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ABSTRACT

Monolithic fuel plates are being developed for application in research reactors throughout the world. These fuel plates are comprised of a U-Mo alloy foil encased in aluminum alloy cladding. Three different fabrication techniques have been looked at for producing monolithic fuel plates: hot isostatic pressing (HIP), transient liquid phase bonding (TLPB), and friction stir welding (FSW). Of these three techniques, HIP and FSW are currently being emphasized. As part of the development of these fabrication techniques, fuel plates are characterized and tested to determine properties like hardness and the bond strength at the interface between the fuel and cladding. Testing of HIPed samples indicates that the foil/cladding interaction behavior depends on the Mo content in the U-Mo foil, the measured hardness values are quite different for the fuel, cladding, and interaction zone phase and Ti, Zr and Nb are the most effective diffusion barriers. For FSW samples, there is a dependence of the bond strength at the foil/cladding interface on the type of tool that is employed for performing the actual FSW process.

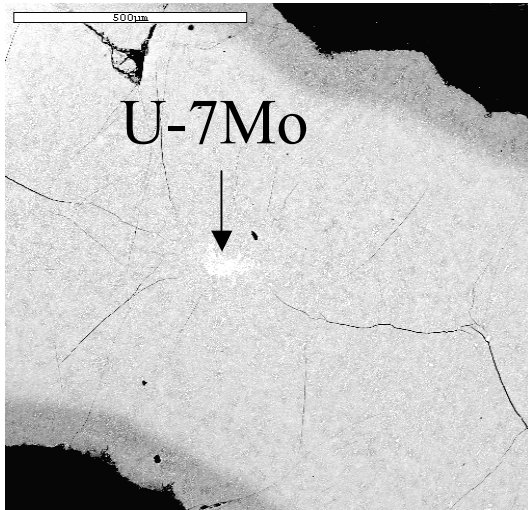
1. Introduction

Monolithic fuel plates are being developed as an LEU fuel for application in research reactors. [1]. To fabricate these fuels, three techniques have been evaluated: friction stir welding (FSW), hot isostatic pressing (HIP), and transient liquid phase bonding (TLPB), and the current emphasis is on FSW and HIP. As part of this evaluation, samples have been generated from fuel plates that were fabricated using the emphasized techniques, which were then characterized and tested. The characterization was performed using both scanning electron microscopy and optical microscopy. The testing was conducted using either hardness testing, which was performed on FSW and HIP samples, or pull testing, which was conducted only on FSW samples. The pull test was employed to investigate the bond strength at the foil/cladding interface in a monolithic fuel plate. This paper will discuss the results from the characterization and testing that was performed on various samples that were produced using FSW and HIP.

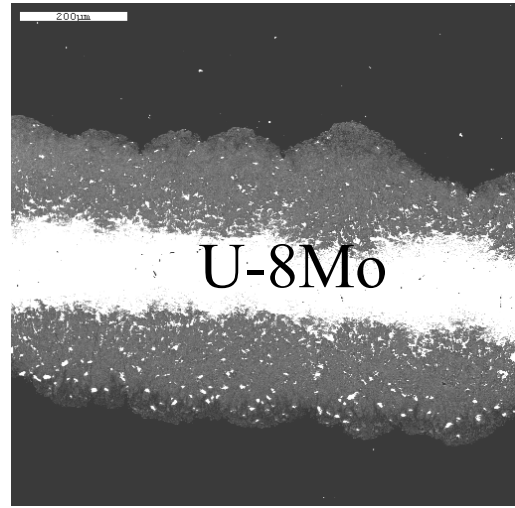
2. Results and Discussion

2.1 Hot Isostatic Pressing

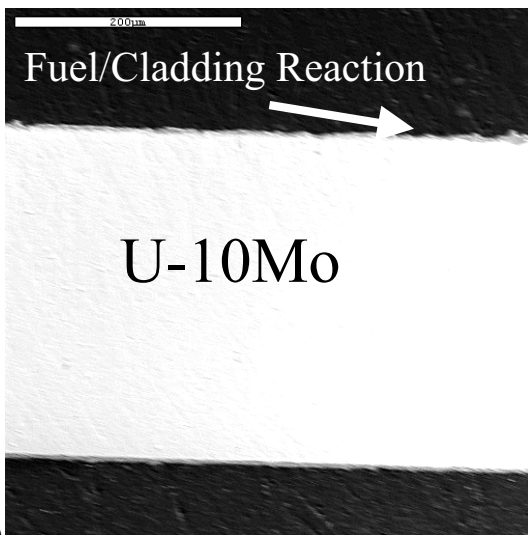
The HIP samples that were characterized were HIPed at 580°C for 90 minutes using a pressure of 15,000 psi. Figure 1 shows SEM micrographs for samples with 6061 Al cladding and U-x wt% Mo (x=7, 8, 10, and 12) fuel alloys. For the U-7Mo sample, the original fuel alloy is completely consumed. Another U-7Mo sample was run for 90 minutes at 580°C that was not completely consumed. Composition analysis of the completely consumed samples showed that the phases that formed were (U,Mo)Al₃ and (U,Mo)_{0.9}Al₄. Only a small region of original U-7Mo alloy remained at the center of the sample. For a U-7Mo sample HIPed at 580°C for half the time, the amount of reaction was less. This sample showed that the main phase to develop in the interaction zone is (U,Mo)Al₃, along with some minor phases at the reaction zone/6061 Al interface that contain component like Mg and Fe (see Figure 2). Nearest the unreacted U-7Mo is a multiphase zone that is commonly seen in diffusion couple experiments (see Figure 3) [2]. The



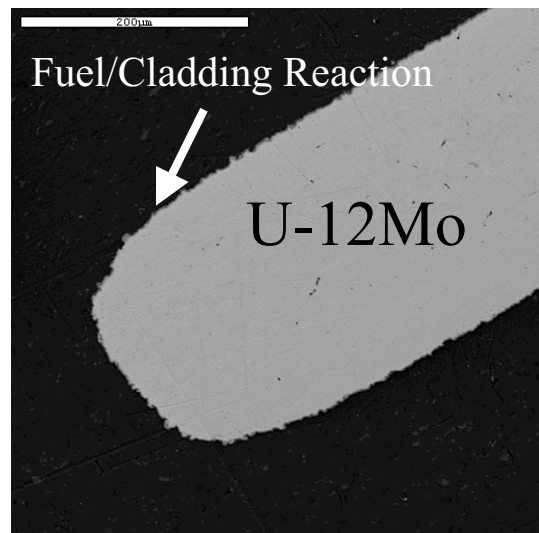
(a)



(b)

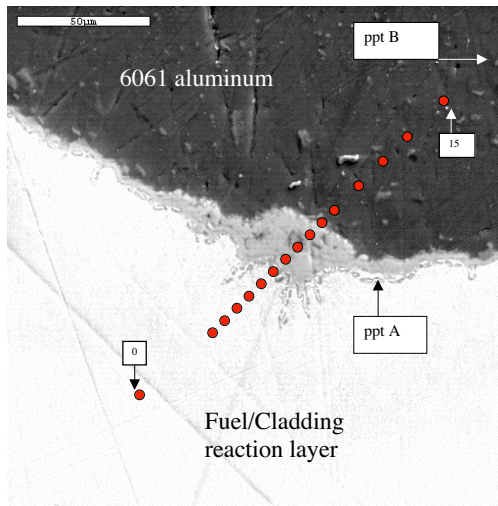


(c)



(d)

Figure 1. SEM micrographs of the (a) U-7Mo (580°C; 3 hrs), (b) U-8Mo (580°C; 1.5 hrs), (c) U-10Mo (580°C; 3 hrs), and (d) U-12Mo (580°C; 1.5 hrs) foils after HIPing. The black areas are 6061 Al cladding and the medium-contrast areas are interaction product.

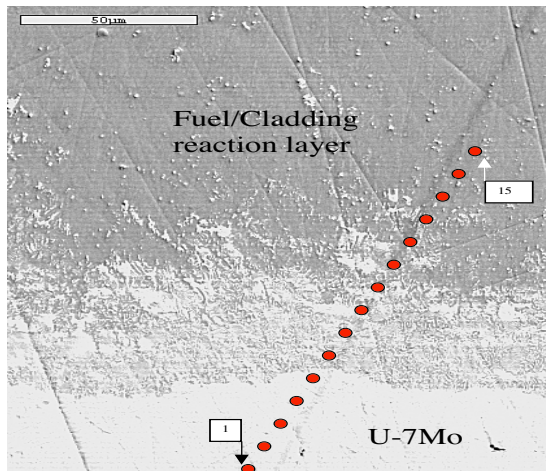


	U (at %)	Mo	Al	Mg	Si	Fe
0	20.44	2.46	75.69	2.58	-	-
1	19.14	2.57	74.71	2.22	2.09	-
2	17.33	2.30	75.95	3.42	1.57	-
3	18.79	2.86	75.66	2.09	0.68	-
4	16.50	1.86	73.66	3.35	4.95	-
5	15.33	3.90	74.53	2.46	4.32	-
6	8.19	2.94	71.54	13.55	0.44	3.33
7	6.58	0.41	79.13	2.06	-	12.61
8	3.73	0.25	90.41	4.23	-	1.84
9	5.48	-	78.86	2.23	0.29	13.51
10	-	-	97.23	3.20	-	-
11	-	0.33	92.44	3.74	-	2.42
12	-	0.35	96.62	2.00	0.82	0.25
13	-	0.11	98.09	1.6	0.34	-
14	-	0.03	98.88	1.29	0.07	-
15	0.03	-	97.31	1.3	1.39	0.13
ppt A	28.18	6.95	47.07	18.95	0.82	-
ppt B	-	0.18	92.59	1.01	2.78	3.67

(a)

(b)

Figure 2. (a) SEM micrograph of interdiffusion zone nearest the unreacted 6061 Al for U-7Mo plate HIPed at 580°C for 90 minutes at 15,000 psi. (b) Table showing compositions at various points in (a) going from 1 to 15.



	U (at %)	Mo	Al
1	80.42	13.73	5.84
2	80.35	15.17	4.49
3	86.33	11.51	2.17
4	82.92	10.95	6.13
5	81.88	15.02	3.10
6	51.69	5.79	42.52
7	49.35	8.13	42.52
8	48.52	9.08	42.39
9	43.11	8.45	48.44
10	29.80	4.82	65.38
11	19.79	4.63	75.53
12	19.75	3.28	76.96
13	20.53	2.11	77.36
14	20.31	4.04	75.65
15	21.44	2.72	75.84

(a)

(b)

Figure 3. (a) SEM micrograph of interdiffusion zone nearest the unreacted fuel for U-7Mo plate HIPed at 580°C for 90 minutes at 15,000 psi. (b) Table showing compositions at various points in (a) going from 1 to 15.

U-8-Mo sample displays less interaction between the fuel and the cladding, but a significant amount of interaction product still forms, and a significant quantity of U-rich precipitates are distributed throughout the interaction zone. For the U-10Mo and U-12Mo samples, only a few-micron-thick layer forms at the foil/cladding interface. From looking at these different samples, it is clear that at 580°C there is a significant correlation between the Mo content of the fuel and the amount of foil/cladding interaction that occurs during HIPing. By looking at the U-Mo phase diagram [3], it can be seen that at around 562°C and at about 10.5 wt% Mo, a eutectoid is present. For the hypo-eutectoid U-7 and 8 wt% Mo alloy compositions, the alloys are in a two-phase region consisting of α -U and γ -U at 580°C. Yet, the U-10 wt% Mo alloy composition is very near the $\gamma/\alpha + \gamma$ phase field boundary and is probably γ -phase. The U-12 wt% Mo alloy is in the $\gamma + \delta$ two-phase region. When HIPing is performed at 560°C, all the alloys will be in the $\alpha + \delta$ two-phase region and how quickly the various alloys transform to these two phases will affect how quickly the alloys react with the 6061Al alloy cladding. Once α -U is present in the alloy microstructure, the amount of interaction should increase, since it has been shown that α -U reacts much more quickly with Al than does γ -U [2].

To reduce reaction between the fuel and cladding a variety of diffusion barriers (Zr, Nb, Ta, C, Si, and Ti) have been evaluated. Figure 4 shows a cross section of a sample where a Zr foil was employed as a diffusion barrier. Bonding between the Zr and the Al was good. Based on ultrasonic testing (UT), there was also bonding between the Zr and the fuel foil. Yet, during the destructive examination process the Zr de-bonded from the fuel foil. This indicates that the bond quality was relatively poor. Ti and Nb exhibited behaviors similar to that of Zr. Ni was tried as a diffusion barrier, but extensive interaction with Al to produce nickel-aluminides precludes using Ni as a diffusion barrier. For C and Mo, UT analysis indicates poor bonding between these materials and the foil and 6061 Al. Initial UT analysis suggests that Ta is a good diffusion barrier, but ongoing SEM examinations must be completed to confirm this result.

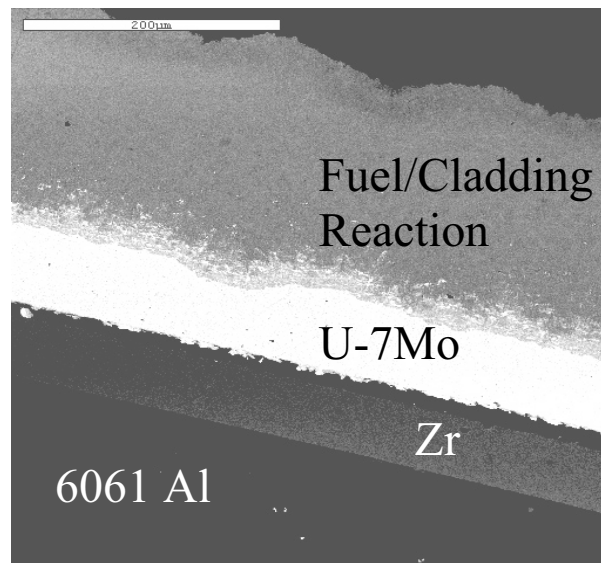


Figure 4. SEM micrograph of a U-7Mo fuel plate with a Zr liner used as a diffusion barrier that was HIPed at 580°C for 90 minutes. There is a gap between the Zr and the U-7Mo foil. Nb and Ti exhibited behavior similar to that of Zr.

Hardness values were measured for the fuel, cladding, and interaction phases for HIPed samples. For the fuel and cladding, measurements were made before and after HIPing. The initial Vickers hardness values before HIPing at 580°C for 3 hours for a 6061 T6 Al alloy and U-10Mo alloy were 107 ± 2 and 263 ± 4 , respectively. After HIPing, the 6061 Al alloy and the U-10Mo alloy registered Vickers hardness values of 47 ± 1 , and 288 ± 4 , respectively. The Vickers hardness values of the two uranium-aluminide layers that formed during HIPing were also measured. The $(U,Mo)Al_3$ phase had a value of 617 ± 3 , and the $(U,Mo)_{0.9}Al_4$ had a value of 653 ± 32 . Based on these results, it can be seen that after HIPing the 6061 Al alloy cladding is softer, while there is little change in the hardness of the U-10Mo alloy. The uranium-aluminide layers that are present after HIPing are much harder than the U-10Mo and 6061 Al cladding.

2.2 Friction Stir Welding

The strength of the bond at the foil/cladding interface is of particular interest in FSW samples. To investigate this parameter, pull tests of friction stir weld (FSW) monolithic fuel plate specimens were conducted with samples fabricated using two different materials as the tool face. A description of the pull test method and procedure may be found in [4], while a description of the FSW process may be found in [5]. The two tool facing materials are defined as Alloy A and Alloy B. In particular, Alloy A has a thermal conductivity of x , while Alloy B has a thermal conductivity of $2.5x$. Thermal conductivity is an important parameter in the FSWing of monolithic fuel plates since increased heat removal away from the weld surface allows higher loads to be applied, resulting in enhanced bonding. For example, if a low thermal conductivity material is used as the tool face, less heat will be removed from the weld face, and in order to avoid a corresponding increase in process temperature and void formation, the applied load must be decreased. Diffusion of atoms across the fuel-cladding interface that results in adequate bonding is driven by two mechanisms: 1) temperature to drive the kinetic reaction and 2) applied load to keep the interface in intimate contact with one another allowing diffusion to occur. Applied load is especially important with the FSW process, since the aluminum cladding is in a plastic state and stirs on top of the fuel foil. Lower applied loads do not result in the intimate contact across the interface therefore limiting the diffusion of atoms, even if the temperature is sufficiently high. This hypothesis is shown graphically in Figure 5, where stress-time plots for Alloy A and Alloy B are provided. Analysis of Figure 5 shows that Alloy B, the higher thermally conductive material, has superior bond strength (greater than 7 times) than that of Alloy A. Reasoning behind this observation is Alloy A, as discussed above, produces mainly a mechanical bond, while Alloy B produces a diffusion enhanced bond, similar to observations made for the HIPing process. It is important to note that the pull test was terminated for the Alloy B specimen not because the foil-clad interface failed, but because the epoxy used to bond the test specimen to the aluminum platens for pull testing failed.

4. Conclusions

Based on characterization and testing described above for monolithic fuel plates fabricated by FSW and HIP, the following conclusions can be drawn:

1. The amount of interaction that will occur during HIPing of monolithic fuel plates will depend on the Mo content of the fuel foil, the temperature and time that is used, and whether or not a diffusion barrier is applied.
2. The hardness of the 6061 Al alloy cladding will decrease during HIPing, while the U-10Mo will maintain about the same hardness. Any uranium-aluminide layers that develop will be relatively hard.

3. Based on pull tests performed on FSW samples, there is a dependence of the bond strength of the fuel plate on the type of FSW tool that is used to fabricate the plate.

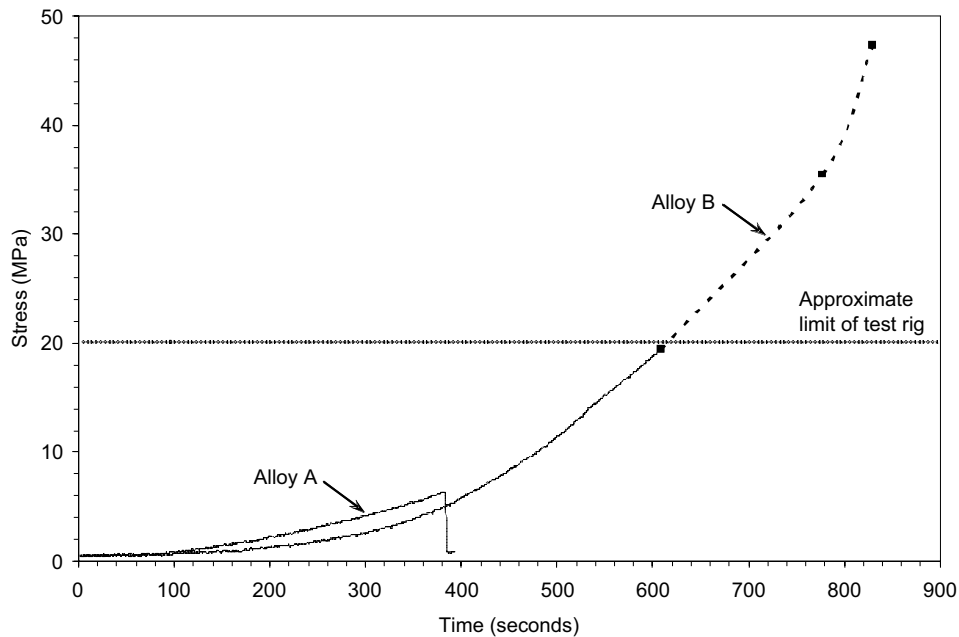


Figure 5. Stress-time plots obtained from pull test specimens fabricated employing FSW tool face materials Alloy A and Alloy B.

Acknowledgments

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References

- [1] C.R. Clark, J.F. Jue, G.A. Moore, N.P. Hallinan, B. H. Park, and D.E. Burkes, 26th International Meeting of Reduced Enrichment for Research and Test Reactors (RERTR), Cape Town, South Africa, October 29-November 2, 2006.
- [2] D.D. Keiser, Argonne National Laboratory Report, ANL-05/14 (July, 2005).
- [3] T. B. Massalski, Ed., Binary Alloy Phase Diagrams, Vol. III, ASM International, Materials Park, OH (1990).
- [4] D. E. Burkes, D. D. Keiser, D. M. Wachs, J. S. Larson and M. D. Chapple, "Characterization of Monolithic Fuel Foil Properties and Bond Strength," these proceedings (2007).
- [5] C. R. Clark, et al., This meeting.