Microstructural development from interdiffusion and reaction between U–Mo and AA6061 alloys annealed at 600°C and 550°C

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HIGHLIGHTS

• Diffusion couples of U–7Mo, U–10Mo, and U–12Mo vs. AA6061 were analyzed by SEM with XEDS.
• The couples were annealed at 600°C for 24 h and at 550°C for 1, 5, and 20 h.
• The interaction regions were more complex than those in diffusion couples of U–Mo vs. high purity Al and Al–Si alloys.
• Analysis showed that the alloying additions of the AA6061 were present in the interaction regions.
• Phase development was significantly influenced by the alloying additions of the AA6061.

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ABSTRACT

The U.S. Material Management and Minimization Reactor Conversion Program is developing low enrichment fuel systems encased in Al-alloy for use in research and test reactors. Monolithic fuel plates have local regions where the U–Mo fuel plate may come into contact with the Al-alloy 6061 (AA6061) cladding. This results in the development of interdiffusion zones with complex microstructures with multiple phases. In this study, the microstructural development of diffusion couples, U–7 wt%Mo, U–10 wt%Mo, and U–12 wt%Mo vs. AA6061, annealed at 600°C for 24 h and at 550°C for 1, 5, and 20 h, were analyzed by scanning electron microscopy with x-ray energy dispersive spectroscopy. The microstructural development and kinetics were compared to diffusion couples U–Mo vs. high purity Al and binary Al–Si alloys. The diffusion couples developed complex interaction regions where phase development was influenced by the alloying additions of the AA6061.

1. Introduction

Diffusional interactions in the U–Mo–Al and U–Mo–Al–Si systems have been extensively researched to develop an understanding of the phase equilibria, kinetics, and irradiation behavior [1,2]. The studies have been carried out to support the U.S. Material Management and Minimization Reactor Conversion Program, originally called the Reduced Enrichment for Research and Test Reactors (RERTR) program, for the development of low-enriched U–Mo fuels encased in Al-alloys [3–6].

In U–Mo fuel alloys, the addition of Mo stabilizes the high temperature (bcc) γ-U phase, Mo has high solubility in γ-U that allows for fuel compositional customization, and the alloy satisfies the fissile-U densities required to maintain fuel performance. Extensive studies and characterization of U–Mo alloys have been carried out to develop an understanding of the phase equilibria [7–29] and thermodynamics [30–34] of this alloy system. The γ-U can be maintained in a metastable state upon quenching of the alloy, where the stability of the phase depends on the Mo content.
Fuel system designs, whether dispersion or monolithic type, place Al-alloy 6061 (AA6061) cladding in contact with the interaction regions of diffusion couples containing U-6061. Mirandou observed the presence of the U3Si phase in diffusion couples containing AA6061 [43] and Al-alloy A356 [66]. The study with the AA6061 [43] alloy observed the dissolution of Mg2Si phase precipitates in the AA6061 at 340 °C, but did not identify Mg within the interaction region. Park [65] considered the effects of Zr additions to the U–Mo and Si additions to the Al in diffusion couples and found Zr and Si in low and high concentrations, respectively, within the interaction region. In a separate study of irradiated U–Mo dispersions in AA6061, Keiser observed low concentrations of Mg within the interaction region [56]. Varela studied Zr and Pt additions to the U–Mo in contact with Al and Al-alloy A356, and found the U3Si2 and Zr2Al3 phases [66] in the interaction regions, indicating that Zr can affect phase development.

Ewh [68] carried out a study on the effect of Nb and Zr additions to γ-U–Mo alloy in diffusion couples of U–Mo–X (X= Nb or Zr) vs. high purity Al, and determined that the element additions did not appear to have an effect on the growth rate of the interaction region in the diffusion couples. Ewh [68] determined that the UAl3 phase dominated the microstructure and that, although not identified, other phases were present within the interaction region. These phases may have included Nb and/or Zr. A recent study by Allenou [69], using micro-x-ray diffraction, considered the effect of Cr, Ti or Zr additions in the γ-U–Mo alloy, and determined that the typical phase constituents within layered microstructures of the U–Mo vs. Al system were not significantly modified. The phase fractions within the interaction region layers were modified. Measurement of the lattice parameters of the relevant phases showed changes indicating the possibility of other phases, such as UTi2Al3 and/or UCr2Al3, or element solubility into the existing phases, i.e., UMo2Al20. Allenou [69] also found that Ti and Zr had a more pronounced effect than Cr, and the thickness of the overall interaction regions was generally increased.

In this study, emphasis is given to the effect of the minor element additions to the AA6061. The phase constituents and the growth of the interaction layers that develop between U–Mo and AA6061 were examined using solid-to-solid diffusion couples, U–7wt%Mo, U–10 wt%Mo, and U–12 wt%Mo vs. AA6061. The couples were annealed at 600 °C for 24 h to develop large interaction regions and avoid decomposition of the γ-U phase. A second set of diffusion couples was annealed at 550 °C for 1, 5, and 20 h to observe the behavior of the interaction region as a function of time. Decomposition of the γ-U phase was observed in diffusion couples of U–7Mo vs. AA6061 annealed at 550 °C. The phase constituents and the thickness of the interaction regions were analyzed via scanning electron microscopy (SEM) and x-ray energy dispersive spectroscopy (XEDS).

2. Experimental details

Metallographical preparation and assembly of diffusion couples were carried out under an Ar atmosphere inside of a glove box to minimize oxidation of the alloys. AA6061 and depleted uranium (DU) alloys consisting of solid solution γ-phase with U–7wt%Mo, U–10 wt%Mo, and U–12 wt%Mo were employed for the experiments. Hereafter, these alloys are referred to as U–7Mo, U–10Mo, and U–12Mo.

The U–7Mo, U–10Mo, and U–12Mo alloys were cast using high-purity DU and Mo via arc melting. They were melted-and-flipped three times to ensure homogeneity and were then drop-cast to form rods with ½ inch (6.35 mm) diameter. The as-cast rods were then homogenized in an Ar atmosphere at 950 °C for 96 h. The U–Mo alloys were water-quenched in ice water after

Pfeil [7], Saller et al. [8–10], Ivanov et al. [14,19], Carrera et al. [11], and Dwight et al. [12] detailed the γ-U → (z-U + δ-U0Mo) decomposition that takes place below 573 °C. Howlett et al. [35], Repas et al. [36], and Goldstein et al. [37] developed Time-Temperature-Transformation (TTT) diagrams for U–Mo alloys ranging from 2.5 to 14 wt% Mo that detail the decomposition process.

The program is developing fuel systems where a γ-U0Mo fuel may come into contact with the Al-alloy 6061 (AA6061) cladding. Fuel system designs, whether dispersion or monolithic type, place γ-U–Mo fuel between AA6061 plates. For the case of dispersion fuels, γ-U–Mo particles are present at the edge of the dispersion where they can contact the AA6061. Diffusion barriers can be included to separate the γ-U–Mo from the AA6061. In these cases, regions where the barrier is not present or may fail, where the U–Mo will then likely contact the AA6061, must be considered.

U–Mo alloys in contact with Al undergo complex diffusional interactions that can produce various phases with undesirable growth kinetics [1,6–50] and irradiation behavior [51–61]. In prior studies by the authors [12,38,44,62], U–Mo alloys, high purity Al [99.999%] and Al–Si alloys were used to assemble diffusion couples to investigate the fundamental interactions that take place in the ternary U–Mo–Al and quaternary U–Mo–Al–Si systems. The interaction regions were determined to consist of a mixture of the UAl3, UAl4, U3Mo4Al12, and U2Mo2Al20 phases. The results were in agreement with the phase development observed in cast U–Mo–Al alloys [63]. When Si was introduced into the Al in the diffusion couples, the UAl3 phase demonstrated solubility for Si and Mo, and the development and/or growth of the UAl4, U3Mo4Al12 phases were suppressed [2]. The previous study by the authors with U–Mo vs. Al–Si alloys determined that the interaction regions were composed of multi-phase regions where all the phases found contained high concentrations of Al. The study observed phase regions with high Si content near the U–Mo alloys. However, it did not identify any phases containing U–Mo–Si compositions. Iltis [64] observed the UMo2Al20, U[Al(Si)3], U3Si3, and a W3Si type phase that was deduced to be a U3Mo(MoSi3–x)Si2 phase in the interaction regions of diffusion couples containing U–7Mo vs. aluminum alloy 4343.

In typical fuel systems, AA6061 is used, in lieu of pure Al, to encase the U–Mo fuel matrix to provide structural stability and to isolate the fuel. Interactions between the U–Mo and the AA6061 take place at high temperature during fuel plate manufacturing and in reactor service accelerated by irradiation. Fuel plate systems that employ diffusion barrier layers have regions where the barriers may not be present. The nominal composition of the AA6061, in wt%, is Al–1.0Mo–0.6Si–0.7Fe–0.25Zn–0.2Cu–0.2Cr–0.15Mn–0.15Ti.

Based on Gibb’s phase rule, the number of alloying components increases the available thermodynamic degrees of freedom, which allows for the possibility for the development of additional phases and more complex microstructures. As observed with the case of the introduction of Si into the U–Mo–Al system [2], additional alloying additions may also significantly modify microstructural development.

Studies using diffusion couples have employed Al-alloys in their research [41–43,53,56,65–67]. Although some of these studies have used nearly pure Al (Al-alloys A1050A, A1060 and A5), small concentrations of alloying additions may still modify the behavior of the system. With the exception of Si additions, most studies have not considered the influence that minor additions may have. A prior study by the authors [2] showed that the Si concentration in the interaction region of diffusion couples of U–Mo vs. Al–Si alloys was significantly higher than that of the terminal Al–Si alloy. This result suggests that minor element additions in AA6061 may also diffuse into the interaction regions in significant concentrations.
homogeneity in composition, phase constituents, and microstructure of the alloys were examined by Rigaku™ DMAX-B x-ray diffraction (XRD) and Hitachi™ 3500N scanning electron microscopy (SEM) equipped with x-ray energy dispersive spectroscopy (XEDS).

The AA6061 was obtained from a commercial source as AA6061-T6, which is in a solution annealed and artificially aged condition. AA6061 has a solutioning temperature of 529 °C. During the diffusion annealing temperatures used in this study, the alloy was expected to solubilize. Detailed characterization of the AA6061 was not carried out in this study.

U–Mo and AA6061 alloy rods were sectioned into discs 6.35 mm in diameter and approximately 3 mm in thickness. The faces of the U–Mo and AA6061 discs were metallographically polished, using 1 μm diamond paste as the final step. The AA6061 discs were then cleaned with concentrated HNO3 to dissolve remaining traces of oxide scale. Significant care was taken to prevent oxidation of the alloy disks during assembly. The prepared surfaces were then placed in contact with each other and held together by two clamping disks with stainless steel rods to form a jig. Jig assemblies were encapsulated in quartz capsules and sealed under an Ar atmosphere after repeated vacuum (10-4 torr) and H2 purges. Ta foil was placed inside the capsules prior to sealing to serve as an oxygen getter. All couples were annealed using a Lindberg/Blue™ three-zone tube furnace. Diffusion couples of U–7Mo, U–10Mo, and U–12Mo vs. AA6061 were annealed at 600 °C for 24 h in order to develop large interdiffusion zones and to avoid decomposition of the α-U phase [7–29,35–37]. A second set of diffusion couples was annealed at 550 °C for 1, 5, and 20 h to observe the behavior of the interaction region as a function of time and any potential coupling with the decomposition of the γ-U phase. After annealing, the diffusion couples were quenched by breaking the quartz capsule in ice water. Each diffusion couple was then mounted in epoxy, cross-sectioned, and polished for microstructural examination, where the final step used 1 μm diamond paste.

For each diffusion couple, SEM analysis was carried out using the same Hitachi™ 3500N SEM mentioned above to examine the quality of the diffusion bond and the thickness of the interaction layer. Selected regions within the interdiffusion zone were examined thoroughly to document the presence of fine-scaled microstructures. The interdiffusion zone thickness for each couple was determined by measuring the area of the interdiffusion zone in the backscatter electron (BSE) micrographs and then dividing the area by the interdiffusion zone’s width. The growth constant, K, defined by the thickness, T = Ke/t, where t is the anneal time, was determined for each diffusion couple based on the assumption of parabolic growth rate. Note that the reported growth constants in this study are used strictly as an approximation so that the growth rates can be quantified. Compositional analysis of discrete phase regions was carried out by semi-quantitative (i.e., standardless) XEDS via SEM. The concentration profiles across the interdiffusion zone were determined by electron probe microanalysis using high purity standards of various metals and UO2.

3. Results

3.1. Diffusion couples annealed at 600 °C for 24 h

Fig. 1 shows BSE micrographs of the interaction regions. The gray contrast regions between the AA6061 (top) and the U–Mo (bottom) developed in each diffusion couple annealed at 600 °C for 24 h. These micrographs demonstrate that each diffusion couple achieved sufficient bonding throughout the width of the diffusion couples. The interaction regions of the couples with terminal alloys U–7Mo, U–10Mo, and U–12Mo developed averaged thicknesses of 700 μm, 659 μm, and 694 μm, respectively. Based on the assumption of diffusion-controlled parabolic growth rate, the growth constants for the diffusion couples containing U–7Mo, U–10Mo, and U–12Mo were calculated to be 2.38, 2.24, and 2.36 μm/s, respectively.

A closer examination of the interaction regions shows complex, fine-grained multiphase microstructures developed within these interaction layers. Figs. 2–4 show detailed micrographs of the interaction regions in the U–7Mo, U–10Mo, and U–12Mo vs. AA6061 diffusion couples, respectively. The micrographs show, in progressive order, the changes in microstructures in the interaction regions from the AA6061 side to the U–Mo side.

The U–12Mo vs. AA6061 diffusion couple annealed at 600 °C for 24 h was analyzed in detail by XEDS to determine the compositions of the “distinguishable” features based on the BSE contrast. The specific locations where XEDS measurements were carried out are represented by Greek letter symbols in the BSE micrographs in Fig. 5. The measured compositions are summarized in Table 1. Because of the experimental uncertainty in the XEDS, measured concentrations lower than 1% are not reported in Table 1. Careful consideration was given to interpret the data correlating the phase contrast (i.e., atomic number) and microstructures in the BSE micrographs with the measured compositions. Up to 8 different groups of compositions were observed and reported in Table 1. Some of the composition groups can be tentatively identified as a distinct phase. Despite the careful analyses, however, some of the reported values represent the average compositions of finely distributed neighboring phases. The BSE micrographs and XEDS results suggest that a larger number of phases may exist in the interaction region.

In Table 1, all of the measured composition-groups, or tentatively distinct phases, contained high concentrations of Al (>50 at.%). Composition-group α represents the bright contrast phase observed in the interaction layer near the AA6061 terminal alloy. The α was depleted of Mo, contained high concentrations of Si, and had measurable concentrations of Cu, Fe, Mg, and Zn. Composition-group β represents the mid-gray contrast phase in the interaction region near the AA6061. It is marked by low U and Mo concentrations and measurable concentrations of Fe, Mg, and Zn. Composition-group γ was similar to β, but distinctively lacked Mo and contained Cr. Composition-group δ is distinguished by a significant concentration of Fe. Composition-group ε contained significant concentrations of U and Si, some Zn, and lacked measurable concentrations of any other alloying additions from the AA6061. Composition-group η contained mainly U, Mo, and Al, with only a low concentration of Cu. Composition-group θ contained U, Si, and low concentrations of Cu, Fe, Mo, and Zn. Assuming that the low concentration elements appear in solid solutions, the composition-group ϑ has an approximate average concentration of (U,Mo)/(Al,Si)3. This average composition has been observed in other studies [2,17,18,62,70]. Composition group κ contained only Al, Mo, and U and contains the typical average composition of the interaction region in the U–Mo–Al system.

Concentration profiles were determined by EPMA for the U–7Mo vs. AA6061 diffusion couple annealed at 600 °C for 24 h. The alloying additions typically found in the AA6061 and U–Mo alloys were measured (i.e., Al, Cr, Cu, Fe, Mg, Mn, Mo, Si, Ti, U, and Zn). Fig. 6(a) shows the measured concentration profiles of Al, Mo, Si, and U, which were found in high concentrations. They were similar to those observed in U–Mo vs. pure Al diffusion couples with the exception that near the AA6061 interface with the interaction region, depletion of the Al concentration was observed. Fig. 6(b) details the components found at low concentrations. Alloying components that did not show measurable concentration profiles in the interaction region are not reported. Cu, Fe, Mg, and Si
concentrations spiked near the interface of the interaction region with the AA6061, gradually decreased towards the direction of the U–Mo alloy, and penetrated approximately 400 μm into the interaction region. The region where these elements accumulated near the interface corresponds to the region where the Al was depleted in the concentration profile and to the area shown in Fig. 2(a and b). Similar regions can be observed for the diffusion couples containing U–10Mo and U–12Mo in Figs. 3(a–b) and 4(a–d), respectively. The composition groups documented in Table 1 for the couple containing U–12Mo show a trend similar to the measured concentration profile for the couple with U–7Mo alloy, where phases in the interaction region near the interface with the AA6061 contained Cu, Fe, Mg, and Si.

Fig. 1. Backscatter electron micrographs of the (a) U–7Mo vs. AA6061, (b) U–10Mo vs. AA6061, and (c) U–12Mo vs. AA6061 diffusion couples annealed at 600 °C for 24 h.

Fig. 2. Detailed backscatter electron micrographs from the interaction region in the U–7Mo vs. AA6061 diffusion couple annealed at 600 °C for 24 h. Changes in microstructures (a through d) are documented from the AA6061 side to the U–7Mo side of the interaction region.
3.2. Diffusion couples annealed at 550 °C for 1, 5 and 20 h

Diffusion couples, U–7Mo vs. AA6061 and U–12Mo vs. AA6061, were assembled and annealed at 550 °C for 5 and 20 h. In addition, a diffusion couple, U–7Mo vs. AA6061 was annealed for 1 h at 550 °C in an attempt to observe the early interactions. The interaction regions in the diffusion couples were consistent throughout the cross-sections of the couples. Figs. 7 through 9 show low-magnification BSE micrographs from the diffusion couples annealed for 1, 5, and 20 h, respectively. Note that the interaction regions in these figures are continuous in all the diffusion couples but are not entirely discernable. The magnifications in Figs. 7–9 were maintained constant so that direct comparisons of the thickness of the interaction regions can be made from the images. Figs. 10 through 12 show detailed BSE micrographs from selected portions of the interaction regions. The micrographs show that all of the diffusion couples developed complex fine grained multi-phase interaction regions.

According to the TTT diagrams for U–Mo alloys [35–37], decomposition of the metastable γ-U phase in the U–7Mo alloy was expected to start near, but before, 1 h at 550 °C. The U–10Mo and U–12Mo alloys were not expected to decompose before 20 h at 550 °C. The annealing times were chosen so that only the diffusion couples containing U–7Mo would experience decomposition of the γ-U phase. Decomposition of the γ-U phase in U–Mo alloys typically results in the development of irregular (e.g., non-planar) interaction regions. The growth of the interaction region when the γ-U phase is in the stable or metastable state is notably different from that when the γ-U phase undergoes decomposition ($\gamma$-U $\rightarrow$ α-U + δ-U$_2$Mo). Where decomposition of the γ-U phase has commenced, the interaction region expands into the U-alloy at an increased rate by fast Al diffusion through the α-U. The interaction region then grows unpredictably, where it typically shows regions with relative low and high interaction rates, presumably as decomposition of the U-alloy proceeds. The microstructural development of the interaction region as decomposition of the $\gamma$-U phase takes place is discussed in more detail in a related publication by the authors [2]. The microstructures of the interaction regions, shown in Figs. 7 through 9 indicate that only the diffusion couples containing U–7Mo experienced decomposition of the U–Mo alloy after 1 h of diffusion annealing. The U–10Mo and U–12Mo alloys did not experience decomposition after 20 h of annealing at 550 °C, and the interaction regions for these couples developed nearly consistent thickness with planar interfaces.

Fig. 7 shows that measurable interactions between the U–7Mo and the AA6061 had taken place after annealing for 1 h Fig. 8(a) shows that the interaction region in the couple containing U–7Mo developed an irregular thickness after 5 h of annealing that is consistent with the microstructures that develop as a result of decomposition of the metastable γ-U phase in the U–Mo alloy. This couple showed regions of low and high interaction. Rapid expansion of the interaction region took place in the direction of the U-alloy. This supports that the observed microstructure of the interaction regions was a direct result of decomposition of the U–7Mo alloy and consequent Al diffusion into the U–Mo through the α-U phase.

The couples containing U–10Mo and U–12Mo, shown in Fig. 8(b and c) respectively, did not experience the decomposition of the γ-U phase. Decomposition was not expected based on the TTT-diagrams [35–37]. Moreover, in the diffusion couples in this study, decomposition of the γ-U phase would be marked by significant expansion of the interaction region into the U-alloy and the development of irregular interfaces between the interaction region and the U–Mo. Figs. 8 and 9 show that even after 20 h of annealing, except for edge effects, the interface between the interaction region and the U–Mo remained relatively planar, indicating that the γ-U phase remained stable.

The diffusion couples with U–10Mo and U–12Mo annealed for 5 h generally developed thin interaction regions with low growth rates. Localized regions of higher interaction, which had the
appearance of “bumps” in the microstructures were observed in these couples, as shown in Fig. 8(b and c), and detailed in Fig. 11(b and c). Because the U–Mo alloys had not experienced decomposition, and expansion of these bumps took place in the direction of the Al-alloy, the mechanism that led to the development of these bumps ought to be different to that observed in the U–7Mo diffusion couple annealed for the same time. The mechanism that resulted in the formation of the bumps was not identified in this study. Extensive care was taken to prevent the presence of oxides or other contaminants on the alloy surfaces during assembly of the diffusion couples. The development of these bumps is not expected to be a result of sample preparation. The authors hypothesize that the locations of the bumps may result from compositional variations in the AA6061. This hypothesis, however, remains a source for further studies.

Table 2 reports the thickness and the growth constants, based on the assumption of parabolic growth rate, of the interaction regions for each diffusion couple. For the cases where local thicker interaction regions, referred to as regions of high interaction, were observed, two values are reported. In the diffusion couples containing U–7Mo, the growth constants between the couple annealed for 1 h and that of the low interaction region in the couple annealed for 5 h were approximately the same. The growth constant in the regions of low interaction at this composition approximately doubled after annealing for 20 h, indicating a probable change in the growth mechanism. The growth rate of the regions of high interaction in the U–7Mo diffusion couple remained nearly identical after annealing for 20 h.

The diffusion couples containing U–10Mo and U–12Mo annealed for 5 h showed relatively small growth constants within the regions of low interactions, and somewhat higher growth constants in the regions of high interaction. Comparison of the couples containing U–10Mo with those containing U–12Mo shows that the growth constants for the low and high interaction regions are very similar. After annealing for 20 h, a significant increase in the growth rate of the interaction regions was observed that is clearly marked by an increase in the growth constant of approximately an order of magnitude. The mechanism that led to a change in the growth rate of these interaction regions, and that of the low growth in the U–7Mo diffusion couples, is not yet understood.

The interdiffusion zone of the U–12Mo vs. AA6061 annealed for 5 h, presented in Figs. 8(c) and 11(c), was investigated in more
Fig. 5. Backscatter electron micrographs of the U–12Mo vs. AA6061 diffusion couple annealed at 600 °C for 24 h showing the locations, based on backscatter electron contrast, where the XEDS data in Table 1 was collected.

Table 1
Composition-groups of the potential phases, developed in the U–12Mo vs. AA6061 diffusion couple annealed at 600 °C for 24 h, determined by XEDS in atomic percent. 

<table>
<thead>
<tr>
<th>Element</th>
<th>α</th>
<th>β</th>
<th>γ</th>
<th>δ</th>
<th>ε</th>
<th>η</th>
<th>θ</th>
<th>κ</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>52.1 ± 1.7</td>
<td>81.3 ± 1.3</td>
<td>83.8 ± 0.7</td>
<td>71.9 ± 0.6</td>
<td>59.7 ± 1.9</td>
<td>83.0 ± 0.9</td>
<td>62.8 ± 2.3</td>
<td>74.5 ± 0.8</td>
</tr>
<tr>
<td>Cr</td>
<td>–</td>
<td>–</td>
<td>3.6 ± 0.4</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Cu</td>
<td>1.9 ± 0.7</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>1.2 ± 0.2</td>
<td>1.1 ± 0.2</td>
<td>–</td>
</tr>
<tr>
<td>Fe</td>
<td>1.1 ± 0.4</td>
<td>1.0 ± 0.2</td>
<td>–</td>
<td>1.6 ± 0.7</td>
<td>–</td>
<td>–</td>
<td>1.1 ± 0.2</td>
<td>–</td>
</tr>
<tr>
<td>Mg</td>
<td>4.3 ± 0.9</td>
<td>4.5 ± 0.4</td>
<td>4.0 ± 0.2</td>
<td>6.0 ± 0.2</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
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<tr>
<td>Mn</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Mo</td>
<td>–</td>
<td>–</td>
<td>5.1 ± 0.2</td>
<td>–</td>
<td>–</td>
<td>7.6 ± 0.6</td>
<td>1.0 ± 0.3</td>
<td>–</td>
</tr>
<tr>
<td>Si</td>
<td>16.9 ± 1.0</td>
<td>–</td>
<td>–</td>
<td>1.3 ± 0.4</td>
<td>10.3 ± 1.6</td>
<td>–</td>
<td>7.4 ± 1.1</td>
<td>–</td>
</tr>
<tr>
<td>Ti</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>U</td>
<td>18.0 ± 1.2</td>
<td>4.1 ± 0.4</td>
<td>3.9 ± 0.2</td>
<td>5.0 ± 0.3</td>
<td>25.1 ± 0.6</td>
<td>5.4 ± 0.3</td>
<td>23.0 ± 1.7</td>
<td>15.4 ± 0.3</td>
</tr>
<tr>
<td>Zn</td>
<td>2.8 ± 1.0</td>
<td>1.1 ± 0.4</td>
<td>0.9 ± 0.3</td>
<td>1.1 ± 0.3</td>
<td>1.6 ± 0.6</td>
<td>–</td>
<td>1.4 ± 0.4</td>
<td>–</td>
</tr>
</tbody>
</table>

a Note that concentrations lower than 1 at.% are not reported.

detail through XEDS and x-ray mapping. Figs. 13 and 14 show x-ray maps from this diffusion couple detailing regions of general and locally accelerated (e.g., bumps) interactions, respectively. While the AA6061 alloying additions (e.g., Si, Mg, Ti, Cr, Fe, Cu, and Zn) were considered along with Al, U, and Mo, the intensities of these elements, with the exception of Si and Mg, were too low for reliable and repeatable observation. Selected regions 1 through 10 are labeled in the SEM micrograph shown in Figs. 13(a) and 14(a), with corresponding compositions reported in Table 3. Since the interaction region, labeled areas 2 through 4 and 7 through 9, contained...
a very-fine multi-phase microstructure, the values reported in Table 3 represent average compositions.

Figs. 13 (b–e) and 14 (b–e) show that the interaction region contained significant distributions of Al, Mo, Si, and U. A significant enrichment in Si concentration was observed near the U–12Mo alloy, as presented in Figs. 13(c) and 14(c), which correspond to the measured composition reported in Table 3 as areas 4 and 9. Mg was detected in low concentrations in the interaction region by XEDS in areas 2, 3, 7, and 8. The interaction region did not contain measurable concentrations of Mg near the U–12Mo alloy where the Si concentration was high. These results indicate that at least some of the minor element additions of the AA6061 penetrated into the interaction region, but do not appear to have penetrated into the Si enriched region near the U–12Mo alloy. The x-ray maps show that the compositions measured in Table 3 extend beyond the areas measured by XEDS. The maps also confirm that the interaction region in this diffusion couple was divided into two sub-layers of low and high Si concentrations.

4. Discussion

The diffusion couples, U–7Mo, U–10Mo, and U–12Mo vs. AA6061, annealed at 600 °C for 24 h, developed large interaction regions with complex microstructures that contained various precipitates with significant compositional differences. The large differences in composition suggest several phases developed within the interaction regions influenced by the alloying additions in AA6061. The results of this study are discussed herein to compare to a previous study that examined diffusion couples, U–7Mo, U–10Mo, and U–12Mo vs. high purity Al [1] annealed at 600 °C for 24 h.

XEDS analysis of the U–12Mo vs. AA6061 diffusion couple, summarized in Fig. 5 and Table 1, and the concentration profile from the U–7Mo vs. AA6061 diffusion couple, shown in Fig. 6, revealed that the Cu, Fe, Mg, and Si alloying additions in the AA6061 penetrated up to 400 μm into the interaction region, particularly with a significant accumulation near the AA6061 alloy. Zn was not consistently observed in the concentration profile, but was detected by XEDS to the same depth. Cr was only detected very near the AA6061 in discrete Cr-containing precipitates. Mn and Ti were not observed in measurable concentrations.

The presence of alloying additions and their partition into various precipitate phases, particularly near the AA6061 alloy, was the major difference in microstructural constituents observed in this study compared to the study carried out with pure Al [1]. In our previous study [1], UAl₆, UMo₂Al₂₀, UAl₃, and U₆Mo₄Al₄₃ were identified by transmission electron microscopy in the diffusion couples that developed layered microstructures with two or three of these phases. When plotted onto the ternary Al–Mo–U phase diagram [63, 71, 72] the profiles from that study were found to accurately predict the phase development of the interaction region based on the diffusion path. Comparison of the U–Mo vs. AA6061 diffusion couples to those vs. pure Al indicates that the microstructures within the interaction zone near the U–Mo alloys were quite similar. In the U–Mo vs. pure Al, the interaction regions near its center and towards the U–Mo alloy contained a three-phase microstructure with the UAl₃, UMo₂Al₂₀, and U₆Mo₄Al₄₃ phases. The concentration profile in Fig. 6 confirmed that the alloying additions of the AA6061 did not penetrate in measurable concentrations into the region near U–Mo alloys. Thus, near the U–Mo alloys, the interaction contains only Al, Mo, and U. The phase development in this region should then be restricted to the ternary Al–Mo–U ternary system [63, 71, 72]. If the relevant section of concentration profile is plotted onto the ternary U–Mo–Al system, the diffusion path can be reasonably used to predict the phases that can be
Fig. 8. Large portions of the interaction regions of the (a) U–7Mo, (b) U–10Mo, and (c) U–12Mo vs. AA6061 diffusion couples annealed for 5 h.

Fig. 9. Large portions of the interaction regions of the (a) U–7Mo, (b) U–10Mo, and (c) U–12Mo vs. AA6061 diffusion couples annealed for 20 h.
present in the interaction region. Based on this analysis, the interaction regions, near the U–Mo alloys, in the U–Mo vs. AA6061 diffusion couples may be estimated to contain the binary and/or ternary phase regions observed in the ternary U–Mo–Al system (e.g., UAl₃, UMo₂Al₂₀, and U₃Mo₉Al₁₃).

The alloying additions in AA6061 appear to have increased the rates of diffusion and/or reaction at 600 °C when compared to high purity Al. Based on the assumption of parabolic growth rate, the interaction zone in the U–7Mo, U–10Mo, and U–12Mo vs. pure Al couples had growth constants of 0.90, 1.84, and 1.20 μm/s¹⁄₂, respectively [1]. The couples with the same U–Mo alloys vs. AA6061 examined in this study had higher growth rates of 2.38, 2.24, and 2.36 μm/s¹⁄₂, respectively, at 600 °C. The mechanism associated with an increase in the rate of interdiffusion and reaction, along with detailed crystallographic characterization of the various phases due to alloying additions in AA6061 warrant further study.

Diffusion couples, U–7Mo, U–10Mo, and U–12Mo vs. AA6061, were annealed at 550 °C for 1, 5, and 20 h in order to examine the effects of γ–U phase decomposition and to compare the interaction kinetics to U–Mo vs. pure Al and U–Mo vs. Al–Si diffusion couples [2]. Decomposition of U–7Mo alloy was expected to start near, but before, approximately 1 h at 550 °C. A diffusion couple was annealed for 1 h to attempt to observe the early stages of decomposition and its effect on the development of the interaction region. After annealing for 1 h, this couple developed a consistent interaction region. The interface between the interaction region and the U–7Mo remained relatively planar. The microstructure in U–7Mo vs. AA6061 couples annealed for 5 and 20 h show that decomposition of the γ–U phase had commenced. Figs. 8(a) and 9(a) show that the interaction region had expanded into the U–7Mo alloy, presumably by fast Al diffusion into the α–U phase. This resulted in roughening of the interfaces between the interaction regions and the U–7Mo alloys, where the interfaces were no longer planar. For the U–10Mo and U–12Mo vs. AA6061 couples, decomposition of the γ–U phase was not observed. The interface between the interaction regions and the terminal alloys remained relatively planar. Some regions with localized thicker interaction zones were observed.

A previous study [2,62] was carried out with diffusion couples of U–7Mo, U–10Mo, and U–12Mo vs. high purity Al, Al–25Si, and Al–55Si alloys annealed at 1, 5, and 20 h. It reported the growth constants of the interaction regions for these couples, which are reiterated in Table 4. Comparison of the growth constants in Table 4 with those in Table 2 reveals similarities and differences in the growth rates of the interaction layers. Comparison of the U–7Mo vs. AA6061 couple with the U–7Mo vs. Al and Al–Si alloys couples, annealed for 1 h, shows that the couple with AA6061 had a growth rate smaller than the couples containing pure Al, and similar to the couples containing Al–Si alloys.

The γ–U phase in the diffusion couple of U–7Mo vs. AA6061 annealed for 5 h began decomposition. This couple developed a relatively thin interaction region, with certain areas showing higher interaction rates, as marked by thicker interaction regions.
Table 2 reports the growth constants of the U–7Mo vs. AA6061. Comparison of the U–7Mo vs. AA6061 couple with all the U–Mo vs. Al–Si couples (Table 4) shows that the thin interaction region in the U–7Mo vs. AA6061 couple had a growth rate similar to that of the couples vs. Al–Si alloys. The regions with greater thickness in this couple were associated with the decomposition of the γ-U phase in the U–7Mo alloy. Comparison of the growth constants for this couple with that of the diffusion couple of U–7Mo vs. pure Al annealed for 5 h shows that the regions of higher interaction in the U–7Mo vs. AA6061 diffusion couple grew at a comparable rate to the diffusion couple with pure Al.

The microstructure of the diffusion couple where the γ-U phase has started decomposition appears controlled by the localised Si-concentrations within the interaction region. The diffusion couples of U–7Mo vs. Al–Si alloys in our previous study [2] experienced decomposition of the γ-U phase after annealing for 5 h. Even after annealing for 20 h, the interaction regions remained small with predictable growth rates defined by their growth constants. The study deduced that the high Si concentration in the interaction regions prevented expansion into the U–Mo by diffusion through the γ-U phase. The AA6061 contains a limited concentration of Si, which does not appear to be sufficient in some regions of the interaction region to deter Al diffusion into the γ-U phase in the U–7Mo vs. AA6061 diffusion couples. The behavior of the interaction region in the U–7Mo vs. AA6061 couple resembles that of U–Mo vs. Al–Si alloys, where it remained thin, and that of U–Mo vs. pure Al, where higher interaction rates were observed.

Mirandou [43] conducted a study with U–7Mo vs. AA6061 alloys annealed at 550 °C for a total of 3 h. The interaction regions that developed in this study were separated into two types of interaction layers defined by areas where the γ-U remained stable or decomposed. Where the γ-U phase remained stable and decomposed, the interaction regions developed average thickness of 6 μm and up to 300 μm respectively. Mirandou carried out crystallographic characterization by x-ray diffraction with a conventional diffractometer and with synchrotron radiation. That study determined that the interaction layers where decomposition of the γ-U phase had started contained the UAl3 and U6Mo4Al13 phases. Where the γ-U phase remained stable, the interaction region was found to contain the U(Al, Si)3 phase. Although the growth rates of the interaction regions in Mirandou’s study, based on the assumption of parabolic growth rate, differed from those observed in the current study, the developed microstructures appear quite similar in both studies. As a result, the microstructures in the current study may be deduced to contain similar phase distributions for the U–7Mo vs. AA6061 where partial decomposition of the γ-U phase was observed.

The U–Mo alloys maintained the γ-U phase in the U–10Mo, and U–12Mo vs. AA6061 diffusion couples annealed for 5 h. Comparison of the growth rates of the interaction regions in these couples with those of U–10Mo and U–12Mo vs. Al–2Si and Al–5Si annealed at the same conditions shows that the regions of lower interaction in the couples containing AA6061 experienced lower growth rates. This indicates that the minor element additions may have reduced interaction rates further than those of diffusion couples with Al–Si alloys, and is in agreement with the study by Allenou [65] where Cr, Ti, and Zr were added to the U–Mo alloy. The regions of higher interaction rates in the U–10Mo and U–12Mo vs. AA6061 diffusion couples experienced similar growth rates to the U–10Mo and U–12Mo vs. Al–2Si and Al–5Si couples annealed for the same time. This suggests that in these regions, Si affected the growth rate, and the other additions of the AA6061 did not have a significant effect. These diffusion couples showed that after 5 h of annealing time, the growth rates of the interaction regions in diffusion couples where the U–Mo alloy had not started to decompose, remained lower than those experienced with Al–Si alloys and significantly lower than the rates in diffusion couples of U–10Mo and U–12Mo vs. pure Al annealed for the same.

After annealing for 20 h, the diffusion couples, U–7Mo, U–10Mo, and U–12Mo vs. AA6061 showed significant increases in the interaction growth rates, as reported in Table 2. Table 4, after 20 h of annealing, only reports the developed thickness of the U–12Mo vs. Al diffusion couple. Nonetheless, the growth constants for the U–7Mo, U–10Mo, and U–12Mo vs. Al diffusion couples...
were reported. Comparison of Table 2 with Table 4 shows that at 550 °C the growth rates of the couples with AA6061 were comparable to the couples with pure Al.

5. Conclusions

The alloying additions of the AA6061 introduce significant complexity into the evolution of the interaction between U–Mo and Al-alloys. Diffusion couples of U–7Mo, U–10Mo, and U–12Mo vs. AA6061 were annealed at 600 °C for 24 h and at 550 °C for 1, 5, and 20 h. At 600 °C, the diffusion couples developed large and complex interaction regions with growth rates higher than those of similar U–Mo vs. Al diffusion couples annealed at the same conditions. The developed interaction regions were significantly more

Table 2
Measured thickness of the interaction region in the diffusion couples annealed at 550 °C for 1, 5, and 20 h. Parabolic growth constants were determined under the assumption of a diffusion controlled process after 20 h of diffusion anneal.

<table>
<thead>
<tr>
<th>Diffusion couple</th>
<th>550 °C for 1 h</th>
<th>550 °C for 5 h</th>
<th>550 °C for 20 h</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness (µm)</td>
<td>(growth constant (µm/sec^0.5))</td>
<td>Thickness (µm)</td>
<td>(growth constant (µm/sec^0.5))</td>
</tr>
<tr>
<td>U–7Mo vs. Al–6061</td>
<td>9.6 ± 1.3 [0.16]</td>
<td>12.8 ± 1.8 [0.10]</td>
<td>59.7 ± 13.9 [0.22]</td>
</tr>
<tr>
<td>U–10Mo vs. Al–6061</td>
<td>N/A</td>
<td>5.1 ± 1.9 [0.04]</td>
<td>348.3 ± 114.2 [1.30]</td>
</tr>
<tr>
<td>U–12Mo vs. Al–6061</td>
<td>N/A</td>
<td>2.0 ± 0.4 [0.01]</td>
<td>12.8 ± 5.1 [0.10]</td>
</tr>
</tbody>
</table>
complex than those observed in diffusion couples of U–Mo vs. pure Al and Al–Si alloys. SEM and XEDS analysis showed that the alloying additions of the AA6061 were present in the interaction regions and led to the development of several phases that contained significant concentrations of these elements.

Analysis of diffusion couples annealed at 550 °C as a function of time showed that the growth of the interaction layer depended on the compositional distributions in the U–Mo alloys. The couples containing U–7Mo vs. AA6061 initially showed growth rates comparable to diffusion couples of U–7Mo vs. Al–Si alloys. After 5 h of annealing, the γ-U phase commenced decomposition, and the diffusion couples showed growth rates similar to diffusion couples of U–7Mo vs. pure Al. Diffusion couples with U–10Mo and U–12Mo did not experience decomposition of the γ-U phase. The couples annealed for 5 h developed thin interaction regions with limited areas showing thicker interactions. The thin and thicker interaction regions showed growth rates lower and similar, respectively, to those determined for the U–Mo vs. Al–Si diffusion couples annealed under the same conditions. After annealing for 20 h, the diffusion couples of U–10Mo and U–12Mo experienced growth rates similar to those of U–Mo vs. pure Al diffusion couples.

Table 3
Standardless semi-quantitative XEDS compositional analysis (at.%) of selected regions of the interaction layer in the U–12Mo vs. Al-6061 diffusion couple annealed at 550 °C for 5 h.

<table>
<thead>
<tr>
<th>Area</th>
<th>Element (at.%)</th>
<th>Al</th>
<th>Mg</th>
<th>Si</th>
<th>Mo</th>
<th>U</th>
<th>Other</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td></td>
<td>97.1</td>
<td>1.6</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>1.3</td>
</tr>
<tr>
<td>2</td>
<td></td>
<td>87.7</td>
<td>2.1</td>
<td>5.4</td>
<td>1.0</td>
<td>3.3</td>
<td>0.5</td>
</tr>
<tr>
<td>3</td>
<td></td>
<td>78.2</td>
<td>2.6</td>
<td>7.4</td>
<td>3.2</td>
<td>8.1</td>
<td>0.5</td>
</tr>
<tr>
<td>4</td>
<td></td>
<td>37.1</td>
<td>–</td>
<td>27.5</td>
<td>8.6</td>
<td>26.2</td>
<td>0.6</td>
</tr>
<tr>
<td>5</td>
<td></td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>24.6</td>
<td>75.4</td>
<td>–</td>
</tr>
<tr>
<td>6</td>
<td></td>
<td>98.0</td>
<td>1.4</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>0.6</td>
</tr>
<tr>
<td>7</td>
<td></td>
<td>67.8</td>
<td>2.1</td>
<td>10.4</td>
<td>4.1</td>
<td>13.6</td>
<td>2.0</td>
</tr>
<tr>
<td>8</td>
<td></td>
<td>68.7</td>
<td>2.0</td>
<td>9.7</td>
<td>4.4</td>
<td>14.2</td>
<td>1.0</td>
</tr>
<tr>
<td>9</td>
<td></td>
<td>21.7</td>
<td>–</td>
<td>32.4</td>
<td>8.9</td>
<td>34.1</td>
<td>2.1</td>
</tr>
<tr>
<td>10</td>
<td></td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>25.6</td>
<td>74.4</td>
<td>–</td>
</tr>
</tbody>
</table>

Fig. 14. (a) Secondary electron micrograph, (b) Al-, (c) Si-, (d) Mo-, and (e) U-x-ray maps of a selected region with higher interaction rates within the interaction layer of the U–12Mo vs. AA6061 diffusion couple annealed at 550 °C for 5 h.
Table 4
Developed thickness and growth constants for the U–7Mo, U–10Mo, and U–12Mo vs. Al, Al–5Si, and Al–5Si diffusion couples annealed at 550 °C for 1, 5, and 20 h [2].

<table>
<thead>
<tr>
<th>Diffusion couple</th>
<th>Growth constant (μm/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U–7Mo vs. Al</td>
<td>N/A</td>
</tr>
<tr>
<td>U–10Mo vs. Al</td>
<td>N/A</td>
</tr>
<tr>
<td>U–12Mo vs. Al</td>
<td>N/A</td>
</tr>
<tr>
<td>U–5Si vs. Al</td>
<td>N/A</td>
</tr>
<tr>
<td>U–10Mo vs. Al–5Si</td>
<td>6.4 ± 0.2</td>
</tr>
<tr>
<td>U–12Mo vs. Al–5Si</td>
<td>2.5 ± 0.5</td>
</tr>
<tr>
<td>U–10Si vs. Al–5Si</td>
<td>8.3 ± 0.2</td>
</tr>
<tr>
<td>U–12Si vs. Al–5Si</td>
<td>11.9 ± 0.5</td>
</tr>
<tr>
<td>U–10Mo vs. Al–5Si</td>
<td>6.4 ± 0.2</td>
</tr>
<tr>
<td>U–12Mo vs. Al–5Si</td>
<td>2.5 ± 0.5</td>
</tr>
<tr>
<td>U–10Si vs. Al–5Si</td>
<td>8.3 ± 0.2</td>
</tr>
<tr>
<td>U–12Si vs. Al–5Si</td>
<td>11.9 ± 0.5</td>
</tr>
</tbody>
</table>

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References


