OUT OF PILE DIFFUSION STUDIES BETWEEN U-7wt%Mo AND Al-Si ALLOYS

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ABSTRACT

New diffusion experiments were performed at 340 ºC between U-Mo and Al6061 and Al A 356 alloys. Better concentration measurements and XRD identification of crystal structures were possible. Experiments to study eventual changes of the interaction-layer due to the consumption of Si while diffusion is taking place are also presented. (U, Mo)(Al, Si)$_3$ with a concentration of approximately 55 at% of Si, is proposed to be the only component of the interaction layer. The zone, surrounding the interaction layer, where Si precipitates are dissolved, already reported at 550ºC, is also very noticeable at 340ºC.

Results for new temperatures were added to the TTT diagram for γ U-7wt%Mo. The influence of Pt as γ-stabilizer is also shown.

1. Introduction

Several failures have been observed during qualifying experiments, in dispersion miniplates or fuel elements, of U-(7wt% to 10wt%)Mo in Al matrix [1,2,3]. Although the problem has not been fully understood, investigations point at the interaction product due to interdiffusion between U-Mo and Al, as being the responsible for these failures.

In order to change the phases that form the interaction layer (IL) between U-Mo and Al, different elements have been added to Al, Si being the most promising [1,4]. Results showing positive changes in the IL have been already identified in out of pile experiments at 550ºC [4]. At this temperature the presence of Si suppressed the formation of (U,Mo)Al$_4$, being (U,Mo)(Al,Si)$_3$ the main component. Experiments to asses this effect at lower temperatures were necessary. Preliminary results at 340ºC were already reported [5], being also promising. They indicated an IL with a phase containing an important quantity of Si. At both temperatures, these phases rich in Si, are formed at expense of the dissolution of Si precipitates in the alloy in a region surrounding the IL, from now on, "dealloyed zone".

This work presents new results of out of pile interdiffusion experiments between U-7wt%Mo and two different Al-Si alloys, containing 0.6wt% and 7.1wt% of Si respectively, performed at 340 ºC. Also the first results of the experiments that are being done to evaluate possible changes on the composition of the IL due to consumption of Si are presented. Results to date are reviewed and summarized.

As it has been mentioned before, heat-treatment of out-of-pile diffusion experiments are time limited due to the decomposition of the metastable γ U-Mo phase towards the equilibrium phases αU and U$_2$Mo. This alters significantly the component phases and the growing kinetics of the IL [6]. Then, it is important to know, as complete as possible, the TTT diagram for the alloy being used. Results for new temperatures are presented. The influence of Pt as γ stabilizer is also shown in this work.

2. Out of pile interdiffusion experiments between U-7wt%Mo and Al-Si alloys at 340ºC.

Diffusion couples were fabricated by Friction Stir Welding (FSW) technique, [7, 8]. Fig. 1 is a schematic view of the experimental set up for this technique, which allows encasing a foil of U-Mo in between a cladding of Al alloy.

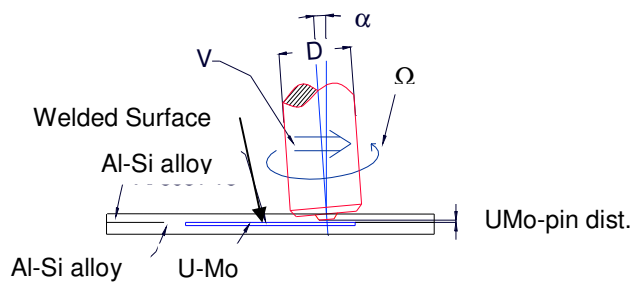


Fig.1. Experimental set up for the FSW technique

Materials employed were an U-7wt%Mo alloy homogenized in composition by a thermal treatment of 2 hours at 1000 ºC and two Al-Si alloys: Al 6061 and Al A356[1] which contain 0.6wt% and 7.1wt% of Si respectively. Thermal treatments were performed at 340 ºC. Details are summarized in Table I.

Sample	*Al-Si alloy*	*Time (days)*	*IL (μm)*	*Dealloyed zone (μm)*	*REF.*
1	A 356	49	21	31	[5]
2	A 356	23	5	33	This work
3	6061	52	2	30	[5]
4	6061	90 (still on going)			

Table I. Thermal treatments at 340ºC. Results for the thickness of the interaction layers and dealloyed zones.

Optical Microscopy (OM) revealed, for all the samples, an IL and their corresponding dealloyed zone on the Al-Si alloy. Thickness are shown in Table I. Back scattered electron images indicated no differences in composition inside the ILs at this temperature, Fig 2.

[1] It was named a AA635 in [4]

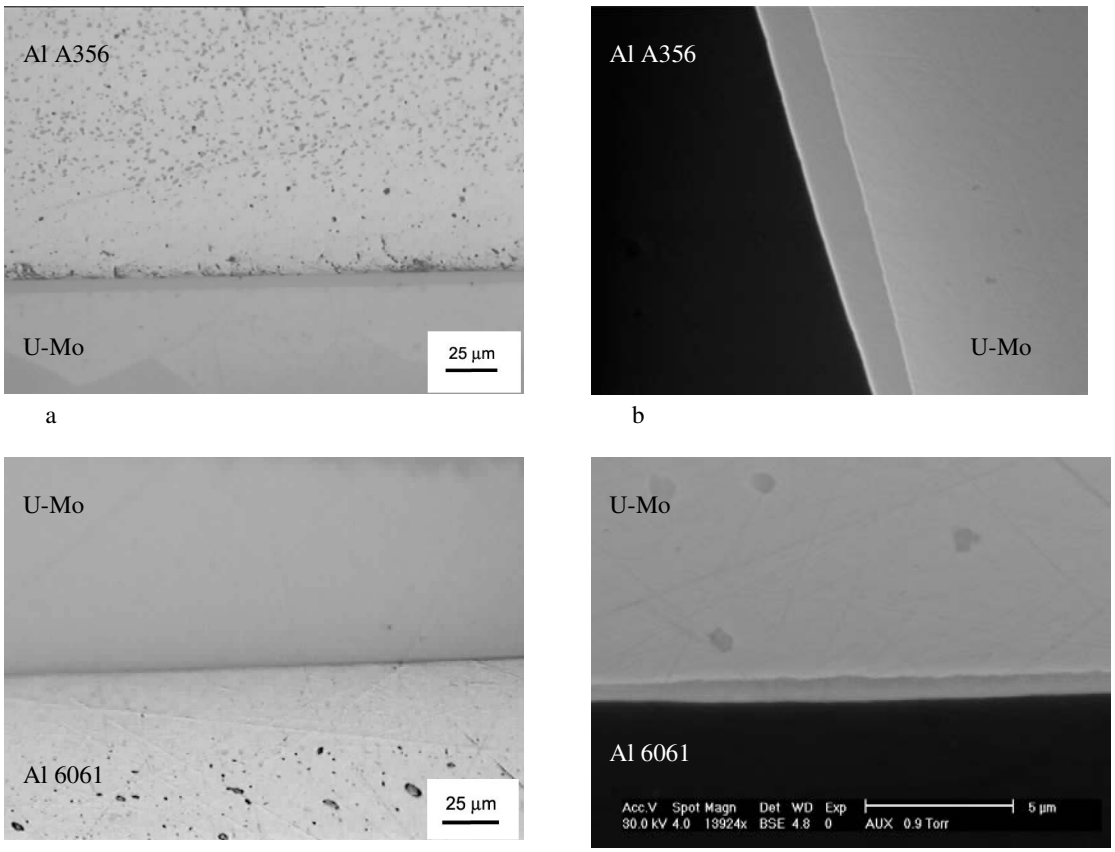


Fig 2. Diffusion experiments at 340 ºC. a) Sample 2, 23 days, OM. b) Sample 2, 23 days, Back scattered electron image. 3) Sample 3, 52 days, OM. d) Sample 3, 52 days, Back scattered electron image.

Electron microprobe with standards or EDS were used to obtain the concentration of each element in the IL. Results are summarized in Table II. Values in Table II are the average of at least five determinations. The concentration of Si resulted very similar in the IL for both alloys.

Sample	*Al-Si alloy*	*Time (days)*	*Technique*	*at % of the elements in the IL*			
				U	*Mo*	*Al*	*Si*
1	A 356	49	EDS	32	4	8	56
2	A 356	23	EPMA	31	5	6	58
3	6061	52	EDS	36	5	9	50
4	6061	90 (still on going)					

Table II. at % of the elements measured on the ILs.

XRD analysis was performed only on sample 2. The XRD pattern, shown in Fig. 3, indicates the presence of structures corresponding to γ-UMo, α-UMo, Al, Si. A set of small and wide peaks are considered to correspond to (U, Mo)(Al, Si)$_3$.

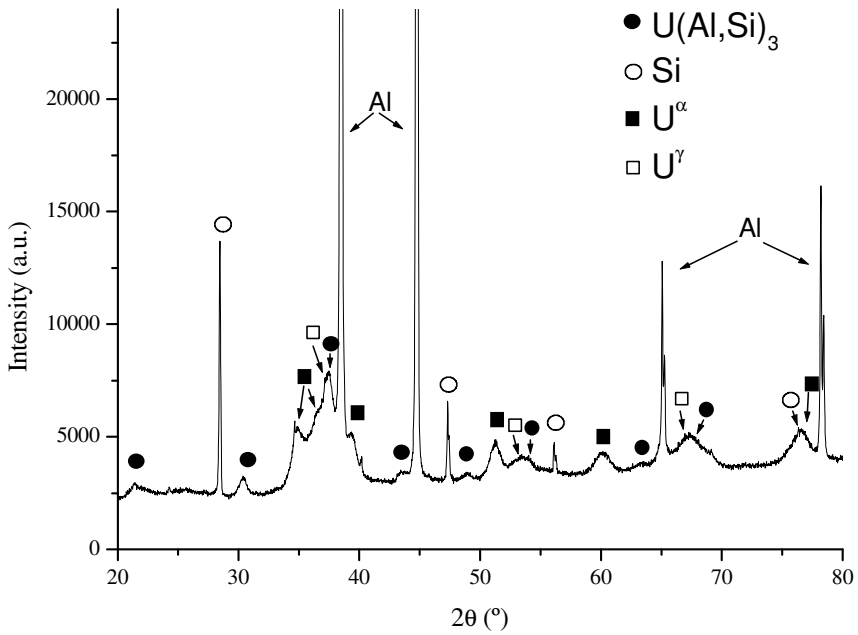


Fig. 3. XRD for Sample 2

Summing up all the information collected from the four techniques, it is shown that the IL between U-Mo and Al A356, at 340ºC, is formed by an only phase. This phase would be (U,Mo)(Al,Si)$_3$ with 57 at% of Si. This implies that going from 550ºC [4] to 340ºC, the two-phase zone disappeared remaining only the (U, Mo)(Al, Si)$_3$ which was found to be stable in dispersion fuel irradiation test for normal irradiation conditions.
Though intended, XRD was not possible for sample 3 and 4 (Al6061) due to the IL being so scarce. However the comparison of the concentrations of the elements in the IL obtained for both alloys, reported in Table II, strongly suggest that the IL would be the same. The only important difference found among the samples containing 6061 and A356 alloy is the thickness of the IL which is, in the first case, ten times thinner, Fig. 2. Longer thermal treatments are being carried out to obtain a zone thicker enough to perform XRD.
At 550ºC, for Al 6061, nor Si was measured on the IL neither a dealloyed zone was observed [4]. This is in agreement with the fact that this temperature is higher than the one at which Si enters in solution in this alloy,[10]. At 340ºC, the clear dealloyed zone observed, Fig. 2c, allowed to predict an IL containing Si even before its identification by SEM.

3. Possible changes on the interaction layer due to the consumption of Si while interdiffusion is still taking place.

Dispersion fuel elements, independently of its size or shape, are made of particles of fissile material surrounded by a pure Al matrix. Distance between particles is variable ranging from 0 to tens of μm. If pure Al were replaced by an Al-Si alloy, Si could be consumed in service

when the dealloyed zone associated to interdiffusion is of the order of the particle spacing. Diffusion would continue, then, with pure Al. Trying to detect if this produces any change in the IL, special samples were prepared. Advantage was taken from the fact that diffusion couples fabricated by FSW have flat and parallel faces by construction. A diffusion couple made with Al 356 was cut in half (Fig. 4 a). In each of them one of the faces was carefully polished to reduce the thickness of the Al alloy up to ~ 50 µm (Fig 4 b and c). This value was chosen from previous diffusion experiences in order to force the lack of Si while diffusion is still taking place. They were subjected to successive thermal treatments at 550ºC and 340ºC. Evolution of the IL was monitored at the end of each one. Details are presented in Table III.

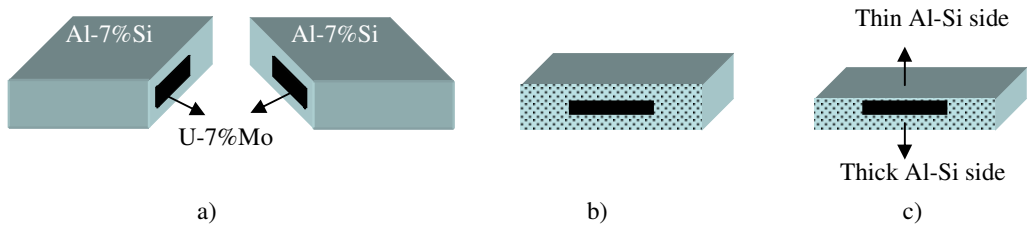
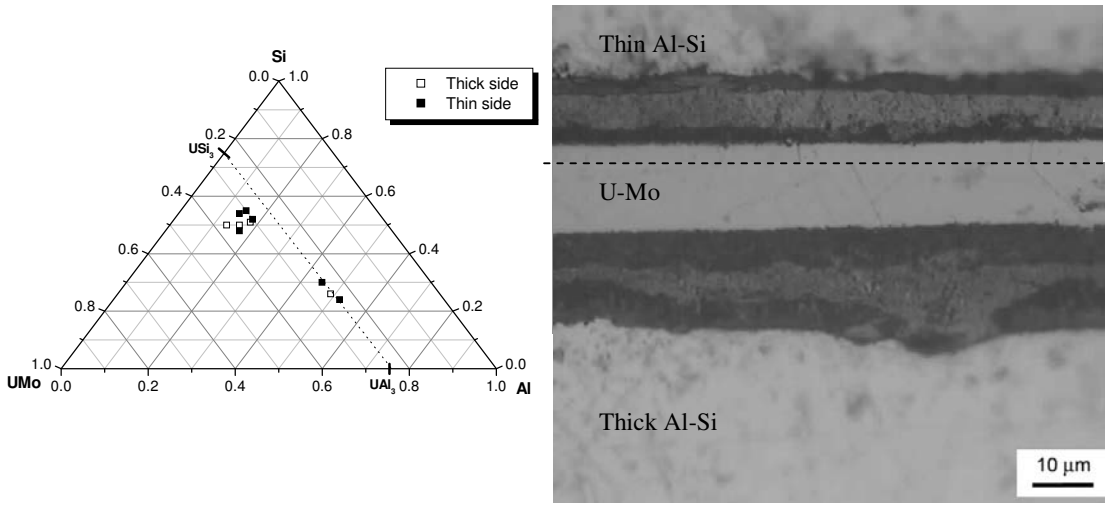


Fig. 4. Schematic representation of sample preparation for point 3.

Sample		Time	Temp. ºC	Thick side		Thin side	
				IL µm	Dealloyed zone µm	IL µm	Dealloyed zone µm
	A	1.5 Hs	550	9	55	8	44
I	**B**	(1.5 + 1.5) Hs	550	13	79	9	43
	C	(1.5 + 1.5 + 1) Hs	550	14	83	11	46
II	**D**	23 days	340	5	33	6	33
	E	(23 + 18) days	340	7	54	8	58

Table III. Thermal treatments and corresponding thicknesses of the interaction layers and dealloyed zones for samples in 3.

At 550 ºC, the IL between U-Mo and the thick Al-Si side grew faster than the other one, Table III. An important increment in the width of the dealloyed zone was observed for the thick side. For the thin Al-Si side, the dealloyed zone is constant, Table III, because Si was exhausted during the first thermal treatment being absent during the other two. OM, SEM and EDAX were used to analyze the IL for both zones. No indication of changes were observed between them, Fig. 5. Phases identified in these ILs are the same already reported [4].
At 340 ºC Si is recently over and no important differences in width or compositions were observed, Fig. 5 and 6. Only a slight Al increment could be infer but another thermal treatment is necessary to define this tendency if it really exist.



a b

Fig 5. Sample I in Table III.550ºC a) Concentration on a Gibbs diagram. b) Optical Metallography

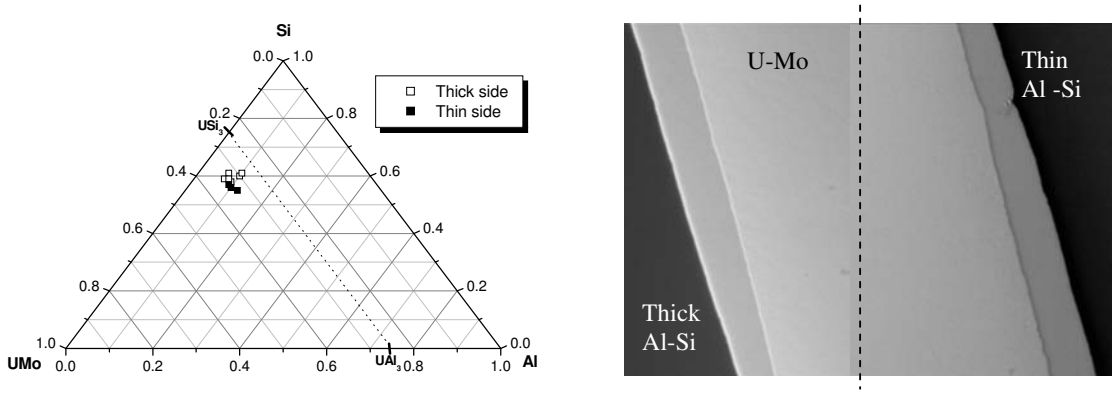


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Fig 6. Sample II in table III. 340ºC. a) Concentration on a Gibbs diagram. b) BSE image.

4. TTT diagram for U-7wt%Mo. Influence of Pt as γ-stabilizer

Other studies concerning properties of U-Mo alloys as part of a wider project are being carried on. One of them related with the diffusion studies, is the stability of γ U-Mo. Data for more temperatures were added to previous TTT diagram [4].

In order to know the influence of Pt as a stabilizing element, two U-7wt%Mo based alloys were studied, one of them with 0.9 wt.% Pt added. They were fabricated in an arc furnace and both homogenized in composition during 2h at 1000 ºC. Then, isothermal treatments at 560, 520, 480, 430 and 350 ºC were performed at periods of time between 1 and 560h. Experimental details were reported in [9].

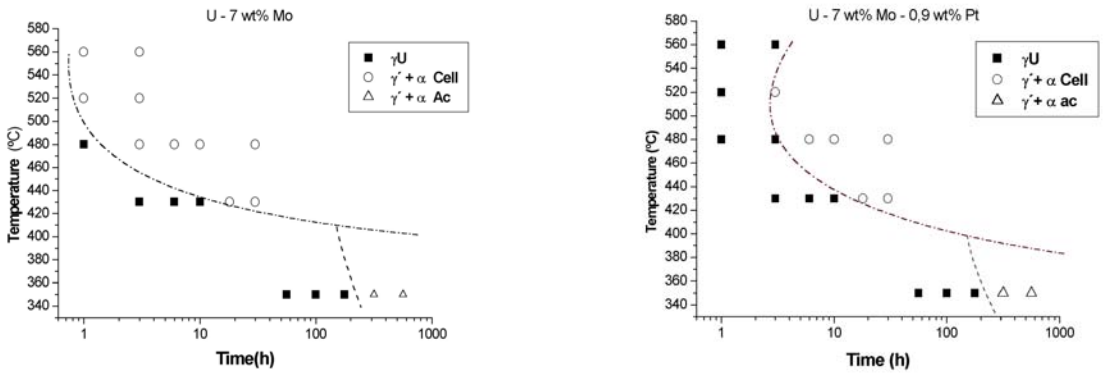

Fig 7. U-7wt%Mo and U-7wt%Mo-0.9wt%Pt alloys. Start of the decomposition γ phase.

Adding Pt, the start of the decomposition of the γ phase is delayed, being the initial grain size of the alloy an important variable which was taken into account.
The interdiffusion between Al and the U-Mo-Pt alloy was studied at 580 ºC. The (U, Mo)Al_3, (U, Mo)Al_4 y $Al_{20}UMo_2$ compounds were identified. This is similar to the result reported in [6] for U-7wt%Mo/Al at the same temperature.
Thus, the added Pt in low amount does not modify the type of intermediate compounds formed in the interdiffusion zone of U-Mo/Al couples at 580 ºC.

5. Conclusions

The results reported in this work for the effect in the interdiffusion U-Mo/Al when Si is added to Al support previous experiments and allow to conclude that

- at 340 ºC , the interaction layer for Al 6061 (0,6wt% Si) and A356 (7 wt.% of Si) is homogeneous in composition, that is, it is formed by only one phase with a concentration of approximately 32 at% of U, 4 at % of Mo, 8 at% of Al and 56 at% of Si.
- the structure of this phase was indexed by XRD for a sample made with the alloy A 356 and it is proposed to be (U,Mo)$(Al,Si)_3$. It has a greater concentration of Si than at 550ºC.
- the only difference between both alloys at 340ºC, is that the IL is ten times thinner for the alloy with less Si, that is Al6061.
- platinum delays the start of the decomposition of γU-Mo phase. In concentrations up to 0.9wt% does not introduce any change in the interdiffusion.

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