

INTERDIFFUSION STUDIES ON HOT ROLLED U-10Mo/AA1050

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ABSTRACT

The U-Mo alloys are investigated with the goal of becoming nuclear material to fabricate high-density fuel elements for high performance research reactors. This enrichment level suggests that the U-Mo alloys should be between 6 to 10wt%, which can give up to 9gU/cm³ as fuel density. Nevertheless, the U-Mo alloys are very reactive with Al. Interdiffusion reaction products are formed since nuclear fission promotes chemical interaction layer during operation, leading to potential structural failure. Present studies were made with treated hot rolled diffusion couples of U-10Mo inserted in Al (AA1050). The U-10Mo/AA1050 pairs were treated in two temperature (150°C and 550°C) with three soaking times (5h, 40h and 80h). From microstructure analyses, rapid diffusion of Al happened inside U-10Mo in the first heating at 540°C during 15 min, reaching 8 at%Al in a range of 200 µm towards U-10Mo. Longer time (5, 40, 80h) at 550°C maintain this level of Al-content up to 1000 µm inside U-10Mo. A minor depth (~1 µm) near the interdiffusion contact had higher Al-content, but not sufficient to form identifiable (U,Mo)Al_x structures. Probably, residual elements reduced drastically the interdiffusion phenomena between U-10Mo and AA1050, maybe due to silicon presence.

Key Words: U-Mo, nuclear fuel, research reactors, interaction layer, Al/U-Mo interdiffusion pair

INTRODUCTION

The need for conversion of HEU research reactors to LEU fronts opened goals to seek alternative fuel elements of high density. The developing of a denser core in uranium leads to higher intensity in the neutron flux and smaller amounts of spent fuel to be stored in repositories. The U-Mo alloy has been investigated with the purpose to be nuclear material for making high-density fuel elements for research reac-

tors of high performance. This alloy could have high density in fuel core up to around 9 gU/cm^3 ^{(1) (2) (3) (4)}.

During fuel plates fabrication, the U-Mo alloy would employ the technology for the current LEU fuel geometry ⁽⁵⁾. U-Mo alloy is very reactive in the presence of aluminum in thermal cycling process. The reaction products are undesirable because they generate a low conductivity IL layer by nuclear fission, leading to potential structural failure ^{(3) (6) (7)}. The thermal experiments are normally carried out below γ -phase temperature formation to simulate the interdiffusion and phases formation in U-Mo-Al ⁽⁸⁾. The more common observed phases have been $(\text{U,Mo})\text{Al}_2$, $(\text{U,Mo})\text{Al}_3$, $(\text{U,Mo})\text{Al}_4$ ⁽⁶⁾. Ryu et al ⁽⁹⁾ simulated heat-treated growth-layer interaction in U-10Mo/Al fuel dispersion with treatments of 550°C during 40h. It was found 3 levels of interaction layer. The more internal layer had a depth of approximately $500 \mu\text{m}$, displaying the UAl_3 (exper. 77.5 at%Al) as the most structured one ($>400 \mu\text{m}$) towards the center of U-10Mo alloy. Near the contact region, there were 2 other layers ($\sim 50 \mu\text{m}$ each), having 81.6 at%Al (UAl_4) and 88.3 at%Al respectively.

Mirandou et al. ⁽¹⁰⁾ studied thermal treatments at 580°C in couples of U-7Mo/Al and found the formation of $(\text{U,Mo})\text{Al}_3$, $(\text{U, Mo})\text{Al}_4$. A very thin layer near the interface was found to be $\text{Al}_{20}\text{UMo}_2$. Mirandou et al. ^{(11) (10)}, studying pairs U-7Mo/Al-6061 and U-7Mo/Al-A356 at 550°C and 340°C , confirmed the presence of Si-phases (U_3Si_5). The interaction layer depth in this work had just few micrometers accounted to a probable participation with Si diffusion. Al-Si stable alloys were formed and hindered the evolution of aluminum interdiffusion into the U-Mo structure. In the present study, U-10Mo alloy was chosen for presenting a more stable γ -phase, since α -phase formation is very little for the Mo-content ⁽³⁾. The use of aluminum alloy AA1050 was elected since it was a regular alloy to fabricate MTR fuel plates and had less latent contaminants than AA6061. In this way, the interdiffusion pair was made



Figure 1 – Uranium ingot, Molybdenum cylinders and Zirconia crucible.



Figure 2 – U-10Mo alloy

by encapsulation and hot rolling U-10Mo/AA1050, at process temperature of 540°C. In this context, it is feasible to check how far the chemical interaction would happen in practical fuel fabrication.

EXPERIMENTAL

Natural uranium metal produced by IPEN's magnesiothermic process⁽¹²⁾ was used to produce U-10wt%Mo. The uranium ingot, with the composition in Table 1, was pickled with nitric acid (65%). Stoichiometric amount of Mo was added as linders (3.175 x 3.175 mm) produced Alpha Aesar (purity 99.95%). The mixture (U + Mo) was loaded in a zirconia crucible and the melting was performed in an induction furnace (ELATEC 15KW). The set of melting is shown in Figure 1. Before induction, cycles of vacuum and argon purging were made. Finally, the melting was made under 2.0×10^2 mbar (argon atmosphere). A U-10Mo ingot was produced with density of 16.80 g/cm^3 , as shown in Figure 2. A homogenization heat treatment 1000°C during 72h was made in the U-10Mo alloy to ensure better compositional variation in metastable γ -phase in U-10Mo. This treatment was made inside a sealed tort of SS310 stainless steel with continuous 1 L/min flow of argon at 2.45

Table 1 - Raw Materials to prepare the inter-diffusion pairs of U-Mo/AA1050

	Uranium	U-10Mo	AA1050
Elem.	$\mu\text{g g}^{-1}$	$\mu\text{g g}^{-1}$	g g^{-1}
Al	110 ± 3	$90,5 \pm 2,7$	$99,4 \pm 0,1$
B	< 0,4	< 0,4	
Ba	< 0,2	$1,34 \pm 0,16$	
C	0.11%	0.09%	
Ca	$6,0 \pm 1,4$	$55,3 \pm 5,6$	
Cd	< 0,10	$3,75 \pm 0,01$	
Co	$0,60 \pm 0,02$	$3,71 \pm 0,02$	
Cr	$17,2 \pm 0,5$	$43,5 \pm 5,3$	
Cu	$20,3 \pm 0,6$	$79,2 \pm 0,2$	$0,033 \pm 0,003$
Fe	423 ± 17	579 ± 9	$0,27 \pm 0,03$
Ga			< 0,01
Li	< 0,1	< 0,1	
Mg	$5,17 \pm 0,15$	$11,5 \pm 1,6$	< 0,01
Mn	196 ± 5	228 ± 7	< 0,01
Mo	< 3,0	$9,74 \text{ g g}^{-1}$	
Ni	$73,0 \pm 2,2$	$75,8 \pm 1,4$	< 0,01
Pb	< 6,0	< 6,0	
S			$0,031 \pm 0,002$
Si	$36,4 \pm 1,9$	500 ± 10	$0,20 \pm 0,07$
Sn	$11,1 \pm 2,5$	216 ± 18	
Ti			< 0,01
U	Balance	Balance	
V	$0,80 \pm 0,21$	$10,1 \pm 0,2$	
Zn	$1,64 \pm 0,28$	<1,0	< 0,01

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Bar. After annealing, the retort was removed from the oven and kept until reaching room temperature. To prepare the diffusion pairs of U-10Mo/AA1050, the samples U-10Mo were cut in tablet format using diamond wheel cutter, followed by metallo-

graphic preparation of interfaces for contact with the aluminum in the diffusion couples. The tablets were polished by diamond paste of 3 μm .

The sets containing the tablets were welded. The set was then heated at 540°C during 15 minutes before being hot rolled, then hot rolled by a single pass of 38% reduction. The final thickness of the plate was 7.35 ± 0.01 mm. The samples were identified and cut by guillotine. X-ray images were taken to guide the pairs positioning. The hot rolling fabrication had no losses. The SEM in Figure 6 (a) shows how perfect was the interdiffusion line formed. The heat treatment of diffusion couples was made in a resistance furnace with no atmosphere conditioning. These treatments are listed in Table 2. The cooling was made in room conditions.

The microstructure of the samples were observed and analyzed with optical microscopy (OM) and scanning electron microscopy (SEM JXA system Jedle 6400) with (EDS). The XRD was made by Rigato Multiflex diffractometer ($\text{CuK}\alpha$ 1.54056 Å). The raw materials were chemically characterized by infrared techniques, ICP-OES, X-ray fluorescence spectrometry and gravimetry.

Table 2 - Sample heat treatments to simulate IL formation

Samples Id	Time (min)	Temperature (°C)
PL (1A)	4800	550
PL (1B)	2400	550
PL (2A)	300	550
PL (2B)	2400	150
PL (3)	15	540

RESULTS AND DISCUSSION

The chemical analyses of the uranium and the aluminum alloy AA1050 brought many constituents to the system of interdiffusion analysis. Silicon impurity is the major concern since the literature cogitates of its hindrance for interdiffusion between Al and U-Mo pairs. Microstructures of U-10Mo alloy prior to heat treatment are shown in Figure 3(a) and (b). In these micrographs, there are punctual regions rich in α -phase. As noted in SEM/EDS, these regions have higher content of U and absence of Mo.

This rich region of α -phase probably was found to be associated with carbides and oxides of uranium.

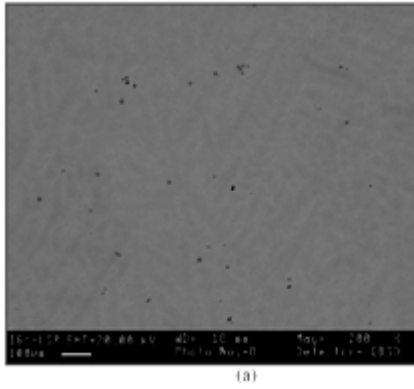


Figure 3- (a) U-10Mo as

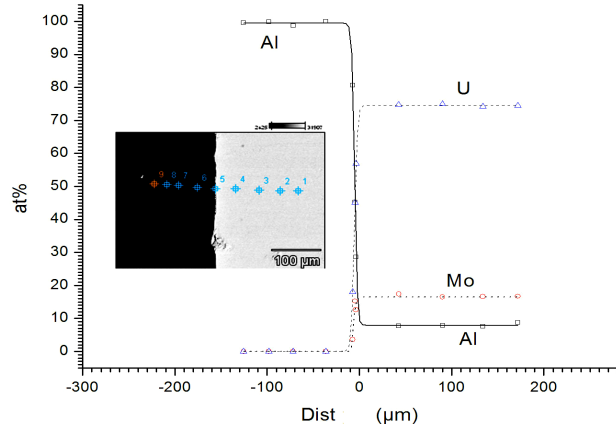


Figure 5 – Interdiffusion curves resulting after 15min soaking at 540°C

The surrounding material is γ -

phase (U, Mo) (light area). This phase, as seen in Figure 3(a), by means of SEM/EDS analyses, revealed a dendritic formation caused by the peritectic solidification of γ -phase. The difference in composition of (U, Mo), estimated qualitatively by EDS was approximately 1 at% U inside this structure. Trying to homogenize this phase, it was performed a heat treatment at 1000°C during 72h. The XRD showed a

better definition for the treated U-10Mo peaks, as shown in Figure 4.

As could be seen, the main diffraction peaks became narrower and moved to the right revealing a better defined interplanar structure. It is reasonable to say that there was an atomic rearrangement in U-10Mo structure reducing crystalline imperfections and vacancies. Seong et al. (13) suggested, by means of neutron

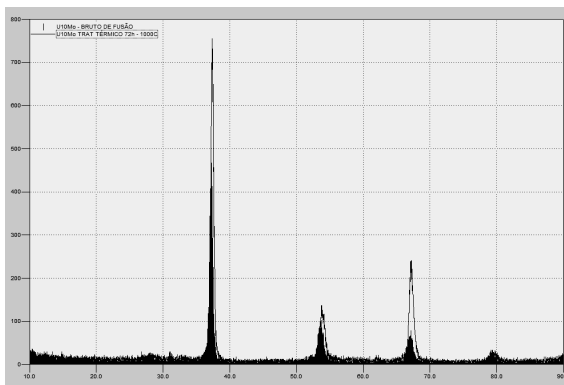


Figure 4 – Xray diffraction pattern showing the as cast (black shaded) U-10Mo and the treated one at 1000°C during 72h (line)

diffraction, that the formed phase in U-10Mo is U_3Mo with structural characteristics of the space group $P-4m2$ (115 tetragonal: $a = b = 4.8305 \text{ \AA}$, $c = 3.418 \text{ \AA}$).

The analysis of heat treated samples allowed preliminary evaluation of the interdiffusion of Al in the area of U-10Mo alloy. The resulting structure revealed a layer of chemical interaction between the constituents AA1050 and $\gamma(U, Mo)$.

The heating time to reach rolling temperature of the samples showed to be very important. Heating of the set during 15 min at 540°C was sufficient to promote a rapid evolution in aluminum diffusion inside U-10Mo, as shown in Figure 5. In this short soaking time, the aluminum diffusion towards the alloy reached a fixed amount of ~8 at% Al in more than 170 μm inside U-10Mo region. Probably, Al easily forms a solid solution inside the U-Mo, occupying structure sites very swiftly, without depending on major structure changes. By EDS analysis, it was detected that the content of Al, with this tiny soaking time,

reached 2at%Al at sample center (~1000 μm from interface). This hot rolled sample preparation simulated the first reduction pass in hot rolling of a MTR fuel plate. A complete set of rolling passes for MTR plate would require 9 passes of hot rolling that would be equivalent to around 150 min of interdiffusion phenomena at operation temperature (480-540°C).

The heating treatments (150°C and 550°C) and at higher time (5h, 40h and 80h) in this study did not reproduced a similar continuous raise of Al contents as would be forecast. With longer periods of soaking, it was observed that the level of aluminum did not surpass the level of 8 at% within minutes of treatment. The apparent change was the increasing of this fixed Al-content thickness towards the sample center. The only real difference in

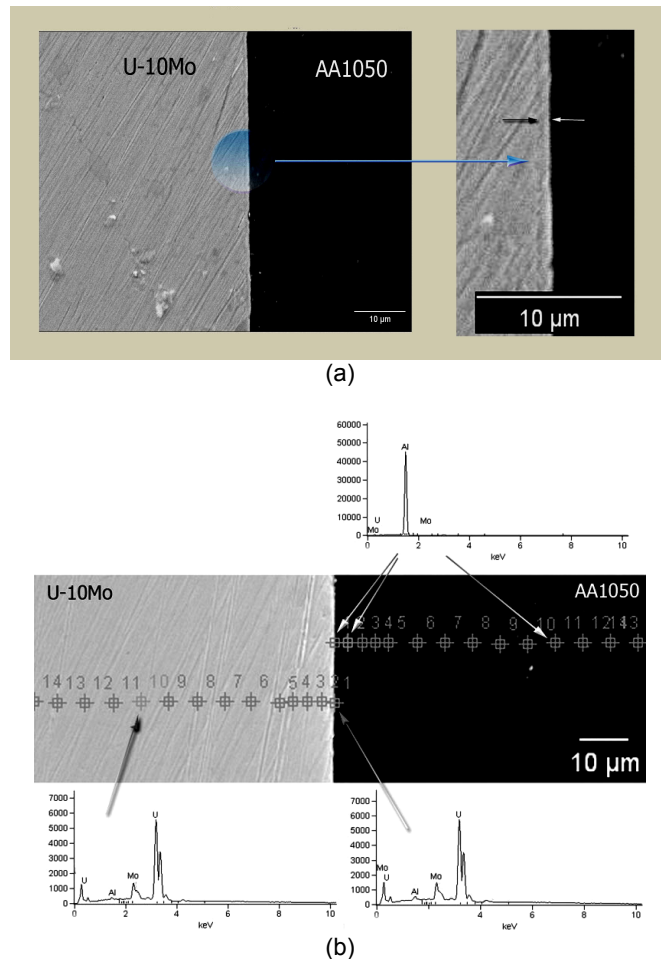


Figure 6 – Interdiffusion pair U-10Mo/AA1050 after heat treatment of 80h at 550°C. (a) General view and magnification at right side showing the interaction layer; (b) EDS analysis of the same sample.

the only real difference in

aluminum concentration occurred very near to the interdiffusion interface that separates the phases.

After 80 hours soaking, the interdiffusion sample at 550°C, as could be seen in Figure 6, showed a very thin region (~1 µm), where Al reached an amount of 11 at% in average by means of qualitative EDS measurements. This amount is not far beyond the 8 at%Al found just ahead of it in the same sample, which stayed constant until the sample center. Comparing this data with Ryu ⁽⁹⁾, it is possible to say that the present study results did not show an intense Al-interdiffusion inside U-10Mo, as expected. A probable explanation to this is the interference of another solid solution element. It might be accounted that the silicon presence, could be the cause of this blockage. This confirms the results of Mirandou et al. ⁽¹¹⁾ in relation to the influence of silicon presence.

In addition, no proper U-Al or (U,Mo)-Al could be perceived, since the formation was very insipient for the available equipment to the researchers. There are still studies being conducted in this context. It is pending more EDS evaluations and microprobe analysis that could elucidate better if any phase is predominant in the region near the interface.

CONCLUSIONS

The produced alloy was basically U-10Mo γ (U, Mo) with less than 1 vol% of α -phase. The structure changed slightly after heat treatment 1000°C during 72h.

The technique of assembling interdiffusion pair by hot rolling method produced good and regular interface for interdiffusion studies. It also simulated adequately the 1st hot rolling pass in production of MTR fuel using U-10Mo with AA1050.

During sample hot rolling operation, within 15 minutes of soaking time at 540°C, there was rapid Al-diffusion inside U-10Mo structure, sufficient to reach steadily 8 at% Al until 200 µm from interdiffusion interface and 2 at% Al at 1000 µm. Longer soaking time of interdiffusion pairs, up to 80h at 550°C revealed some composition variation of U, Mo, Al in approximately 1 µm region close to interface plane. No proper phases of U-Mo/Al were identified. Probably happened only Al solid solution happened inside U-10Mo with structure change. This experimentation showed that interaction layer between U-10Mo/AA1050 is highly prevented and limited probably due to silicon presence in AA1050.

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