PNNL-25365

Pacific Northwest
NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

U-10Mo/Zr Interface Modeling using a Microstructure-Based FEM Approach

April 2016

A Soulami
Z Xu
V Joshi
D Burkes
C Lavender
E McGarrah

U.S. DEPARTMENT OF ENERGY

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

DISCLAIMER

PACIFIC NORTHWEST NATIONAL LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC05-76RL01830

Printed in the United States of America

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information,
P.O. Box 62, Oak Ridge, TN 37831-0062;
ph: (865) 576-8401
fax: (865) 576-5728
email: reports@adonis.osti.gov

Available to the public from the National Technical Information Service
5301 Shawnee Rd., Alexandria, VA 22312
ph: (800) 553-NTIS (6847)
email: orders@ntis.gov <http://www.ntis.gov/about/form.aspx>
Online ordering: http://www.ntis.gov

This document was printed on recycled paper.
(8/2010)

PNNL-25365

U-10Mo/Zr Interface Modeling using a Microstructure-Based FEM Approach

A Soulami
Z Xu
V Joshi
D Burkes
C Lavender
E McGarrah

April 2016

Prepared for
the U.S. Department of Energy
under Contract DE-AC05-76RL01830

Pacific Northwest National Laboratory
Richland, Washington 99352

Executive Summary

Pacific Northwest National Laboratory has been investigating manufacturing processes for a plate-type fuel made from a uranium alloy with 10 weight percent molybdenum (U-10Mo). This work supports the Reactor Conversion Program of the U.S. Department of Energy National Nuclear Security Administration's Office of Material Management and Minimization. Use of metallic fuel is required to meet the objectives of the Reactor Conversion Program for the fleet of high-performance research reactors in the United States.

The U-10Mo in low enrichments (LEU) has been identified as the most promising alternative to the current highly enriched uranium (HEU) used in the United States' fleet of high performance research reactors (USHPRRs). The nominal configuration of the new LEU U-10Mo plate fuel comprises a U-10Mo fuel foil enriched to slightly less than 20% U-235 (0.08" to 0.02" thick), a thin Zr interlayer/diffusion barrier (25 μm thick) and a relatively thick outer can of 6061 aluminum. Currently the Zr interlayer is clad by hot roll bonding. Previous studies and observations revealed a thinning of the zirconium (Zr) layer during this fuel fabrication process, which is not desirable from the fuel performance perspective. Coarse UMo grains, dendritic structures, Mo concentration segregation, carbides, and porosity are present in the as-cast material and can lead to a nonuniform UMo/Zr interface. The purpose of the current work is to investigate the effects of these microstructural parameters on the Zr coating variation.

A microstructure-based finite-element method model was used in this work, and a study on the effect of homogenization on the interface between U-10Mo and Zr was conducted. The model uses actual backscattered electron–scanning electron microscopy microstructures, Mo concentrations, and mechanical properties to predict the behavior of a representative volume element under compressive loading during the rolling process. The model successfully predicted the experimentally observed thinning of the Zr layer in the as-cast material. The model also uses results from a homogenization model as an input, and a study on the effect of different levels of homogenization on the interface indicated that homogenization helps decrease this thinning. This model can be considered a predictive tool representing a first step for model integration and an input into a larger fuel fabrication performance model.

Acknowledgments

This work was funded by the U.S. Department of Energy and the National Nuclear Security Administration's Office of Material Management and Minimization and performed at Pacific Northwest National Laboratory under contract DE-AC05-76RL01830.

Acronyms and Abbreviations

°C	degrees Celsius
BSE	backscattered electron
EDS	energy dispersive spectroscopy
FEM	finite-element method
Mo	molybdenum
PNNL	Pacific Northwest National Laboratory
RVE	representative volume element
SEM	scanning electron microscopy
U-10Mo	uranium alloy with ten weight percent molybdenum (22 atomic percent Mo)
U-Mo	uranium-molybdenum
UTS	ultimate tensile strength
wt%	weight percent
Zr	zirconium

Contents

Figures

1.0 Introduction

In support of the U.S. Department of Energy National Nuclear Security Administration's Office of Material Management and Minimization, Pacific Northwest National Laboratory (PNNL) has been investigating manufacturing processes for uranium-10% molybdenum (U-10Mo) alloy plate fuel. U-10Mo fabricated with low-enriched uranium has been identified as the most promising alternative to the current highly enriched uranium used in the United States' fleet of high-performance research reactors.

One of the defects observed in U-10Mo fuels is nonuniform zirconium (Zr) coating thickness, which can be detrimental to the fuel performance. The Advanced Test Reactor (ATR) Full-size plate In center flux trap Position (AFIP)-7 fuel is one example where this Zr thickness nonuniformity was revealed. AFIP-7 consisted of a monolithic U-10Mo fuel meat with a target thickness of 0.013 in. and a 0.001 in. thick Zr coating on either side of the foil, resulting in an overall foil target thickness of 0.015 in. The foils were fabricated using an as-cast microstructure and later utilizing a two-stage, hot then cold, rolling process to bond the Zr on the U-10Mo. Edwards et al. (Edwards DJ, 2012) performed a detailed microstructure characterization of the rolled fuel through optical microscopy and scanning electron microscopy (SEM). While reviewing these micrographs, a non-uniform thickness of the Zr layers on both sides of the U-10Mo fuel was observed.

Figure 1 represents optical micrographs, at 200× magnification, of the transverse section of a Los Alamos National Laboratory (LANL) fabricated U-Mo foil revealing a mixture of relatively small, elongated grains interspersed with a small fraction of abnormally coarse elongated grains. The Zr layers were found to have a significantly nonuniform thickness, in some cases regions that could be as thick as 35–40 μm as well as localized regions that tapered down to as low as 6–10 μm.

In addition to the abnormally coarse grains, other microstructural features can cause the nonuniform UMo/Zr interface during rolling. Recently, Joshi et al. (Joshi V. V., 2015) characterized various U-10Mo coupons and observed various microstructural aspects that could explain the variability of the Zr coating thickness. Abnormally coarse grains, dendritic structures, or carbides are among these possible causes. Joshi et al. also demonstrated that homogenization can lead to a uniform Mo concentration and elimination of the abnormally coarse grains, which can decrease the U-10Mo/Zr interface roughness. **Figure 2** shows the dendritic structure with Mo-rich regions at the cores of the grains, and Mo-lean regions at the grain boundaries. The micrograph also reveals the presence of carbides and porosity, which when present at the UMo/Zr interface, could lead to Zr thinning or depression on the UMo side during the hot roll bonding process. The graph at the top of **Figure 2** represents Mo concentration from a line scan and shows segregation with concentrations ranging from 3% to 12 wt%.

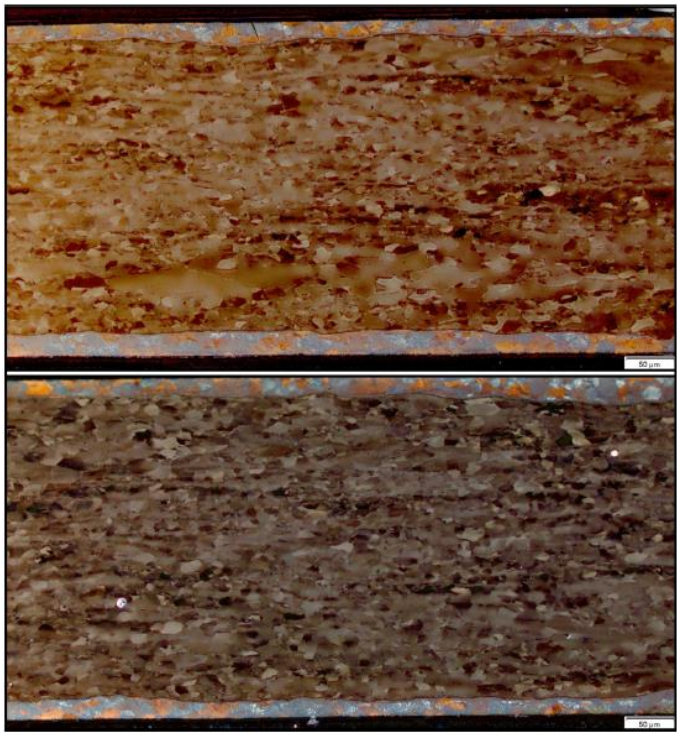

Figure 1. Optical Micrographs Taken at 200× (hydrogen peroxide/colloidal silica final polish) of the Transverse Section of a LANL fabricated U-Mo Foil (Edwards DJ, 2012). The Zr layers are the gray areas above and below the brown fuel.

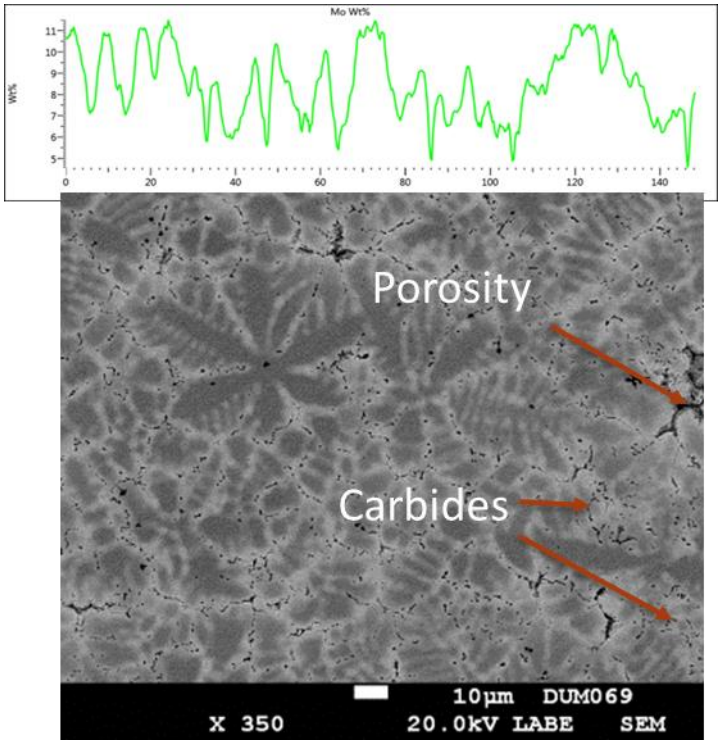

Figure 2. Line Scan of Mo Concentration (top); Backscattered Electron (BSE)-SEM Image of a U-10Mo As-Cast Sample with Mo Concentration Ranging from 3 to 12 wt% (bottom).

To better understand the potential variables associated with the zirconium thickness, a microstructure-based finite-element method (FEM) model was developed to study the effect of homogenization on the U-10Mo/Zr interface. This model uses the homogenization studies by Joshi et al. (Joshi V. V., 2015) (Joshi V. V., 2015) and reconstructed microstructures with Mo concentrations from Xu et al. (Xu, 2016) as an input. Actual microstructures from samples in three conditions will be considered in the current work: 1) as-cast, 2) homogenized for 4 hours at 800°C, and 3) homogenized for 16 hours at 800°C. The model also uses results from a homogenization model as an input, and a study on the effect of different levels of homogenization on the interface indicated that homogenization helps decrease this thinning. This modeling effort will serve as an input to a larger fuel fabrication and performance modeling effort.

2.0 Model Description

The microstructure-based FEM model was developed using actual SEM images and correlated Mo-concentration virtual microstructures. BSE-SEM micrographs for three conditions (as-cast, homogenized at 800°C for 4 hours, and homogenized at 800°C for 48 hours) along with energy dispersive spectroscopy (EDS) and x-ray diffraction data were used to develop a correlation method to reconstruct the Mo concentrations from experimental SEM images. Details about this technique can be found in (Xu, 2016). Figure 3 summarizes this process, starting from an SEM image in Figure 3a; line-scan gray-scale data were extracted and plotted with the Mo concentration from EDS (Figure 3b); then a nonlinear relationship between the image gray scale and Mo atomic concentration was established and plotted (Figure 3c); finally, the Mo concentration map of the entire image was obtained (Figure 3d) by reading the BSE-SEM image and converting the gray scale to the Mo concentration pixel-by-pixel based on the nonlinear relationship from Figure 3(c). Note the Mo concentration segregation in this as-cast material, with Mo-rich regions in red color and Mo-lean regions in yellow color.

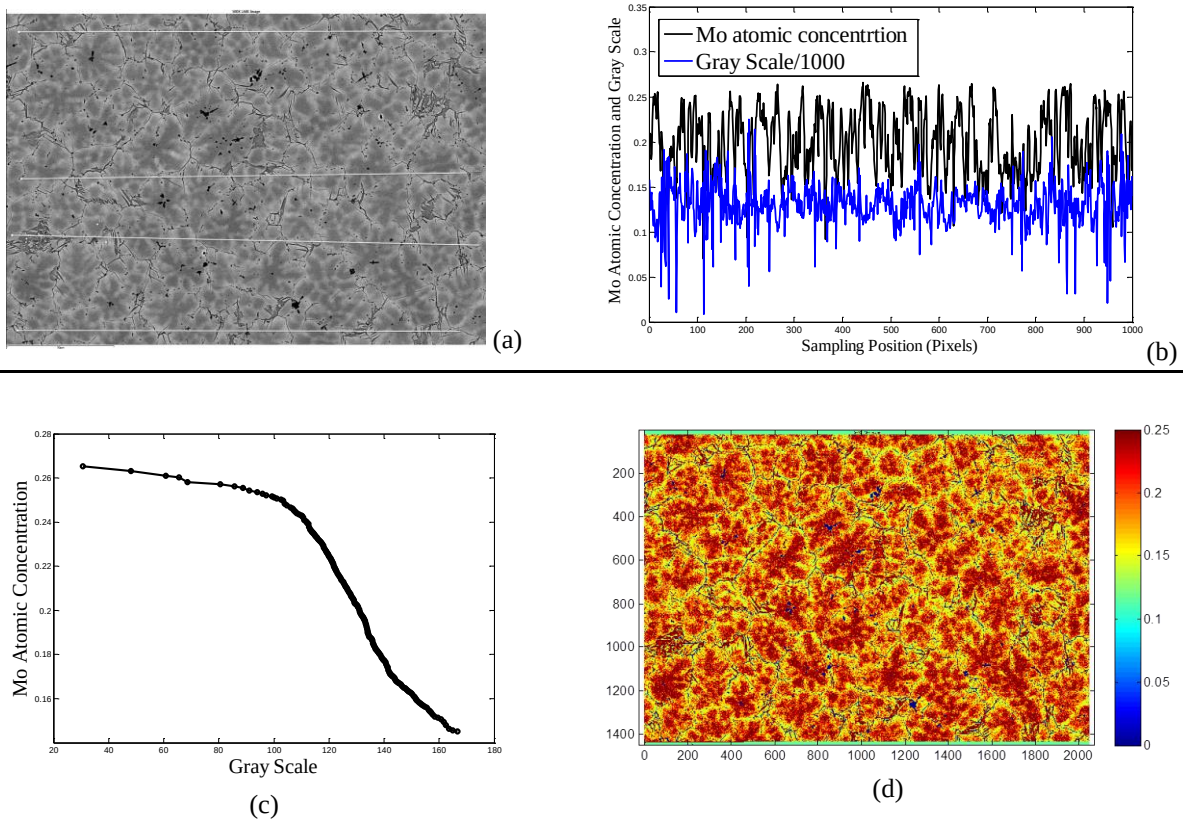

Figure 3. Steps to Obtain Reconstructed Microstructure with Mo Concentrations. (a) BSE-SEM micrograph of as-cast U-10Mo with line scans; (b) EDS Line-scan data of gray scale and Mo concentration; (c) Nonlinear relationship between the image gray scale and Mo atomic concentration; (d) Reconstructed Mo atomic concentration map corresponding to (a); axis units in (d) are the number of pixels. (Xu, 2016)

Next, in order to build the microstructure-based FEM model, the Mo concentration was correlated with mechanical properties. Many authors have published studies where mechanical properties were measured

for U-Mo alloys with various Mo concentrations (Beghi, 1968) (Eckelmeyer, 1976) (Peterson, 1964) (Waldron, 1958) (Burkes, 2009) (Hills, 1964). A detailed literature search on mechanical properties and microstructures of UMo alloys with various Mo concentrations is published in a separate report (McInnis, 2016). Ultimate tensile strength (UTS) as a function of Mo concentration is plotted in Figure 4 using Burkes' (2009) and Hills' (1964) data for room temperature, and (Beghi, 1968) and (Waldron, 1958) data for 600 C. Linear regression fit lines were used to establish the relationship between Mo concentration and UTS of the UMo alloy.

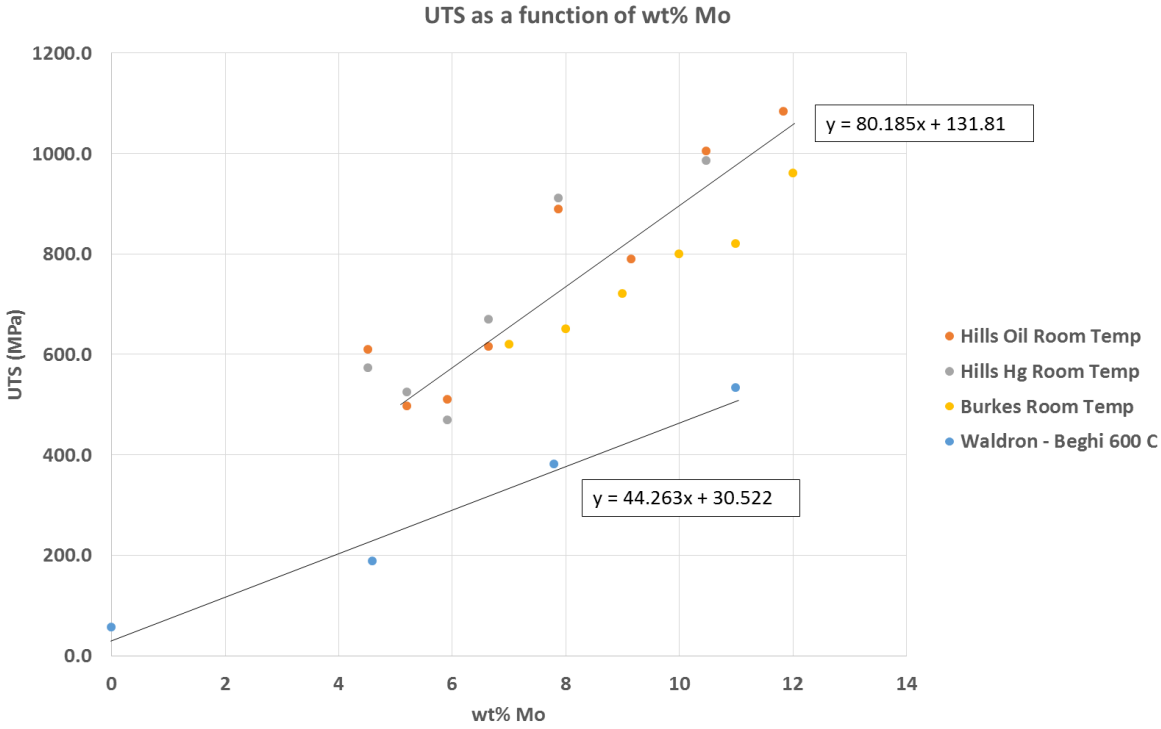

Figure 4. UTS as Function of wt% Molybdenum. Data points from (Beghi, 1968) (Burkes, 2009) (Hills, 1964) (Waldron, 1958)

A 2D FEM model was created using fine mesh with 1 μm × 1 μm element size. This representative volume element (RVE) contains 22,500 elements with a size of 150 μm × 150 μm. Each element was assigned mechanical properties based on its Mo concentration using the correlation functions displayed in Figure 4. A MATLAB script was programmed to generate the FEM model input file. Commercial FEM code LS-DYNA was used to conduct the simulations using explicit formulation. Elements with Mo concentration below 0.1 % were considered carbides with an elastic behavior.

Figure 5 represents the FEM model of the 150 μm × 150 μm RVE. Each color represents one element with a given Mo concentration and the corresponding mechanical properties. Note that the colors are just to differentiate the elements and are not indicative of a particular set of Mo concentration/mechanical properties.

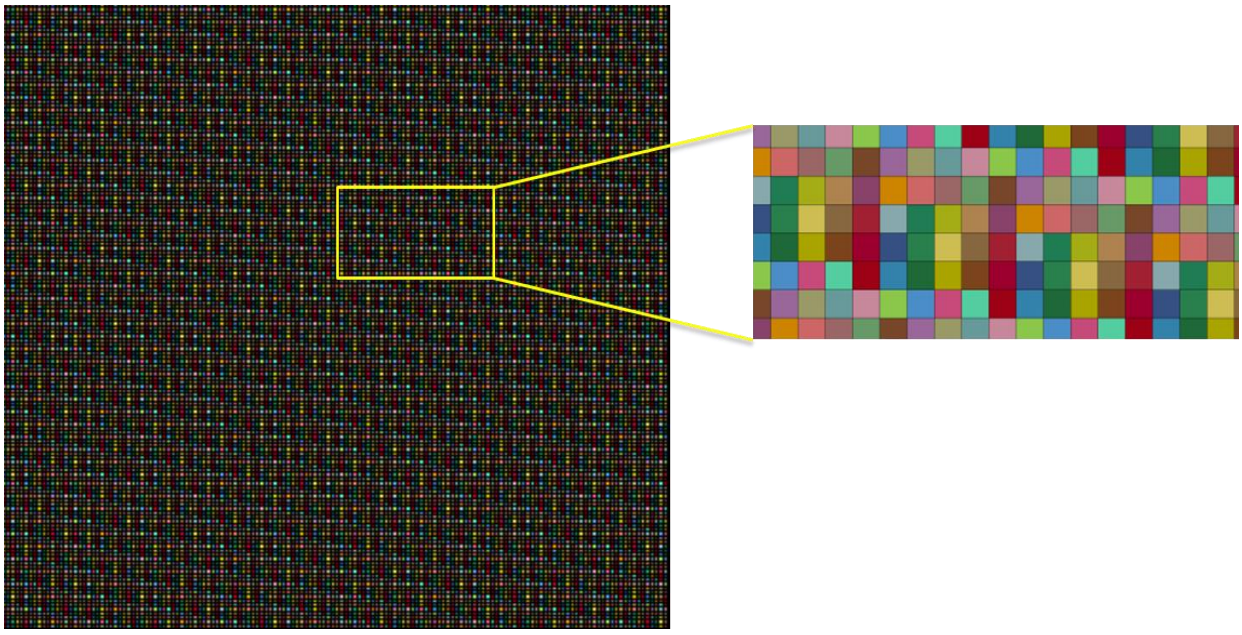

Figure 5. FEM Model. Each color represents a 1 μm × 1 μm element.

3.0 Results and Discussion

3.1 Effect of Homogenization on the Microstructure

As Joshi et al. (Joshi V. V., 2015) showed, homogenization greatly affects the microstructure by changing the carbides' structure, eliminating the dendritic structure of the Mo, and decreasing the segregation in Mo content between the cores of the grains and the grain boundaries. In order to investigate the effect of such homogenization on the U-10Mo/Zr interface using the microstructure-based FEM model, three cases were selected: as-cast material and two levels of homogenization: homogenized for 4 hours at 800°C, and homogenized for 48 hours at 800°C. FEM models were created for each of the three conditions, and slight compression corresponding to 4% thickness reduction was applied to these RVEs. Results show that the Mo segregation was captured by the models in terms of differences in local stresses.

In Figure 6, images 1-a, 1-b, and 1-c represent the Mo-concentrations in the as-cast and homogenized conditions. Images 2-a, 2-b, and 2-c represent the local Von-Mises stresses in the microstructures after compressive loading. We observe that homogenization eliminates the dendritic structure, mainly present in the as-cast condition, where a segregation in Mo-concentrations caused high stresses in the core of the grains (Moly rich region), and low stresses at the grain boundary (Moly lean region). It was also observed that the inclusions (carbides) corresponding to Mo-lean areas (blue color) with concentrations less than 2% are present in the FEM model as well and have lower stresses. Grains are clearly marked in the homogenized microstructures and are also revealed in the FEM model (images 2-b and 2-c). These results (Figures 2-a, 2-b, and 2-c) suggest that a large gradient in material strength within the U-10Mo alloy is not desirable and may be detrimental to the Zr/U-10Mo interface uniformity. In fact, homogenization would improve the UMo/Zr interface given the fact that a uniform stress distribution in the UMo is the best way to avoid Zr thinning. This will be discussed in the next section.

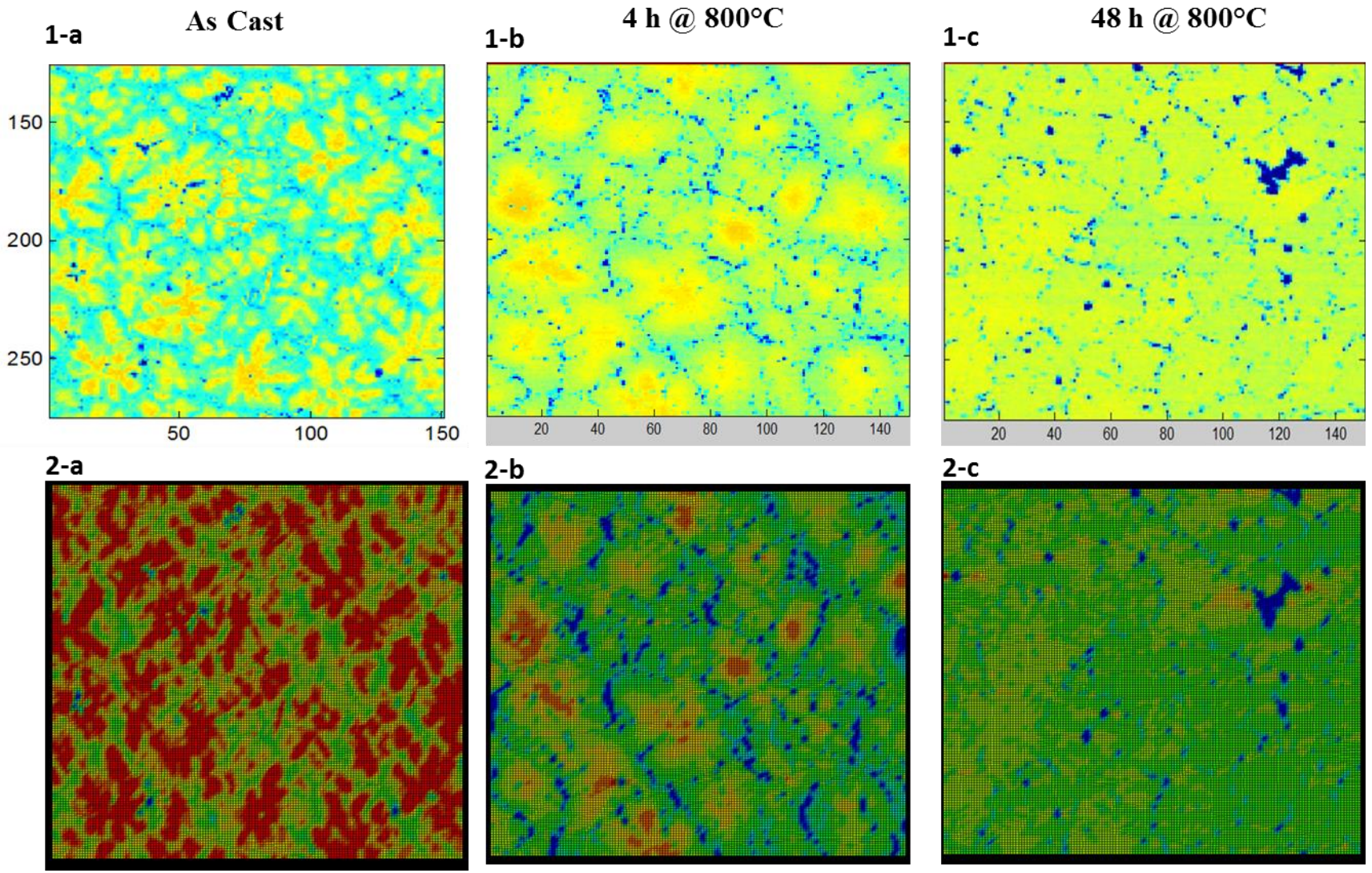

Figure 6. Mo Concentrations (top) and von Mises Stresses (bottom) in the FEM Model after Compression Simulation for As-Cast, Homogenized at 800°C for 4 Hours, and Homogenized at 800°C for 48 Hours.

3.2 Investigation of the UMo/Zr Interface

A 25 µm thick Zr layer was added on top of the U-10Mo RVE. Compressive loads corresponding to 7% thickness reduction were applied to the top of the Zr layer in order to simulate rolling conditions and investigate the effect of the deformation on the UMo/Zr interface at 600 C. Isotropically uniform mechanical properties, extracted from (Garde, Chung, & Kassner, 1977), for Zircaloy-4 were assigned for the Zr. U-10Mo mechanical properties at 600 C are presented in Figure 4. Figure 7 shows the FEM model with the Zr coating, and the U-10Mo fuel.

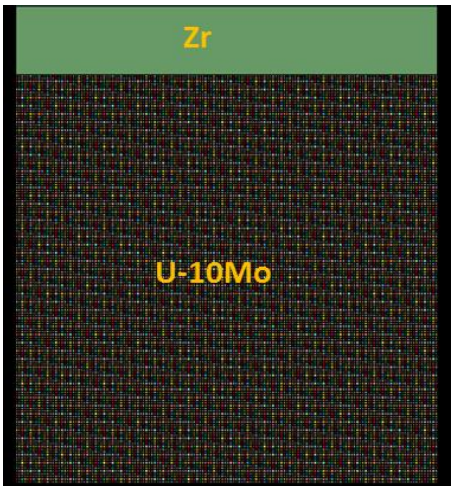

Figure 7. FEM Model Representing U-10Mo with Zr Layer on Top

A closer look at the UMo/Zr interface shows that the model predicted a nonuniform interface in the case of as-cast U-10Mo. A waviness is observed in Figure 8, which is due to the segregation of Mo concentration, and therefore is apparent in stresses in the RVE. The thinning of the Zr layer is less pronounced for the two levels of homogenization studied in this work: 800°C for 4 hours and 48 hours, which are represented in Figure 9 and Figure 10, respectively. Homogenization reduces the variability of the zirconium layer thickness.

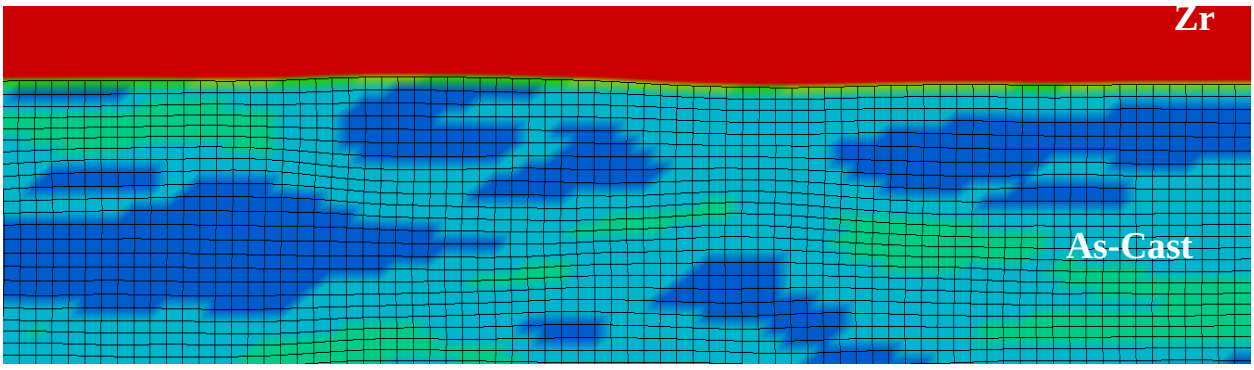

Figure 8. Zr (red) and U-10Mo Interface in the As-Cast Condition

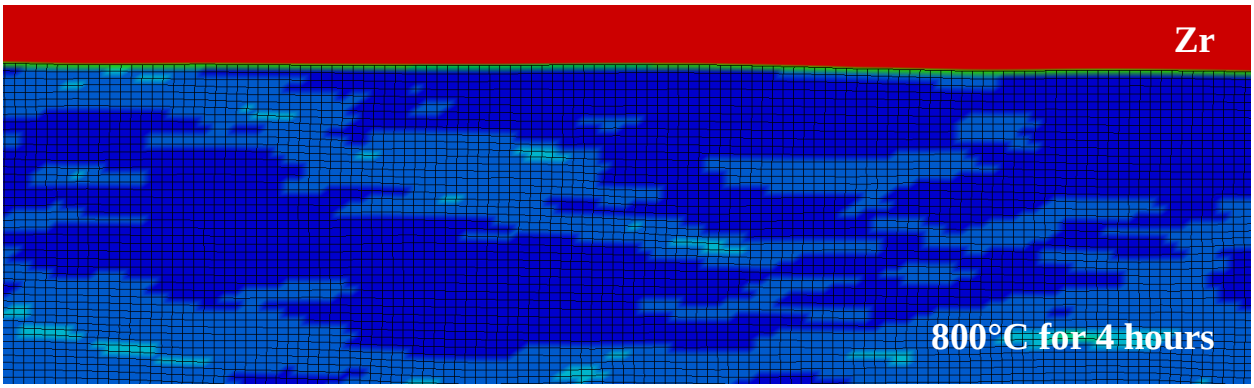

Figure 9. Zr (red) and U-10Mo Interface in the Homogenized at 800°C for 4 Hours Condition

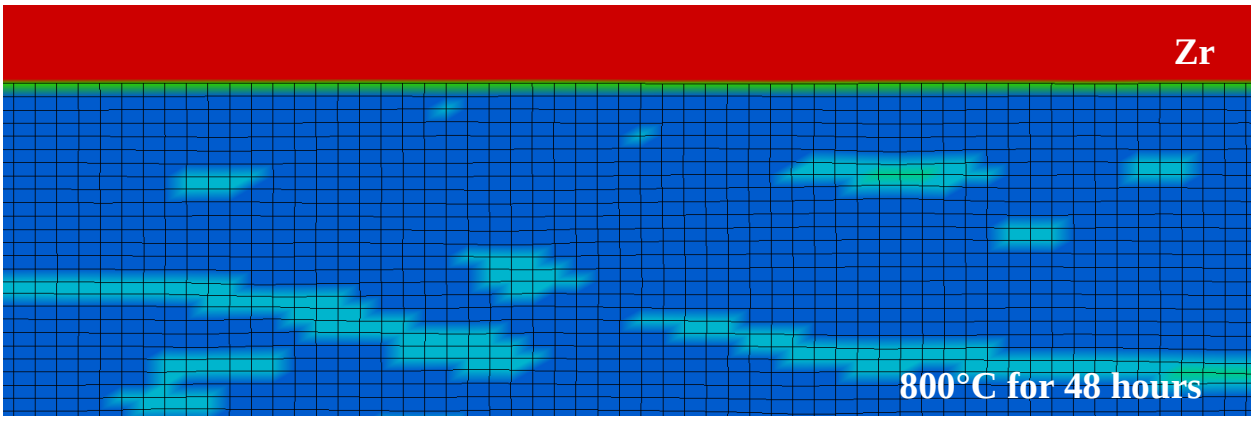

Figure 10. Zr (red) and U-10Mo Interface in the Homogenized at 800°C for 48 Hours Condition

In Figure 11, the nonuniformity of the U-10Mo interface was quantified by plotting the Zr layer thickness along the longitudinal cross-section of the rolled RVE. For the as-cast model, the zirconium layer varied up to 4 μm; the 800°C for 48 hours model varied less than 1 μm.

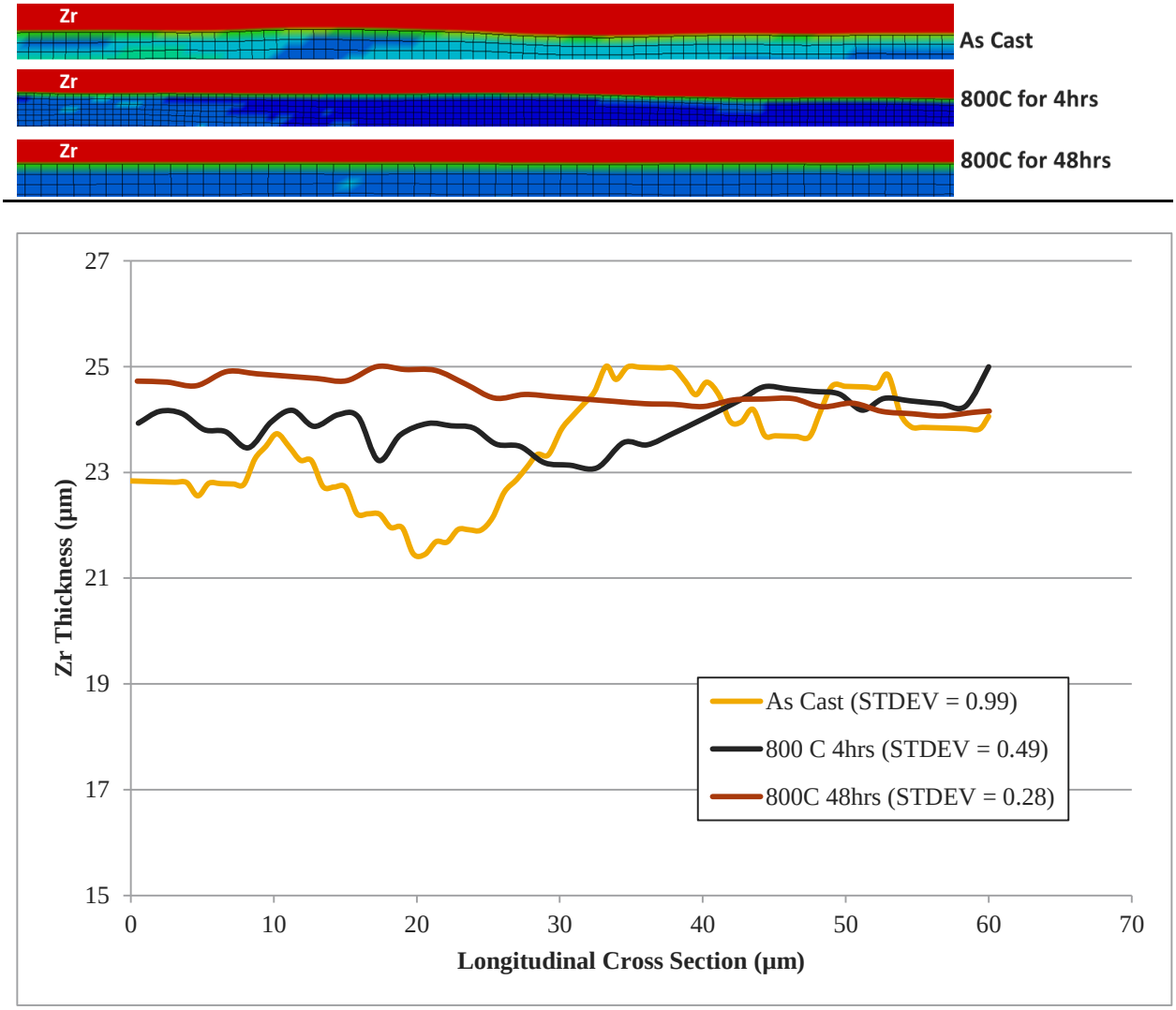

Figure 11. Zr Thickness Variation along the Longitudinal Cross-Section

Grain size is an important aspect of the microstructure that can significantly impact the Zr thinning. Large grains can cause thinning even if the material is homogenized and Mo concentration segregation is diminished. Future work will include a study on the effect of grain size and grain orientation on the UMo/Zr interface.

Carbides are also important and their distribution and morphology crucial to the UMo/Zr interface. Indeed, a large carbide at the surface of UMo can cause significant waviness of the interface. However, small carbides (1 to 2 µm) do not significantly affect the UMo/Zr interface.

4.0 Summary

The goal of this work was to present a microstructure-based FEM approach to investigate the UMo/Zr interface during rolling and study the effect of homogenization on the interface.

1. BSE-SEM images were used with a reconstructed Mo concentration map to create the FEM model, with different mechanical properties for every element based on its Mo concentration.
2. The FEM model accounted for various features present in the actual microstructures, including dendritic structures, grain shapes, and inclusions.
3. Compression simulations using the multilayer model, with Zr on top of U-10Mo, were conducted, and results show the influence of the homogenization on the U-10Mo/Zr interface.
4. Model predictions revealed that homogenization reduces the U-10Mo/Zr interface roughness and results in a uniform Zr layer thickness.
5. Zr thinning was predicted and was more pronounced in the as-cast sample, decreased after homogenization for 4 h at 800°C, and almost disappeared in the material homogenized at 800 for 48 h.

5.0 Future Plans

- As the first attempt of model integration, the model presented in this study shows promising features by integrating the homogenization model with the rolling process model.
- This model will be improved with many new features currently under development.
- Carbides will be added to the microstructure-based FEM model using their appropriate mechanical properties and constitutive behavior during deformation.
- A carbide fracture model will also be implemented in the future; this will allow carbides to fracture under certain conditions.
- The model will also be coupled with an ongoing microstructure-based model that studies carbide distribution during the rolling process.
- Finally, a macro-micro technique will be used to assign more-realistic boundary conditions to the RVE, reproducing the actual rolling conditions.

6.0 References

Beghi. (1968). Gamma Phase Uranium-Molybdenum Fuel Alloys. 6-80.

Boyd, G. A. (1985). The effect of temperature and strain rate on the tensile strength of a fast-gas-cooled uranium-2 wt% molybdenum alloy. *Journal of Nuclear Materials*, 181-191.

Burkes, D. E. et al. (2009). Mechanical properties of DU-xMo alloys with x = 7 to 12 weight percent. *Metallurgical and Materials Transactions*, 1069-1079.

Eckelmeyer, K. A. (1976). The effect of aging on the mechanical behaviours of U-0.75 wt.% Ti and U-2.0 wt.% Mo. *Journal of Nuclear Materials*, 37-49.

Edwards DJ, R. E.-K et al.?. (2012). *Characterization of U-Mo Foils for AFIP-7.* Richland, WA: PNNL-21990, Pacific Northwest National Laboratory.

Garde, A. M., Chung, H. M., & Kassner, T. F. (1977). *Uniaxial Tensile Properties of Zircaloy Containing Oxygen: Summary Report.* Argone National Laboratory, ANL-77-30.

Hills, R. B. (1964). The mechanical properties of quenched uranium-molybdenum alloys. *Journal of Nuclear Materials*, 149-162.

Joshi, V. V. et al.?(2015). Thermomechanical process optimization of U-10 wt% Mo – Part 1: high-temperature compressive properties and microstructure. *Journal of Nuclear Materials*, 805–813.

Joshi, V. V. et al?(2015). Thermomechanical process optimization of U-10wt% Mo – Part 2: The effect of homogenization on the mechanical properties and microstructure. *Journal of Nuclear Materials*, 710-718.

McInnis, C. S. et al?(2016). *Mechanical Properties of Uranim-Molybdenum Alloys at Different Concentrations and Temperatures.* Richland, Wa: PNNL.

Peterson, C. A. (1964). Mechanical Properties of Some Uranium Alloys. *Lawrence Radiation Laboratory*.

Waldron, M. R. (1958). The mechanical properties of uranium-molybdenum alloys. *United Kingdom Atomic Energy Authority*.

Xu, Z. J. et al?(2016). Modeling the homogenization kinetics of as-cast U-10wt% Mo alloys. *Journal of Nuclear Materials*, 154-164.

Distribution

No. of Copies

Department of Energy National Nuclear Security Administration
Office of Material Management & Minimization
1000 Independence Ave.
Washington, DC 20002
Mr. Brian Waud
Mr. Scott Ravenhill

Idaho National Laboratory
P.O. Box 1625
Idaho Falls, ID 83415
Mr. Kenneth Rosenberg
Dr. Barry Rabin

Argone National Laboratory
9700 S Cass Ave.
Argone, IL 60439
Dr. Erik Wilson

No. of Copies

Local Distribution
Pacific Northwest National Laboratory

Jared M. Wight	K8-34
J. Mark Henderson	K8-46
Curt Lavender	K2-44
Vineet Joshi	K2-03
Ayoub Soulami	K2-03
Zhijie Xu	K7-90
Douglas Burkes	K8-34
Eric McGarrah	K8-46

Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** *Since 1965*

U.S. DEPARTMENT OF
ENERGY

902 Battelle Boulevard
P.O. Box 999
Richland, WA 99352
1-888-375-PNNL (7665)

www.pnnl.gov