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

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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.

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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 → (α-U + δ-U_2Mo) 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-UMo 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 [1,38–50] and irradiation behavior [51–61]. In prior studies by the authors [1,2,39,44,62], 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 UAl_3, UAl_4, $U_6Mo_4Al_{43}$, and UMo_2Al_{20} 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 UAl_3 phase demonstrated solubility for Si and Mo, and the development and/or growth of the UAl_4, $U_6Mo_4Al_{43}$ 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 UMo_2Al_{20}, $U(Al,Si)_3$, U_3Si_5 and a W_3Si_5 type phase that was deduced to be a $U_4Mo(Mo_xSi_{1-x})Si_2$ 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 barriers have regions where the barriers may not be present. The nominal composition of the AA6061, in wt%, is Al–1.0Mg–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.

In addition to the typical phases that develop in the interaction regions between U–Mo alloys and Al (UAl_3, UAl_4, $U_6Mo_4Al_{43}$, and UMo_2Al_{20}), Mirandou observed the presence of the U_3Si_5 phase in diffusion couples containing AA6061 [43] and Al-alloy A356 [66]. The study with the AA6061 [43] alloy observed the dissolution of Mg_2Si 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 U_3Si_5 and Zr_5Al_3 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 UAl_3 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 UTi_2Al_{20} and/or UCr_2Al_{20}, or element solubility into the existing phases, i.e., UMo_2Al_{20}. 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 reduced.

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 alloys 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 ¼ 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