases.

DISCUSSION AND CONCLUSION

To date, both small- and large-scale U-10Mo monolithic fuel plates have been successfully irradiated in the ATR. Samples from these fuel plates have been characterized using a variety of techniques to improve understanding of how the fuel plate microstructure evolves under different irradiation conditions and in fuel plates of different sizes that were fabricated using different methods. Data have been generated with respect to the microstructural evolution of the U-10Mo and at the different interfaces present in fuel plates irradiated to different levels of burnup. Characterization results suggest that U-10Mo monolithic fuel exhibits good mechanical integrity, dimensional stability, and irradiation performance. This is based on information generated for samples that were fabricated using nonoptimized fuel fabrication methods. Future work will focus on characterizing irradiated fuel plates where the methods employed to fabricate the plates have been optimized.

ACKNOWLEDGEMENT

This submitted manuscript was authored by a contractor of the U.S. Government under DOE Contract No. DE-AC07-05ID14517. Accordingly, the U.S. Government retains and the publisher, by accepting the article for publication, acknowledges that the U.S. Government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this manuscript, or allow others to do so, for U.S. Government purposes. Funding was provided by National Nuclear Security Administration.

REFERENCES

1. D.D. Keiser Jr., S.L. Hayes, M.K. Meyer, and C.R. Clark, *JOM* 55, 55 (2003).
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, *Nucl. Energy Technol.* 46, 169 (2014).
3. G.A. Moore and M.C. Marshall, *Co-Rolled U10Mo/Zirconium–Barrier-Layer Monolithic Fuel Foil Fabrication Process*. Idaho National Laboratory Report, INL/EXT-10-17774 (2010).
4. J.-F. Jue, B.H. Park, C.R. Clark, G.A. Moore, and D.D. Keiser Jr., *Nucl. Technol.* 172, 204 (2010).
5. J.-F. Jue, D.D. Keiser Jr., C.R. Breckenridge, G.A. Moore, and M.K. Meyer, *J. Nucl. Mater.* 448, 250 (2014).
6. J.-F. Jue, T.L. Trowbridge, C.R. Breckenridge, G.A. Moore, M.K. Meyer, and D.D. Keiser Jr., *J. Nucl. Mater.* 460, 153 (2015).
7. E. Perez, B. Yao, D.D. Keiser Jr., and Y.H. Sohn, *J. Nucl. Mater.* 402, 8 (2010).
8. Y. Park, J. Yoo, K. Huang, D.D. Keiser Jr., J.F. Jue, B. Rabin, G. Moore, and Y.H. Sohn, *J. Nucl. Mater.* 447, 215 (2014).
9. E. Perez, D.D. Keiser, and Y.H. Sohn, *J. Nucl. Mater.* 477, 178 (2016).
10. A.J. Clarke*, K.D. Clarke, R.J. McCabe, C.T. Necker, P.A. Papin, R.D. Field, A.M. Kelly, T.J. Tucker, R.T. Forsyth, P.O. Dickerson, J.C. Foley, H. Swenson, R.M. Aikin Jr., and D.E. Dombrowski, *J. Nucl. Mater.* 465, 784 (2015).
11. Y. Park, N. Eriksson, R. Newell, D.D. Keiser, and Y.H. Sohn, *J. Nucl. Mater.* 480, 271 (2016).
12. Y. Park, N. Eriksson, D.D. Keiser Jr., J.F. Jue, B. Rabin, G. Moore, and Y.H. Sohn, *Mater. Charact.* 103, 50 (2015).
13. P.E. Repas, R.H. Goodenow, and R.F. Hehemann, *Trans. Am. Soc. Metals* 13, 150 (1964).
14. M.L. Bleiberg, L.J. Jones, and B. Lustman, *J. Appl. Phys.* 27, 1270 (1956).
15. G. Beghi, *Gamma Phase Uranium–molybdenum Fuel Alloys*. European Atomic Energy Community-Euratom, EUR 4053e (1968).
16. J. Gan, D.D. Keiser Jr., D.M. Wachs, A.B. Robinson, B.D. Miller, and T.R. Allen, *J. Nucl. Mater.* 396, 234 (2010).
17. J. Gan, D.D. Keiser Jr., B.D. Miller, A.B. Robinson, J.F. Jue, P. Medvedev, and D.M. Wach, *J. Nucl. Mater.* 424, 43 (2012).
18. A. Leenaers, W. Van Renterghem, and S. Van den Berghe, *J. Nucl. Mater.* 476, 218 (2016).
19. E.R. Smith and P.W. Frank, *Recoil Range of Fission Fragments in Zirconium*. AEC Research and Development Report. WAPD-TM-198 (1959).
20. E.S. Fisher, *J. Alloys Compd.* 213/214, 254 (1994).