**Microstructural Anomalies in Hot-Isostatic Pressed U-10 wt.% Mo Fuel Plates
with Zr Diffusion Barrier**

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Abstract

Microstructural anomalies in the co-rolled-and-HIP'ed U–10 wt.% Mo (U10Mo) metallic fuel plate with Zr diffusion barrier assembly were examined as a function of HIP temperature (from 520 to 580°C) and duration (45, 60, 90, 180 and 345 minutes) by scanning and transmission electron microscopy. The anomalies observed in this study are carbide/oxide inclusions within the U10Mo fuel alloy, and regions of limited interaction between the U10Mo alloy and Zr barrier, frequently associated with carbide/oxide inclusions. In the U10Mo alloy, the cF8, Fm3m (225) UC phase ($a = 4.955 \text{ \AA}$) and cF12, Fm3m (225) UO₂ phase ($a = 5.467 \text{ \AA}$) were observed through out the U10Mo alloy with an approximate volume percent of 0.5 to 1.8. The volume percent of the UC-UO₂ inclusions within the U10Mo alloy did not change as functions of HIP temperature and time. These inclusions phases, located near the surface of the U10Mo alloy, were frequently observed to impede the development of interdiffusion and reaction between the U10Mo alloy and Zr diffusion barrier. The regions of limited interaction between the U10Mo and Zr barrier decreased with an increase in HIP temperature, however no noticeable trend was observed with an increase in HIP duration at constant temperature of 560°C.

Keywords: Uranium alloy, Zirconium, Microstructure, Transmission electron microscopy

1. Introduction

U-Mo alloy, in the metastable γ phase, is being considered as the fuel for Global Threat Reduction Initiative (GTRI) program with an objective of developing low-enriched uranium (LEU) fuel to replace high-enriched uranium (HEU) fuel in research and test reactors [1-5]. For GTRI, two types of U-Mo fuels systems, dispersion and monolithic designs, are being developed to increase the U density in LEU fuels. Typically, the dispersion fuel of U-7 wt.% Mo with Al matrix has an Uranium loading of 8 g/cm^3 in Al-alloy matrix [6]. During irradiation, development of fission gas bubbles [7,8] and metallurgical reaction [7-15] has been reported. For higher Uranium loading, up to 15.6 g/cm^3 , monolithic fuel system with U-10 wt.% Mo, which require a Zr diffusion barrier to minimize the adverse metallurgical reaction between fuel alloy and Al-alloy cladding is being considered [16-19]. Robinson *et al.* [20] reported results of tests and qualifications for monolithic fuel plates to find an optimized fabrication process. The U10Mo alloy was recommended as the optimal fuel because of low swelling tendency, excellent co-rolling properties with Zr laminate, and good stability of γ -U at high temperature.

Previous investigations have reported microstructural development in monolithic fuel that consists of U10Mo fuel alloy, Zr diffusion barrier and AA6061 cladding [16,21]. The fuel system is fabricated by sequential process of co-rolling ($\sim 650^\circ\text{C}$) and hot-isostatic press ($\sim 520^\circ$ to 580°C) as schematically illustrated in Figure 1. Perez *et al.* [16] identified several phases, such as UZr_2 , γ -U(Zr), Zr and Mo_2Zr , associated with interaction between the U10Mo and Zr after the co-rolling and HIP process. Park *et al.* [21] further examined phase constituents and microstructure changes as function of temperature (520, 540, 560 and 580°C for 90 minutes) and duration (45~345 minutes at 560°C) of HIP during fabrication. Between the U10Mo and Zr barrier, the interaction zone mainly consist of α -U, Mo_2Zr and UZr_2 phases, which did not grow

significantly during the HIP. Jue *et al.* [22] also evaluated the HIP-bonded monolithic fuels with Zr diffusion barriers. Microstructure of decomposed U-Mo region (e.g., $\gamma\text{-U} \rightarrow \alpha\text{-U} + \text{U}_2\text{Mo}$ eutectoid) depended on the rolling direction. Jue *et al.* [22] also postulated that the Mo_2Zr precipitates formed by the reaction between UZr_2 and the U10Mo matrix. Jue *et al.* [22] furthermore reported complex (i.e., distorted and smeared) microstructure near the edge of the fuel within the cladding and none-Zr-coated fuel region. Huang *et al.* [23] examined the diffusion couples between U10Mo alloy and pure Zr, but were unable to document any significant interaction at temperature below 700°C.

Orlov *et al.* [24] demonstrated the introduction of the carbon impurity by annealing Uranium ingot with carbon isotope additions from 550 to 950°C. They concluded that the carbon impurity diffused into the uranium, and formed both the UC within uranium and U(CO) on the surface of the uranium. According to Nomine *et al.* [25], low solubility of each impurity in uranium causes chemical compound such as the randomly distributed UC phase in U-Mo alloy. Burkes *et al.* [25] examined the mechanical behaviors of U10Mo alloy with carbon, nitrogen and oxygen impurities, and concluded that the failure mode from tensile test was sensitive to impurity content. Hoge [27] also found that carbon content below 250 ppm is desired to prevent reduction of ductility and tensile strength, based on deleterious mechanical effects of carbon on U10Mo alloys. The focus on current investigation is on the influence of UC and UO_2 on the diffusional bonding of U10Mo and Zr through co-rolling and HIP.

In this study, anomalous microstructures in HIP fuel plates were examined by scanning electron microscopy (SEM), transmission electron microscopy (TEM) and X-ray energy dispersive spectroscopy (XEDS). The anomalous features are the U10Mo-Zr bonding with limited interdiffusion and reaction (i.e., absence of distinguishable interaction zone consist of α -

U, Mo₂Zr and UZr₂) typically impeded by the UC and UO₂ phases randomly present in the U10Mo alloy.

2. Experimental details

The U10Mo alloy used in this investigation was fabricated by arc-melting the U (depleted) and 99.95% pure Mo. **Prior to melting, the U was cleaned with 30% nitric-acid.** The U10Mo alloy was flipped/turned and remelted three times to ensure the homogeneity of compositions in a vacuum of 1.3×10^{-3} Pa (10^{-5} torr) having less than 50 ppm oxygen. Then, the alloy was cast into coupon-shaped ingots using a graphite mold. The alloy had approximate dimension of 88.8 mm $\times$ 48.9 mm $\times$ 3.2 mm and a carbon content of approximately 700 ppm. This ingot was then acid cleaned using a mixture of nitric and hydrofluoric acid (2.5% HF, 35% HNO₃ and 62.5% H₂O), and then laminated, in a carbon steel can, using pure Zr (99.9% pure) foil with a starting thickness of 250 micrometer on each surface. **The commercially-available reactor-grade pure Zr was etched using ~30% nitric acid solution prior to lamination.** The Zr-laminated U10Mo coupon was pre-heated at 650°C for 30 min in a furnace, and then co-rolled 15 times to a thickness of approximately 0.46 mm. A post-rolling heat treatment was then performed at 650°C for 45 min. The hot-rolled/annealed foil was then removed from the can, and cold-rolled to a final thickness of 0.33 mm. The dimension of the final as-rolled foil was **approximately 746 mm $\times$ 65 mm $\times$ 0.33 mm.** The total time exposure at 650°C was estimated at approximately 130 minutes. The final thickness of the Zr diffusion barriers on each surface were approximately 25 μ m. The final as-rolled U-Mo foil was then sectioned into smaller foils. All the small U-Mo foils used in this study were sectioned from the same parent foil. **Each foil sample, in general,**

exhibited the same microstructure of the co-rolled U10Mo/Zr laminate [21]. The as-rolled/sectioned sample before the HIP process is labeled as sample AR in this study.

Each U10Mo/Zr mini foil was polished, cleaned using nitric and hydrofluoric acid (2.5% HF, 35% HNO₃ and 62.5% H₂O), rinsed with ethanol, and stacked with AA6061 (in wt.% nominal composition of 0.6Si-0.35Fe-0.28Cu-0.08Mn-1.0Mg-0.20Cr-0.13Zn-0.08Ti-Al balance) cladding pieces and HIP'ed at various temperatures (520, 540, 560 and 580 °C) and durations (45, 60, 90 180 and 345 minutes). Before the HIP'ing process, the AA6061 cladding was etched using a 1.85 M NaOH rinse, followed by pickling in 30% nitric acid in order to remove the residual oxide scale on the Al alloy surface. The U10Mo/Zr/AA6061 assemblies were then inserted into stainless steel HIP cans with tool steel strong-backs that were welded with corners open for degassing in an argon atmosphere glove box. The assemblies were leak checked and vacuum degassed at 315 °C for 3 h. The HIP can was then loaded into a laboratory-size HIP, and heated to the target temperature with a ramp-up and cool-down rate of 280°C per hour. The isostatic pressure of the HIP process was kept constant at 103 MPa (15 ksi) in an argon environment. Schematic illustrations of the co-rolling and HIP processes are presented in Figure 1 [21], and the samples examined in this study are listed in Table I.

The fabricated fuel plates were then cross-sectioned and mounted in epoxy. The mounted fuel plates were polished using silicon carbide papers (240, 600, 800 and 1200 grit) with an ethanol lubricant and subsequently final polishing was carried out utilizing diamond pastes (3 and 1 µm) with an oil lubricant in a glove box under Ar atmosphere, preventing oxidation and contamination. Microstructural features at the U10Mo-Zr interface were examined by using Zeiss ULTRA-55 FEG SEM with XEDS. Transmission electron microscopy (FEI/Tecnai™ F30) equipped with Fischione™ high angle annular dark-field (HAADF) and XEDS was used to

examine the detailed microstructure and identify the phase constituents via selected area electron diffraction (SAED) and high resolution TEM (HRTEM). The TEM samples were prepared by focused ion beam and in-situ lift-out was utilized for extraction (FIB-INLO; FEI™ TEM200) using site-specific capability. Typical phase constituents and microstructural development in these samples as a function of HIP temperature and time were reported previously [21]. This paper reports, in detail, the anomalies, as defined above, observed in these fuel plate samples. Quantitative measure of microstructural features, such as areal percent of inclusions phases within the U10Mo alloy and linear percent of interface with limited interdiffusion-reaction, was carried out for the entire cross-sectional areas (25.4 mm x 0.33 mm) and interfacial length (25.4 mm on both sides) using various and appropriate magnifications.

3. Phase Constituents and Microstructure of Anomalous Features

Figure 2(a) shows typical BSE micrographs from the U10Mo monolithic fuel clad in AA6061 with Zr barrier. Typically, the interdiffusion zone between the U10Mo alloy and α -Zr diffusion barrier consisted of UZr_2 layer, discrete Mo_2Zr and α -U layer near the U10Mo/Zr interface [21] as shown in Figure 2(b) with relatively uniform thickness. However, there were several randomly-distributed inclusions (identified as UC and UO_2 by TEM) observed within the U10Mo alloys, and some located near the interface between the U10Mo alloy and α -Zr diffusion barrier appeared to influence the phase and microstructural development. Figure 3 presents BSE micrographs that are considered “microstructural anomalies” in this investigation: Figure 3(a) presents a BSE micrograph from the region where apparent presence of these inclusions has limited the interdiffusion and reaction between U10Mo and Zr diffusion barrier; Figure 3(b) presents a BSE micrograph from the region where limited interdiffusion and reaction was

observed between U10Mo and Zr diffusion barrier without the apparent presence of the inclusions, at least by SEM. Two TEM samples were prepared by FIB-INLO as indicated in in Figure 3 to elucidate the phase constituents of the inclusions and characterize the regions of limited interdiffusion and reaction. The region of limited interaction in Figure 3(a) to the right-hand-side of the apparent UC and UO₂ inclusions, is assumed to be similar to that presented in Figure 3(b) studies by TEM.

Figure 4 presents (a) HAADF scanning TEM (STEM) and (b) BF micrographs of inclusions in TEM sample prepared from Figure 3(a). The UC phase was identified via SAED patterns along the $[\bar{1}12]$ and $[1\bar{1}\bar{1}]$ beam directions as shown in Figure 5. The UO₂ phase, consisted of many fine grains, some containing a significant amount of C, was identified by HRTEM micrographs with fast Fourier transformation (FFT) analyses as presented in Figure 6. The UC and UO₂ phases were identified as cF8, Fm-3m (225) with $a = 4.955 \text{ \AA}$ and cF12, Fm-3m (225) with $a = 5.467 \text{ \AA}$, respectively. The crystallographic details for the UC and UO₂ phases along with typical chemical compositions observed by semi-quantitative standard-less XEDS are reported in Table II.

Figure 7 shows HAADF micrograph from the TEM sample that had limited interdiffusion and reaction between the U10Mo and Zr diffusion barrier without the apparent presence of the inclusions. Although not observed by SEM, a significant amount of UC and UO₂ phases were identified on the U10Mo fuel next to Zr barrier as “varying” contrasts (i.e., composition and orientation sensitive) in Figure 7. Figure 8(a) shows that the presence of UC and UO₂ phases near the surface of U10Mo in contact with Zr barrier by BF TEM micrograph. Both UC and UO₂ phases were confirmed by FFT analyses as presented in Figure 8(b) and 8(c). Figure 9 shows the XEDS elemental mapping from the region shown in Figure 7. Presence of UC and UO₂ phases

near the Zr within the γ -phase U10Mo alloy is distinguished by Mo depletion, while distinction between the UC and UO_2 is apparent by contrast in U content: higher in UC than UO_2 .

4. Microstructural Anomalies and Fuel Fabrication

The UC and UO_2 phases in the U10Mo alloy matrix were observed in all fuel plate samples listed in Table I. Figure 10 shows BSE micrograph with highlights that distinguishes the UC and UO_2 phases. Although the morphology of the UC and UO_2 phases clearly influenced by the co-rolling, it is assumed that the area percent by image analysis approximate the volume percent for these phases with limited plasticity. Area percent of the UC and UO_2 estimated for all samples examined in this study are listed in Table I. The amount of these inclusions did not change during HIP: as a function of time at 560°C as presented in Figure 11(a); and as a function of temperature after 90 minutes as presented in Figure 11(b). Therefore the UC and UO_2 phases are presumed to come from the initial casting process, and their amount is not altered by HIP.

In all samples examined in this study, regions of limited interdiffusion and reaction were observed as presented in Figure 3(b). They were observed typically with the presence of UC and UO_2 phases as presented in Figures 7, 8 and 9. The linear length of these regions and the total length of the fuel alloy in contact with Zr barrier were measured to estimate the linear percent of regions where the interdiffusion and reaction were significantly impeded. Figure 12 shows an example to illustrate how the fractional linear length of the limited interdiffusion-reaction zone was measured (i.e., L_i/L_T) over the entire fuel plate sample. Table I reports the percent of length associated with limited interdiffusion and reaction. Standard deviation in percent measurement was obtained by three independent measurements.

Figure 13(a) presents the amount of region with limited interdiffusion and reaction as a function of HIP temperature with a constant holding time of 90 minutes. Despite the interference of the UC and UO_2 phases on the interdiffusion and reaction, there is a noticeable decrease in this microstructural anomaly as a function of temperature. However as a function of HIP time at 560°C , no observable trend was observed as shown in Figure 13(b). A decrease in the amount of regions with limited interdiffusion and reaction may involve complex phenomena including oxide/carbide break-up, thermally-activated interdiffusion via lattice and interfaces with geometrical barriers, nucleation and growth. Within the parameters of temperature and time variation in this study, only the temperature dependence was observed.

The UC and UO_2 phases in the U10Mo alloy appear to originate from the C and O impurities during the casting based on this investigation [24,25]. Amount of these inclusions did not change during HIP within the U10Mo alloy. Their presence near the surface of the U10Mo alloy in contact with Zr diffusion barrier impeded the interdiffusion and reaction that produce UZr_2 layer, discrete Mo_2Zr and $\alpha\text{-U}$ layer after HIP. However the region of limited interdiffusion and reaction disappeared noticeably with an increase in HIP temperature, however not with longer HIP duration examined in this study.

Distribution of UC and UO_2 inclusions in the U10Mo fuel alloy can cause unexpected physical, mechanical and chemical behavior of the monolithic fuel plate [24-27]. This manuscript reported some anomalies in chemical interactions between the fuel alloy and Zr diffusion barrier. Table II, in addition to crystallographic details and compositions, lists some of the physical and mechanical properties for the UC, UO_2 and γ (U-10 wt.% Mo) phases [24-34]. During the fabrication of the monolithic fuel plate, large differences in elastic modulus and

hardness may cause crack formation within the low-toughness phase. Cracks were frequently observed in the UO_2 phase as presented in Figures 3 and 4 in this study.

5. Summary

The microstructural anomalies in the U10Mo fuel alloy and near the interface between the U10Mo fuel alloy and Zr diffusion barrier were examined by SEM and TEM/STEM. The cF8, Fm-3m (225) UC phase and cF12, Fm-3m (225) UO_2 phase were identified to be distributed through out the U10Mo alloy. Volume percent of these inclusions ranged from 0.5 to 1.8, and did not change as functions of HIP temperature and time. The presence of UC and UO_2 inclusions near the surface of the U10Mo alloy in contact with Zr diffusion barrier produced regions of limited interdiffusion and reaction observed by cross-sectional microscopy. HIP at higher temperature partially removed these regions of impeded interdiffusion and reaction. **However within the HIP duration examined in this study at constant temperature of 560°C, no noticeable change was observed as a function of time.** Difference in physical, mechanical and chemical properties/interactions of UC, UO_2 , and γ -U10Mo phases can influence structural integrity of the U10Mo monolithic fuel with Zr diffusion barrier.

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Table I. HIP plate samples examined in this study along with estimated quantity of microstructural anomalies.

Sample	Temperature (°C)	Hold Time (min)	Area percent of the UC and UO ₂ phases in U-10Mo (%)	Limited interdiffusion-reaction zone (%)
AR	N.A.	N.A.	1.24	18.10 ± 0.54
52-90	520	90	1.53	19.90 ± 0.49
54-90	540	90	0.49	16.65 ± 0.20
56-45	560	45	1.78	14.99 ± 0.39
56-60	560	60	1.18	20.47 ± 0.47
56-90	560	90	1.39	11.05 ± 0.16
56-180	560	180	1.41	21.71 ± 0.51
56-345	560	345	1.64	10.90 ± 0.38
58-90	580	90	1.32	9.75 ± 0.15

Table II. Crystallographic details, properties, and chemical compositions for the UC, UO₂ and γ (U-10wt.%Mo) phases [26, 28-37]. The chemical compositions reported are typical values observed in this study by semi-quantitative standard-less XEDS.

Phase		UC	UO ₂	γ (U-10wt.%Mo)
Pearson Symbol		cF8	cF12	cI2
Space group		Fm3m (225)	Fm3m (225)	Im3m (229)
Lattice parameter (Å)		4.955	5.467	3.409
Elastic modulus (GPa)*		215.7 [28]	172.6 [29]	89.2 [26]
Thermal expansion coefficient (/K)		~10 ⁻⁵ [28]	~10 ⁻⁵ [29]	~10 ⁻⁵ [26,30]
Melting point (°C)		2275 [31]	2878 [32]	~1250 [33]
Density (g/cm ³)*		13.63 [34]	10.27 [35]	17.2 [36]
Hardness (GPa)*		9.1 [31]	6.5 [29]	3.1 [37]
Composition (at.%)	U	30.2	33.1	78.4
	Mo	2.3	2.1	21.6
	Zr	0.4	1.0	N.A.
	O	0.0	10.2	N.A.
	C	67.1	53.6	N.A.

* At room temperature

List of Figures

- [1] Schematic diagrams of (a) co-rolling and (b) hot-isostatic pressing employed in this investigation to produce U-10 wt.% Mo monolithic fuel encased in AA6061 with Zr diffusion barrier [21].
- [2] Backscatter electron micrographs of the U-10 wt.% Mo monolithic fuel encased in AA6061 with Zr diffusion barrier: (a) overall fuel plate assembly, and (b) the interface between U-10 wt.% Mo and Zr diffusion barrier, produced by co-rolling and hot-isostatic pressing (sample 56-345, hot-isostatic pressed at 560°C for 345 min).
- [3] (a) Anomalous microstructure associated with the UC and UO_2 phases, and (b) the imperfect reaction zone in U-10 wt.% Mo alloy. (From sample 56-345, HIP'ed at 560°C for 345 min).
- [4] (a) High angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) micrograph and (b) bright field (BF) micrograph from the UC and UO_2 inclusion phases. (TEM specimen prepared from Figure 3(a) from sample 56-345, HIP'ed at 560°C for 345 min).
- [5] Selected area electron diffraction patterns from the UC phase in (a) [112] and (b) [111] beam direction. (TEM specimen prepared from Figure 3(a) from sample 56-345, HIP'ed at 560°C for 345 min).
- [6] High resolution TEM micrographs with the fast Fourier transform analysis along the $\begin{bmatrix} 01\bar{1} \end{bmatrix}$ and $\begin{bmatrix} 001 \end{bmatrix}$ beam direction for the UO_2 phase. (TEM specimen prepared from Figure 3(a) from sample 56-345, HIP'ed at 560°C for 345 min).
- [7] HAADF STEM micrograph of the region with limited interdiffusion and reaction near the interface between the U10Mo alloy and Zr diffusion barrier. Contrast developed by composition and orientation shows UC (white arrows) and UO_2 (black arrows) phase. (TEM specimen prepared from Figure 3(b) from sample 56-345, HIP'ed at 560°C for 345 min).
- [8] (a) Bright field micrograph and high resolution TEM micrographs with the fast Fourier transform analyses for the (b) UC and (c) UO_2 phases. (TEM specimen prepared from Figure 3(b) from sample 56-345, HIP'ed at 560°C for 345 min).
- [9] XEDS elemental mapping of Figure 7. (TEM specimen prepared from Figure 3(b) from sample 56-345, HIP'ed at 560°C for 345 min).

- [10] The measurement of area percent for the UC and UO_2 phases distributed in the U-10 wt.% Mo alloy.
- [11] Estimate volume fraction of the UC and UO_2 inclusion phases distributed in the U-10 wt.% Mo monolithic fuel after hot-isostatic pressing (a) at 560°C as a function of time, and (b) after 90 minutes as a function of time. “AR” refers to the estimated volume fraction in as-rolled sample prior to HIP.
- [12] Illustration of the linear measurement for regions with limited interdiffusion and reaction between the U-10 wt.% Mo alloy and Zr diffusion barrier.
- [13] Estimated area of limited interdiffusion and reaction between the U-10 wt.% Mo and Zr diffusion barrier during HIP as (a) a function of temperature after 90 minutes and (b) a function of time at 560°C . “AR” refers to the estimated volume fraction in as-rolled sample prior to HIP.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Figure 13