

INTERDIFFUSION BETWEEN U-Mo ALLOYS and Al

M. I. Mirandou**, S. N. Balart*, M. Ortiz*, M. S. Granovsky*, G. L. Hofman***

*Dpto. Materiales, CAC, CNEA, ARGENTINA ;

** I.T. J. Sábato UNSAM-CNEA ARGENTINA;

*** Argonne National Lab. ILL, USA

Abstract

During the fabrication and / or irradiation of the dispersion fuel elements, the fuel particles react with the surrounding Al matrix. This reaction results in the formation of a zone consisting of intermetallic compounds. The low thermal conductivity of these compounds has a major effect on the fuel temperature as well as on the swelling of the fuel.

Interdiffusion between U-Mo /Al is being investigated using chemical diffusion couples. In this paper the first results obtained with optical and scanning electron microscope, electron microprobe and X-Ray diffraction are presented.

Investigation of the effect on the formation of the interdiffusion zone of small additions of Mg to Al is the primary purpose of this study.

Introduction

For the past several years research in the fuels area led to the development of aluminium- based dispersion fuels containing U-Mo as fuel particles.[1]. These fuel elements allow fulfillment of requirements to reduce the enrichment of U used in research and test reactors.

Results of irradiation test are promising , how ever, some efforts are focused on the problem of the reaction between the Al matrix and U-Mo particles [1,2].

It is well known, when two metals are in contact an interaction zone may develop formed by new phases with the composition in between that of the terminal components. This produces

changes in the specific volume and in the thermo mechanical properties of the couple. This has implications in the fabrication processes as well as the behaviour in service.

The effect of irradiation temperature on the extent of the fuel-aluminium interdiffusion and the diminution of matrix has been shown to be important [1]

Early diffusion experiments done with diffusion couples were found only concerning pure U and Al [3, 4]. And more recently experiments with dispersion fuel samples of U-Mo and pure Al [5]

Characterization of the reaction zone for U-Mo / Al and its eventual control would be valuable. In this work the interdiffusion between U-Mo /Al is being investigated using chemical diffusion couples, more specifically, the effect on the interaction zone of small additions of Mg to the Al is studied.

This study is in its initial stage and therefore only provides an indication of the effect of Mg on the diffusion rates. A more complete set of samples, including various compositions of Al-Mg is planned for various annealing times and temperatures.

Experimental.

Materials employed in this study were high-purity Aluminium (99,99%), Al -Mg commercial alloys and two arc-melted U - 7% Mo alloys, fabricated from two kind of U(a and b). Table 1 and 2 give the composition of the Al alloys and relevant impurities in the two kind of U respectively.

TABLA 1 : composition of comertial Al alloys (wt%)

	Mg	Cu	SI	Cr
Al 6061	1.0	0.27	0.6	0.2
Al 5052	2.5	---	---	0.25

TABLA 2: content of relevant impurities of U (ppm)

	Al	Mg	Fe	Si
U(a)	> 200 *	< 3D	60 -120	~ 25
U(b)	< 10 D	~ 60	~27	~ 24

* EPMA measurements gave ~2 at.%

γ phase U-Mo alloy was obtained by heat treating the as cast alloy at 1000°C during 24 hs and quenched to room temperature. The U-Mo grain size obtained by this treatment was 500 to 1000 μ m.

Pieces of approximately 2 x 5 x 5 mm were cut from the original materials to form the couples. Samples to be analysed by X-Rays diffraction (XRD) were 2 x 8 x 15 mm.

Pieces were mechanical polished up to 1 μ m diamont paste and ultrasonically degreased in ether ethilic. In the case of aluminium a pretreatment in dilute sodium hydroxide solution was used to remove the oxide layer.

Diffusion couples consisted in one piece of Al and one piece of U-Mo alloy or a double sandwich Al/ U(Mo)/Al(Mg). They were kept in tight contact during treatments by a stainless-steel mechanical clamp. Thermal treatments were done in sealed quartz tubes with high purity Ar atmosphere.

Interdiffusion anneals were performed between 400°C and 580°C to obtained measurable interphase layers in reasonable times.

After the diffusion anneals, the couples were sectioned parallel to the diffusion direction. For XRD successive surfaces at a small angle from the perpendicular to the diffusion direction were exposed by a careful polishing in an abrading machine specially design to keep paralelism between each layer, and a spectrum was taken each time. An spectrum taken of a cross section of the reaction layer would give the lines corresponding to all the species present.

The characterization of the samples was performed using optical microscopy (OM), sanning electron microscopy (SEM), energy dispersive analysis (EDAX), electron microprobe microanalysis with standarts (EPMA) and X-Ray diffraction (XRD).

Results

Interaction layers of appropriate width were obtained for anneals up to 4 hs at 580 °C. At 475°C at least 26 hs was needed. The γ phase in the U-Mo alloys was retained during this periods of time.

U-Mo/Al interdiffusion

Figure I shows an optical micrograph of a typical section of a couple. The intermediate layer appears uniform with planar interface specially at the U-Mo side. No cracks were visible. Mechanical polishing did not reveal different phases in the reaction layer. An electrolytic polishing with opalu B followed by chemical etching with hydrofluoric acid solution, revealed two different colored zones with a fine microstructure, Figure II.

The width of the interdiffusion layer varies from 25 microns for a 0.5 hs anneal to 175 microns for 4 hs at 580°C.

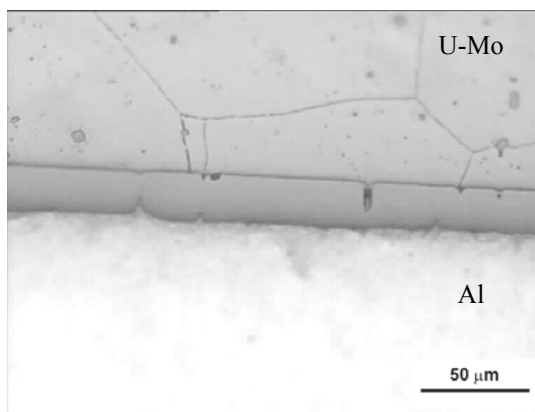


Fig. I. Interdiffusion layer. 0.5 hs 580°C.
Mechanical polished.

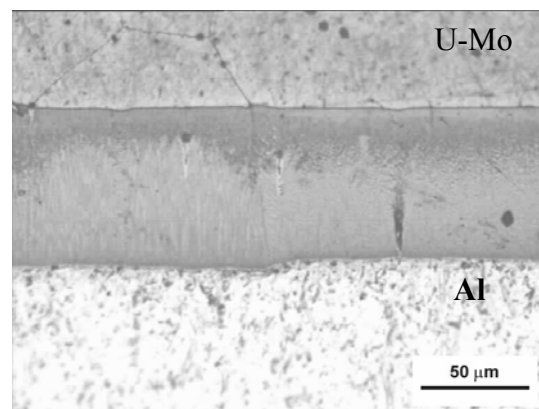


Fig. II. Interdiffusion layer. 2 hs 580°C
Chemical etched

The quantitative distribution of each element across the reaction layer was measured with EPMA against standarts. It is shown in Figure III. Measurements done close to the reaction layer did not shown the diffusion of Al into U-Mo nor U or Mo into Al.

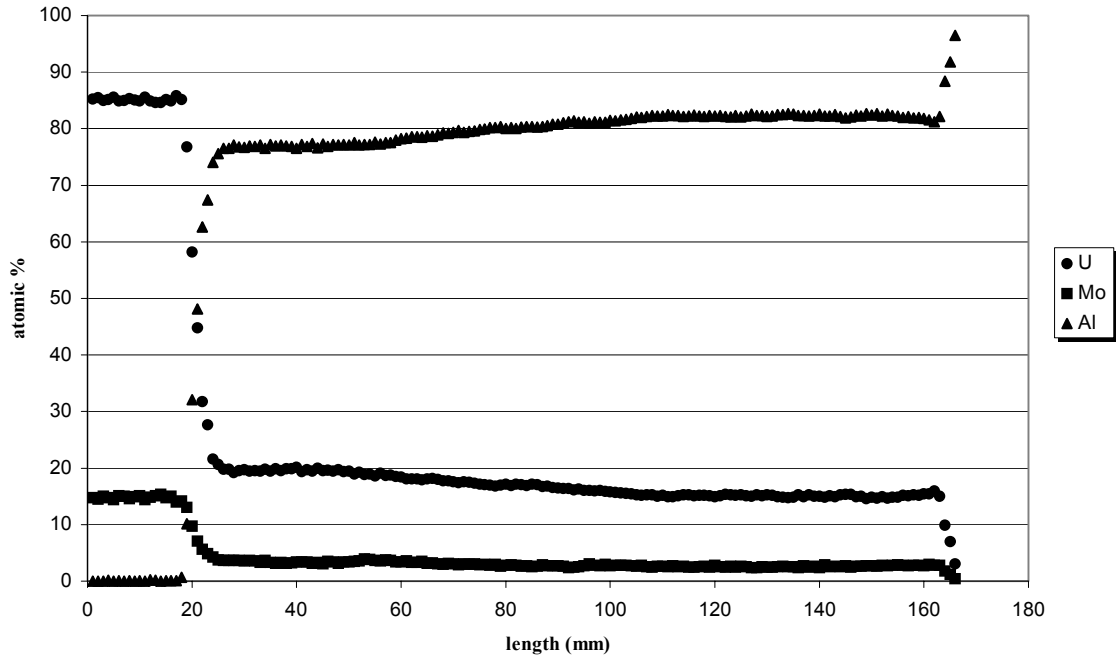


Fig.III. Quantitative distribution of each element across the interaction layer.

XRD was performed to identify the compounds present in the layer. Figure IV shows some of these spectra. The presence of UAl_4 as the first intermetallic from the aluminium side is clearly depicted. Up to the analysed depth the presence of UAl_3 was also identified, however, in small proportions.

U-Mo/Al-Mg interdiffusion

When dilute alloys of Al with Mg were used as counterpart the microstructure revealed by metallographic results were similar to the case of pure Al, that is, mechanical polishing showed a uniform layer with planar interface and two distinguishable coloured zones when chemically etched. (Figure V).

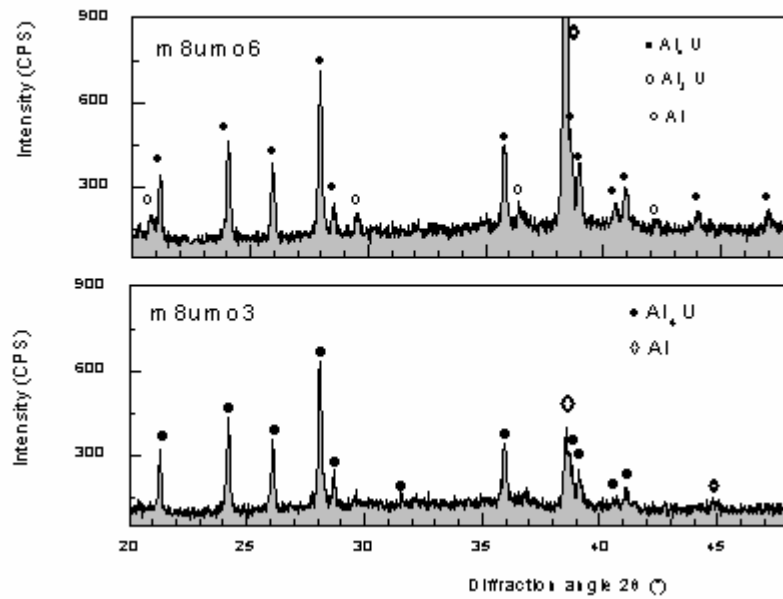


Fig.IV. XRD spectra at different depth in the interdiffusion layer. Bottom: first intermetallic close to the Al. Above: deep inside the layer.

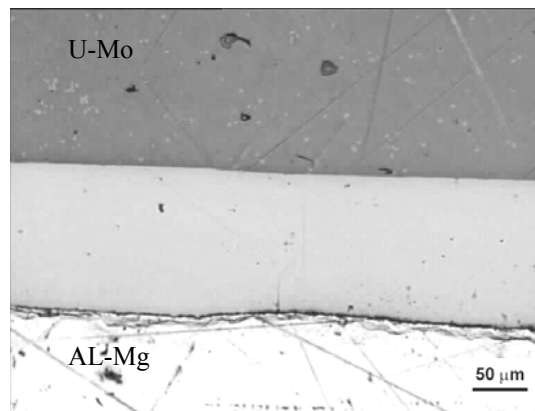


Fig. V. Interdiffusion zone for U-Mo/Al-Mg. 2hs, 580 °C. Chemical etched.

Even though a careful handling, several couples broke apart at AlMg side specially for Al 6061 alloy. Concerning the width of the layer was difficult to establish a systematic behaviour at

580°C. At 475°C no interaction was observed at the Al-Mg side meanwhile 3 microns were measurable at the Al side.

Discussion and conclusion

The reaction between U-Mo and Al was detected as soon as these alloys began to be tested as a new high density LEU fuel. Out of pile experiments on meats also allowed to measure the kinetics at different temperatures [5], but the constituents of the layer were not unambiguously identified. The results of the present study confirm the fact that provided γ phase is not decomposed, the interaction layer is uniform and has smooth or planar interface, similar to what is observed in fuel irradiation test. XRD shows the presence of at least two intermetallics UAl_4 and UAl_3 . Taking into account the results from EPMA (Fig. III), both contain amounts of Mo. That figure also shows an Al distribution consistent with the phases composition. There is no measurable Al content in U-Mo. The chemical etch revealed two zones that would correspond to those intermetallics.

As shown in Fig VI there appears to be a difference in the diffusion kinetics for the two uranium alloys used, presumably due to the different impurity levels of the uranium (see Table 2). However, in all cases the measured width of the interdiffusion zone with either Al-6061 or Al-5052 alloy are smaller than with pure Al. The number of experimental samples is at this point insufficient to draw more than this preliminary conclusion.

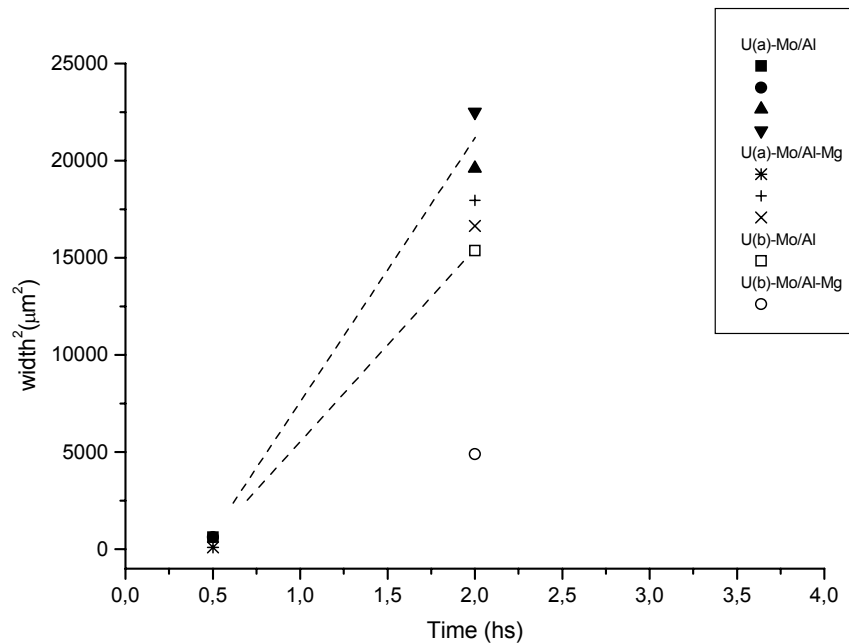


Fig.VI. Square width of the interdiffusion layer vs time at 580°C

Acknowledgments

The authors thank the staff of the Lab. of Metallography, SEM-EDAX Lab., DRX Lab., Electron Microprobe Lab. and Glass Shop for their help.

This work was partially supported by PIP 1064/98 CONICET, Argentina.

REFERENCES

- [1].- G.L. Hofman and J.L.Snelgrove, L.Hayes and M.K.Meyer, "Progress in development of low enriched U-Mo dispersion fuels". International Meeting on Research Reactor Fuel Management, March 17 –20, 2002, Ghent, Belgium.
- [2].-M. Meyer, G. Hofman, R.Starin, C.Clark and J.Stuart, Proc. 23er. RERTR International Meeting, Las Vegas USA. Oct. 1-6, 2002
- [3]D. Subramayan, M. Notis and J. Goldstein. Metallurgical Transaction A, 16^a, 1985, pp589.
- [4].- L. S. Castleman, Jof Nucl. Mat. 3, 1 (1961), 1-15
- [5].- K-H.Kim et all. Proc. 23rd RERTR Meeting, Las Vegas USA. Oct. 1-6, 2002