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MICROSTRUCTURAL CHARACTERIZATION OF DISPERSION FUEL MINIPLATES MADE OF HYDRIDED U + 7wt% Mo POWDER.

L. Olivares, J. Marin, J. Lisboa and M. Barrera.
Nuclear Materials Department
Chilean Nuclear Energy Commission - CCHEN
95 Amunategui St.,
Postal Code 6500687
Santiago – Chile.

ABSTRACT

This paper presents the characterization results of miniplates based on hydrided U+7wt%Mo powder developed at CCHEN´s fuel facility. Two hydriding methodologies were applied and the powders produced were dispersed in Al+4wt%Si matrix to produce miniplates with uranium density of 8gU/cm^3 with either LEU or natural uranium, according to RERTR design and specs guidelines. Metallography of longitudinal cross-section has shown some irregularities in cladding thickness. SEM+EDS inspection indicated the formation of an interaction layer composed by U-Mo-Al around UMo particles; this phenomena started at the degassing - annealing stage of the compact and then continued during the course of the fabrication processes. Out-of-pile swelling test revealed moderate volume increasing, less than 1%, which remained stable until the end of the vacuum annealing (350°C/37 hrs). The homogeneity examination did not exhibited fuel particles segregation, white points or stray particles, and UT scanning showed acceptable bonded meat and cladding and evidences no delamination, but nevertheless indicated some differences in cladding thickness towards both ends of miniplates. Fuel/matrix contact areas showed interaction layer formation whose growth advanced by reactive diffusion through the entire particles. In conclusion, some critical questions about the potential application of hydrided UMo powders in dispersion fuel are still open. Additionally, some fabrication processes must be adjusted to assure the integrity of cladding and also to improve the homogeneity of fuel/matrix distribution

1. Introduction

Nowadays it is generally accepted that hydriding is a third route to obtain powder from U-Mo ductile alloys [1]. An alternative phyrochemical process has been developed in addition to methodologies based on physical fragmentation, mainly atomization or based on mechanical forces as grinding or machining of ingots. The main variables in hydride – dehydride process are the temperature and time of heat treatment, hydrogen pressure, dehydriding conditions and additional annealing for homogenization, and quenching and passivation of powders. A previous study of hydriding allows concluding that is possible to produce UMo powders with characteristics proper for dispersion type fuel fabrication [2]. Irregular morphologies, reactivity due to pyrophoricity, relevant residual contents of oxygen and hydrogen, brittleness of particles, interaction with Al matrix and as-fabricated porosity appears as relevants during fabrication and inspection of test miniplates [3,4,5] and could predict its behavior under irradiation. It seems that the only way to obtain definitive answers in relation to applicability of this type of powders is to apply irradiation tests according to a well designed irradiation, monitoring and follow-up qualification program, as the RERTR series experiments. This paper summarizes the main aspects of manufacture and characterization of miniplates fabricated recently in CCHEN and shown the relevant results of inspections for LEU and NU miniplates. In the final part of this paper, results of interaction tests and out-of-pile swelling test are discussed.

2. Experimental set-up

Uranium + 7 wt% molybdenum (UMo) alloys were prepared through induction melting, starting from either natural uranium or LEU (19,75% U-235) plus the addition of Mo metal (99,9%). The UMo powders were obtained by two hydriding methodologies: through either hydrogen decomposition of UMo alloy into $\alpha + \gamma'$ phases (550°C/4 hours) for porous powder, or hydrogen solubilization in gamma phase (800 °C/1hour), cooling and a long thermal cycle at low temperature for slow decomposition and hydrides consolidation (120°C/24 hours) for producing dense powder. The fragmented alloys were sieved below 150 μm and the oversize material was ground by rotating mill, sieved and added to fine powder. Since 2009, 30 miniplates has been fabricated with this type of powder, and complying with RERTR design and specifications. Seven of them using LEU and other 23 containing NU. The matrix material was prepared by blending pure Al powder (80% <45 μm) with silicon powder (100%<15 μm), followed by a vacuum annealing degassing and then sieving for dissagregation. The cladding material was always Al-6061.

In section 3 are tabulated the fabrication parameters and some manufacturing results of the miniplates, and whose characterization included dimensional meat controls, homogeneity, cladding thickness by means of metallographic inspection, bending test in same cases, and bonding test using UT scanning and blister annealing.

For evaluation of the interaction between Al-Si matrix and fuel particles, four samples were taken from the meat zone of UMo-54 LEU miniplate (prepared with HMD dense powder). The interaction tests were performed through vacuum annealing (500°C/ 4 and 6,5 hours). Transverse cross-section microstructures specimens were obtained and inspected using scanning electron microscopy (SEM). The elemental composition of the interaction layers were revealed by using energy dispersive X-ray spectroscopy (EDS) analysis. Finally, miniplate UMo-52 NU was submitted to an out-of-pile test, i.e., it was annealed in air (350°C/ up to 36 hours). For all fabricated miniplates, the compacting pressure was 23 Tons applied to a 4 cm^2 area (564 MPa).

3. Results and Discussion

According to the results summarized in table 1, some NU miniplates could not strictly comply with the specifications, mainly because they were manufactured for the purpose of final processes adjustments. Major efforts were focused in the approval of LEU miniplates.

Table 1. Fabrication parameters and results for selected test miniplates manufactured in CCHEN under RERTR design

Miniplate number		UMo-52	UMo-54	UMo-55	UMo-58	UMo-64	UMo-65	UMo-66	UMo-67	UMo-68	UMo-70
Uranium type		NU	LEU	NU	NU	LEU	LEU	LEU	LEU	NU	NU
Hydrided UMo Fuel (U + 7wt% Mo)		Dense	Dense	Dense	Dense	Dense	Dense	Porous	Porous	Dense	Dense
Fuel density g/ cm³		16,15	16,15	16,15	16,15	16,15	16,15	11,45	11,45	16,15	16,15
Nom Loading gU/cm³		8,0	8,0	8,0	8,0	8,0	8,0	8,0	8,0	8,0	7,0
Real Loading gU/cm³		7,53	7,53	7,53	7,53	7,53	7,53	6,3	6,3	7,53	6,6
UMo Fuel weight (g)		6,40	6,40	6,40	6,40	6,40	6,40	6,4	6,40	6,40	6,18
Matrix (Al+4wt%Si) (g)		1,05	1,05	1,05	1,05	1,05	1,05	1,05	1,05	1,05	1,27
Compact measures (mm)	Length (mm)	22,33	22,40	22,41	22,35	22,34	22,37	22,35	22,37	22,33	22,34
	Wide (mm)	17,98	17,96	17,96	17,90	17,88	17,89	17,91	17,94	17,89	17,89
	Thickness (mm)	2,45	2,78	2,80	2,54	2,51	2,52	2,42	2,43	2,59	2,75
Meat measures (mm)	Length (mm)	93,5	67,8	83,0	105,8	78,2	79,5	78,7	77,4	84,2	82,4
	Wide (mm)	20,2	18,3	18,3	20,1	18,6	18,7	18,5	18,5	19,3	19,5
	Thickness (mm)	0,63 (3)	0,88 (3)	0,71 (3)	0,63 (1)	0,69 (3)	0,67 (3)	0,65 (3)	0,66 (3)	0,68 (3)	0,74 (3)
Miniplates measures (mm)	Length (mm)	(2)	(2)	(2)	(1)	(2)	(2)	(2)	(2)	(2)	(2)
	Wide (mm)	(2)	(2)	(2)	(1)	(2)	(2)	(2)	(2)	(2)	(2)
	Thickness (mm)	1,38	1,82	1,48	1,18	1,50	1,47	1,47	1,48	1,48	1,54
Total Reduction (%)		74,4	68,3	74,6	78,7	72,6	73,3	73,0	72,9	73,7	73,1
Reduction rate		1 : 3,91	1 : 3,15	1 : 3,94	1 : 4,69	1 : 3,65	1 : 3,75	1 : 3,70	1 : 3,69	1 : 3,80	1 : 3,72
Main Characterization		Out of pile Swelling	Interact Layer	UT Scanning	Metallo graphy	Fabricated for Irradiation tests					

(1) metallographic inspection; (2) No final cut; (3) Calculated values

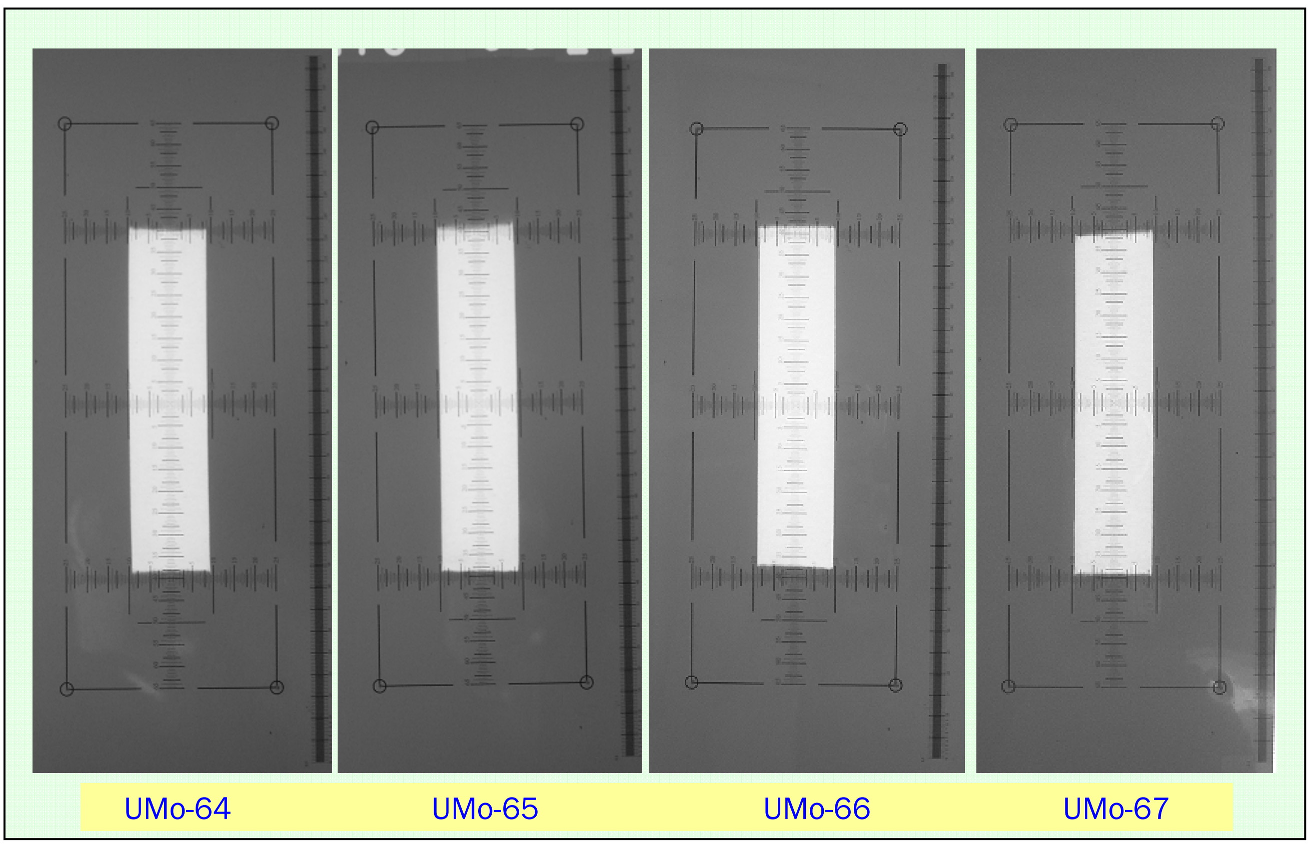


Figure 1. Radiographs for metrology, location and homogeneity of meat in LEU miniplates

The images showed in figure 1 correspond to LEU miniplates. The dimensional control of the meat satisfied the limits included in pattern transparency placed over the radiographs. But it must be considered that these miniplates were in a fabrication stage prior to the blister test and cold rolling stage. In relation to homogeneity, and based on visual inspection and homogeneity evaluation using optical densitometry, the results were considered qualitatively satisfactory because it was not detected evidence of segregations, white spots or stray fuel particles (out of meat zone).

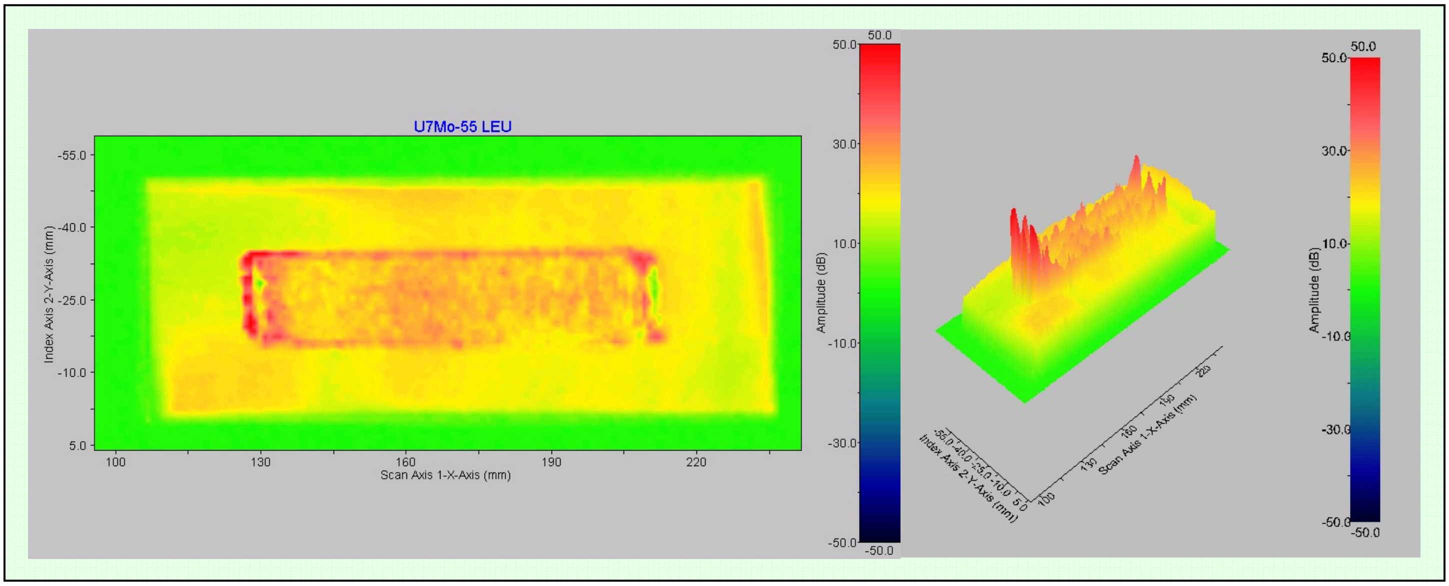


Figure 2.- UT, bonding test applied to UMo-55 LEU test miniplate

Figure 2 presents the UT scanning results for the UMo-55 LEU test miniplate. After hot rolling and blister test applied to this miniplate, non bonding, blisters or pillowing spot were not observed.

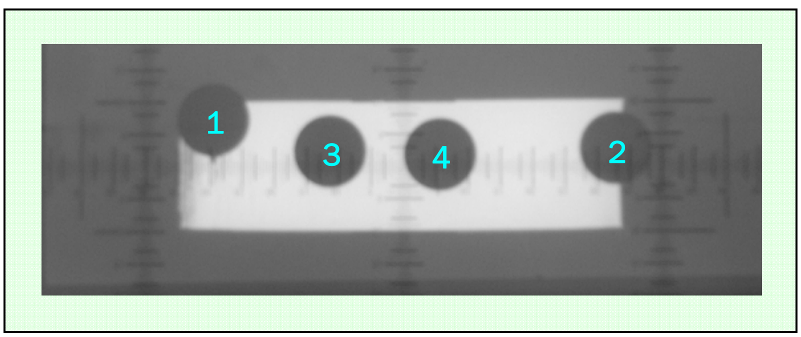


Figure 3. Radiograph of UMo-54 LEU miniplate showing the location of the specimens taken by punching for the interaction test. Specimens 1 and 2 before annealing, specimen 3 annealed 4 hours and specimen 4, annealed by 6,5 hours

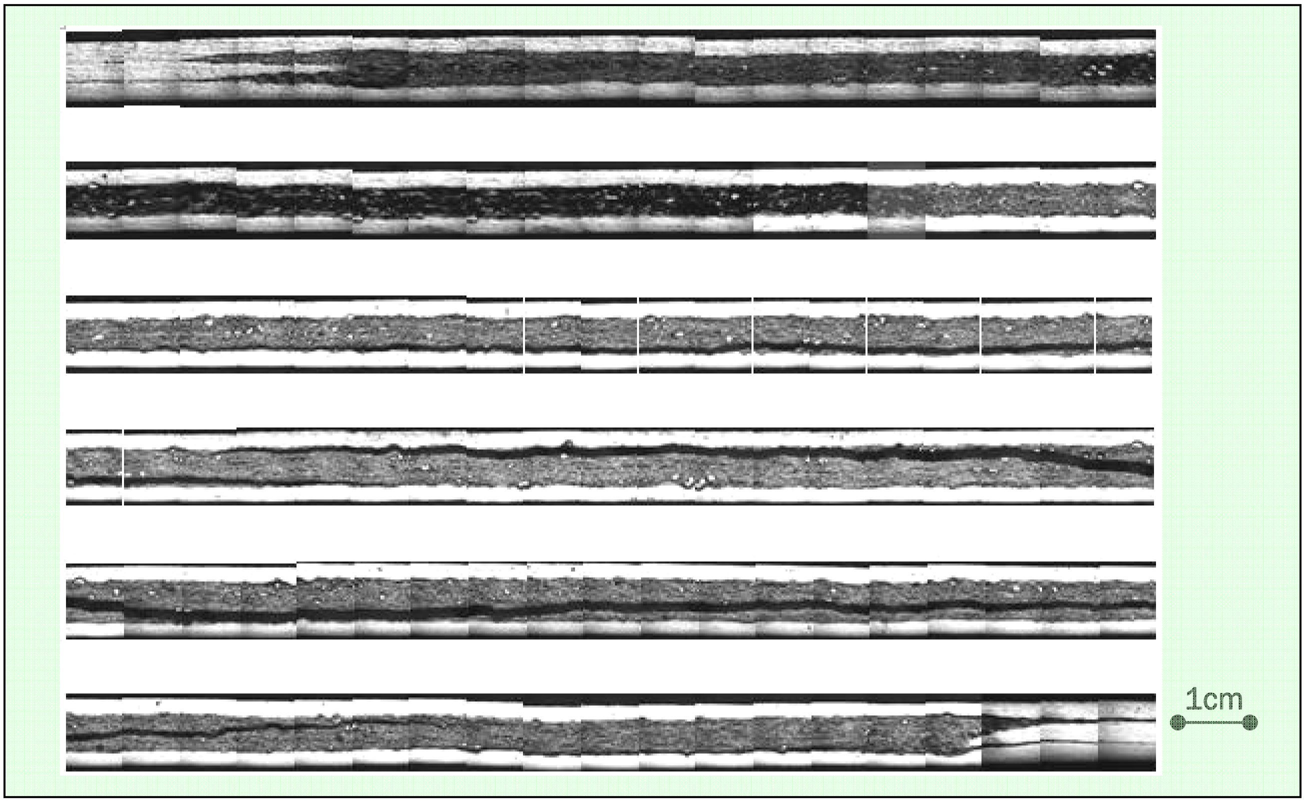


Figure 4. Micrographs of the longitudinal cross-section along the meat for the UMo-58 miniplate.

Optical micrographs showed in figure 4 reveals typical defects for dispersion fuel, as tail fish and dog bone, on both end sides of the meat. In these zones the minimum cladding was 0.20 mm. Also is possible to see points of aluminum accumulation. Towards the center of the miniplate, appears an open zone due to residual stress of the forces applied during cutting and metallographic preparation; however it can not be assumed as meat / cladding non-bonding. The average measured thickness values were 0,66 mm for the meat, 0,31 mm for the cladding and 1,28 mm for the UMo-58 miniplate.

Figure 5 shown the bend test results for UMo - 58 NU miniplate. There was not evidence of delamination in the samples taken from areas adjacent to meat zone.

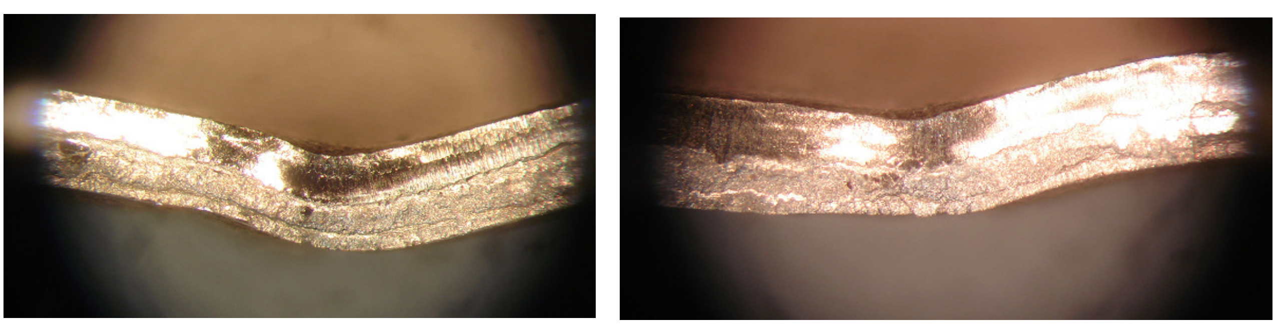
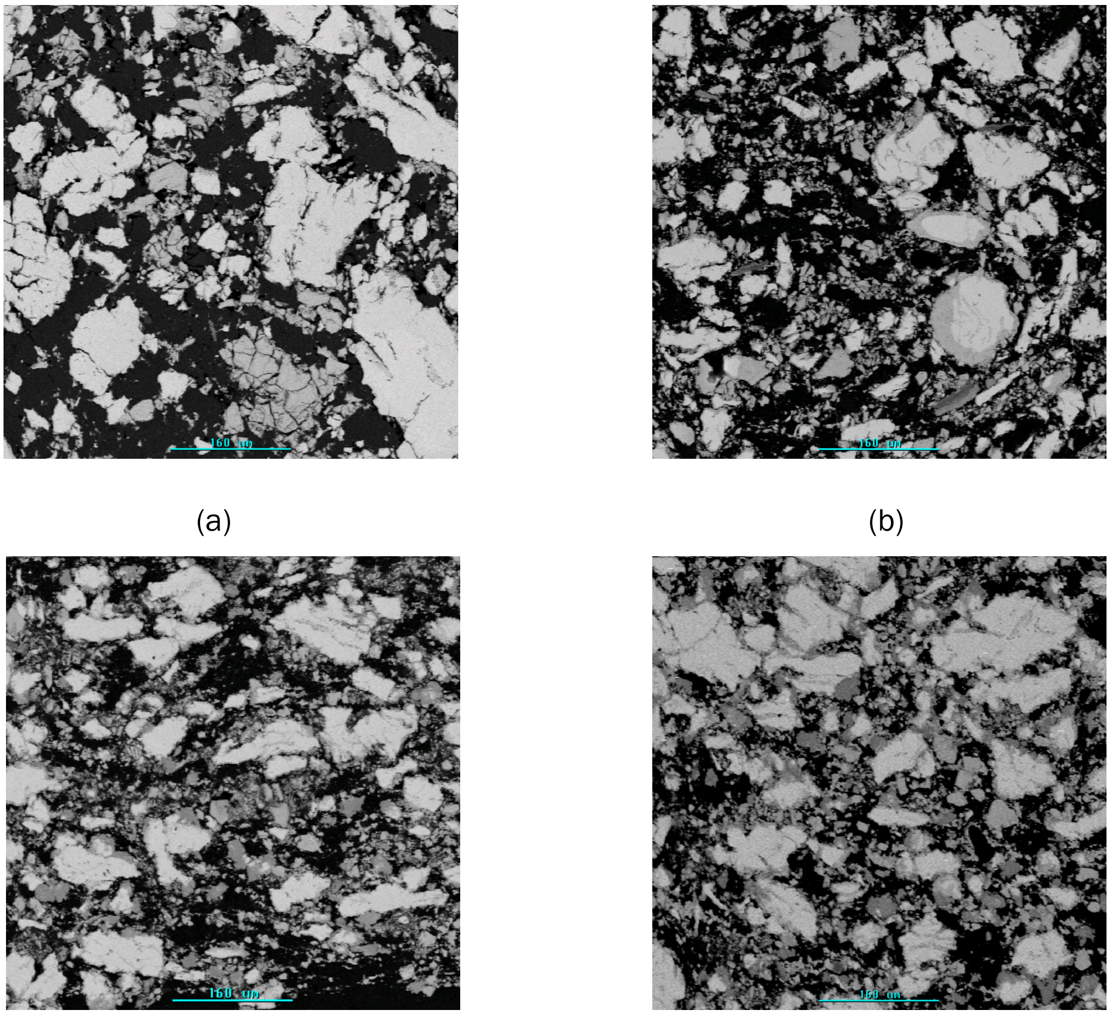


Figure 5. Bend test applied to UMo-58 NU miniplate

In figure 6, SEM micrographs shown in (a) the specimen as-compacted, before framing and rolling showing no evidences of interaction layer surrounding fuel particles, although it appears broken into many fine particles, this most probably due to uniaxial force applied during compacting. In (b) an after rolling micrograph, taken, there are evidences of reactive diffusion activated by temperature during fabrication process, producing interaction layers with irregular thickness around fuel particles. Here, the coarser particles appear surrounded by fine particles generated by mechanical fragmentation, as a consequence of the application of the plastic deformation process. In micrographs (c) and (d), apparently the annealing applied for the interaction test resulted in coalescence of finest particles, for these samples, and composed almost completely by (U,Mo) aluminides (grey phase). In figures 7, 8, 9 and 10 is possible to identify three phases, the lines in SEM micrograph indicates the place where the linescan compositional analyses was taken and at the right are shown the concentration profiles for fuel and matrix constituents.



(a) (b) (c) (d)

Figure 6. SEM micrographs of UMo-54 LEU samples: (a) as-compacted (before framing and rolling), (b) as-fabricated, (c) annealed (4 hrs/500 °C) and (d) annealed (6,5 hrs/500 °C).

The interaction produced by the reactive interdiffusion seems to advance by the grain boundaries according to what is observed in figures 7, 8, 9 and 10. The Al and Si inside the fuel particle come from the matrix and diffuse inside the fuel particles through reactive interdiffusion mechanisms. Besides, the U and Mo profiles indicate evidences of movement although migration of these elements outside the particles boundaries was not detected.

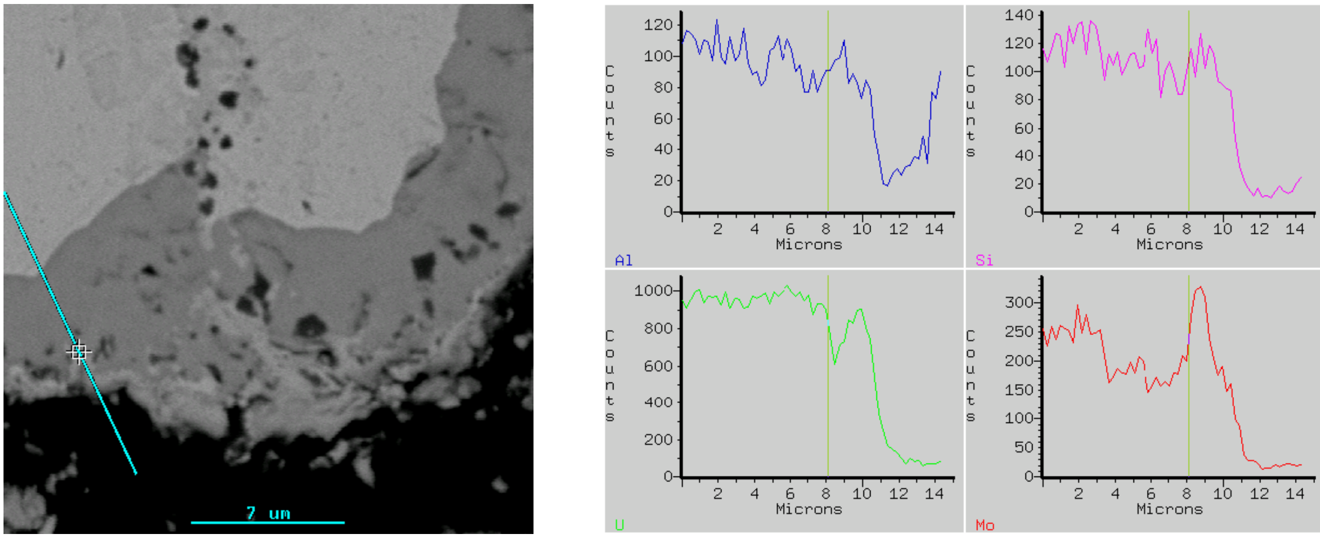


Figure 7. SEM image and EDS linescan elemental profiles from UMo particle to Al-Si matrix through IL for UMo-54 LEU miniplate as- fabricated condition, before interaction annealing.

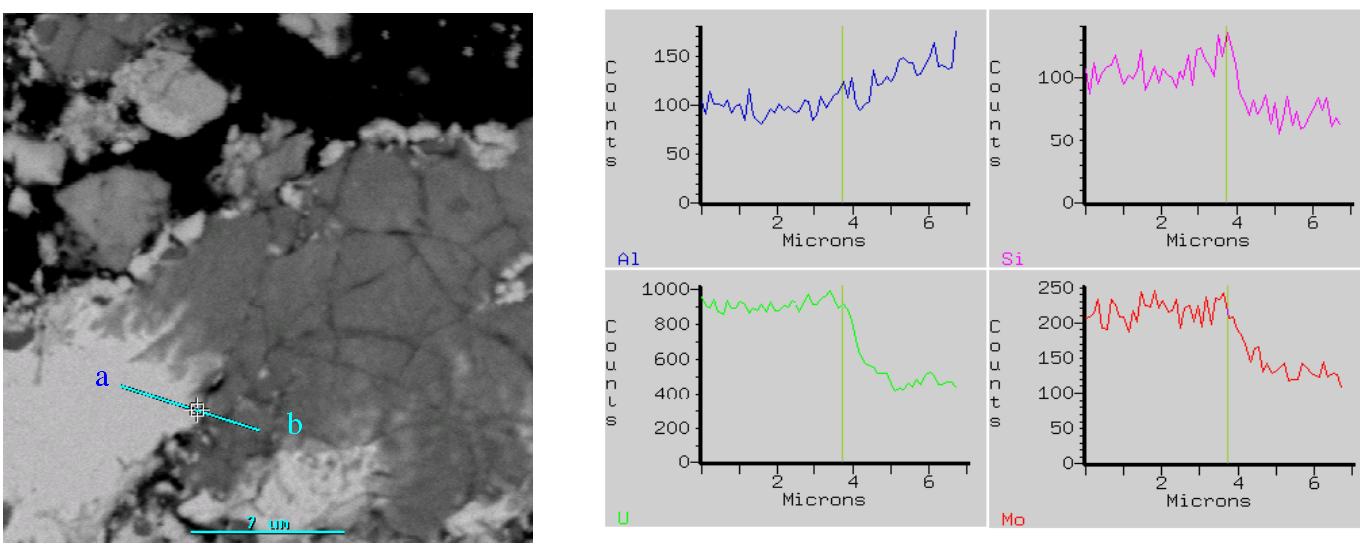


Figure 8. SEM image and EDS linescan elemental profiles from unreacted UMo white phase to grey phase (U,Mo) applied to UMo-54 LEU miniplate annealed (4 hrs/500 °C)

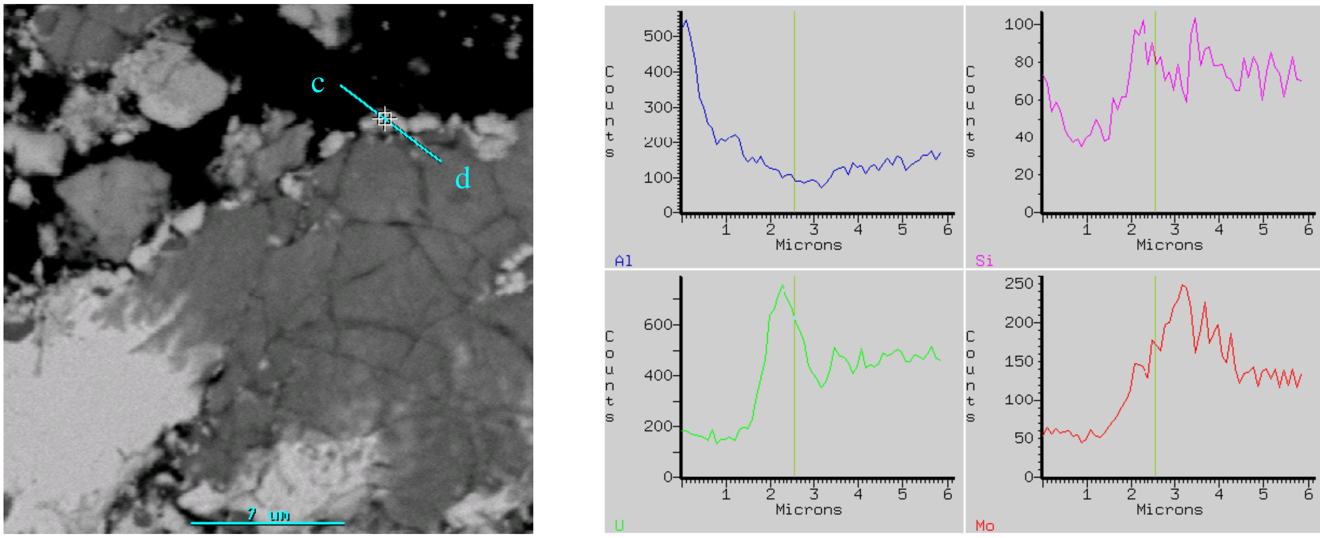


Figure 9. SEM image and EDS linescan elemental profiles from Al-Si matrix (dark phase) to grey phase (U,Mo)Al through IL-2 (point c) applied to UMo-54 LEU miniplate annealed (4 hrs/500 °C)

The interaction seems to start at the grain boundaries according to what is observed in figure 9

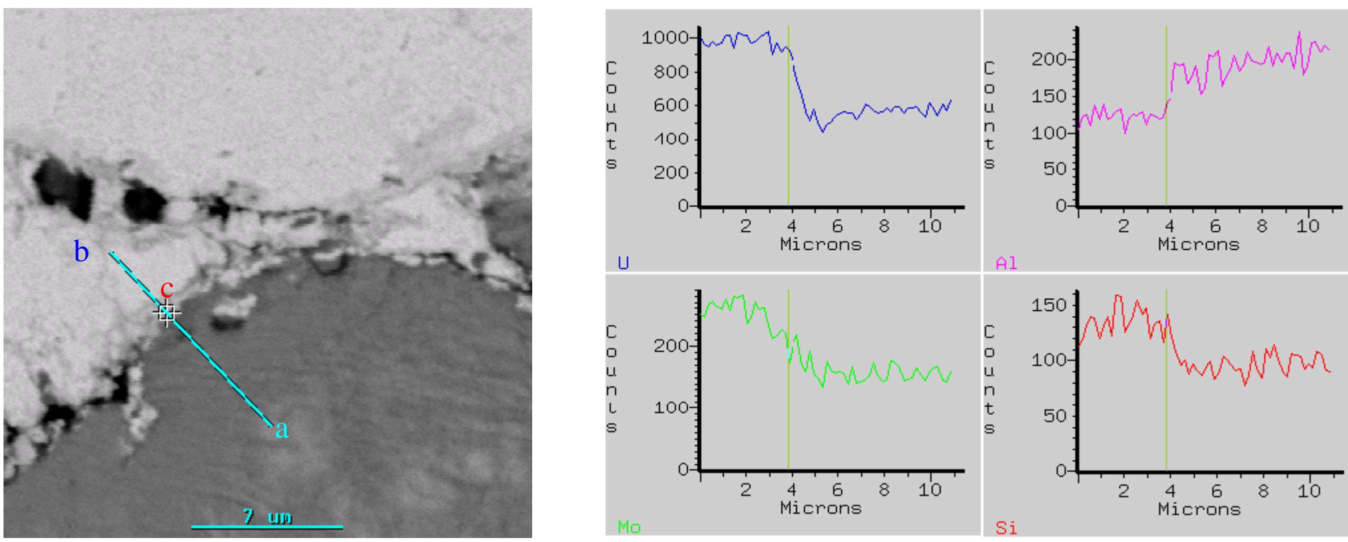


Figure 10. SEM image and EDS linescan elemental profiles from (U,Mo)Al (grey phase) to UMo (white phase) through point c applied to UMo-54 LEU miniplate annealed (6,5 hrs/500 °C)

Summarizing the interaction characterization, the starting microstructure was a matrix or Al-Si (dark phase in SEM) in which were dispersed fine particles of hydrided UMo fuel powder (white phase in SEM) The first signs of phase transformation appear during the manufacturing processes, probably after the compact degassing or after hot rolling. As a consequence of these transformations appears a third phase in the UMo/matrix interlayer (gray phase in SEM), whose growth occurs rapidly inside the fuel particles due to a reactive interdiffusion mechanism. According to the EDS linescan results and spot-scanning results, the constituents of this third phase might be (U,Mo) aluminides. According to the SEM micrographs comparison, these interdiffusion phenomena, besides the phase transformations, may produce fragmentation and coalescence of fuel particles.

Table 2. Summary of results for EDS Point-scanning analyses applied to the interaction specimens taken from UMo-54 LEU miniplate

Results for EDS Point-scanning composition analyses

EDS Analysis	Specimen Type	Element measured - atomic % Al	U	Mo	Si	Possible compounds	Comments
UMo-54 c – 1 IL grey phase	As-fabricated before annealing	80,59	12,13	2,13	5,15	aluminides (U,Si,Mo)Al	Point scanning in middle of grey phase or Interact. Layer Figure 6
UMo-54b-1 general view	Interaction anneal. 4 hours/500°C	76,29	17,21	4,28	2,22		EDS scanning in whole area of micrography Figure 5 (c)
UMo-54b-2a UMo original white phase	Interaction anneal. 4 hours/500°C	2,85	83,12	14,03	-------	UMo + (U,Mo)Al	EDS scanning inside UMo particle Figure 7
UMo-54b-2b IL grey phase	Interaction anneal. 4 hours/500°C	66,65	19,3	3,72	10,33	aluminides (U,Si,Mo)Al	EDS scanning inside grey phase or IL Figure 7
UMo-54b-2c Dark Phase Al-Si Matrix	Interaction anneal. 4 hours/500°C	64,79	3,29	1,0	30,93	(U,Si)Al	EDS scanning inside dark phase, matrix Figure 8
UMo-54-1 general view	Interaction anneal 6,5 hours/500°C	72,8	18,79	3,29	5,12		EDS scanning in whole area of micrography Figure 5 (d)
UMo-54 2a IL grey phase	Interaction anneal 6,5 hours/500°C	64,39	18,26	1,42	15,93	aluminides (U,Si,Mo)Al	EDS scanning inside grey phase or IL Figure 9
UMo-54 2b UMo original white phase	Interaction anneal 6,5 hours/500°C	-------	86,95	13,05	-------	UMo	EDS scanning inside UMo particle Figure 9
UMo-54 2c IL-2 IL grey phase/UMo	Interaction anneal 6,5 hours/500°C	5,61	86,94	6,42	1,03	UMo + (U,Mo)Al	IL-2 between IL-1 and UMo particle Figure 9

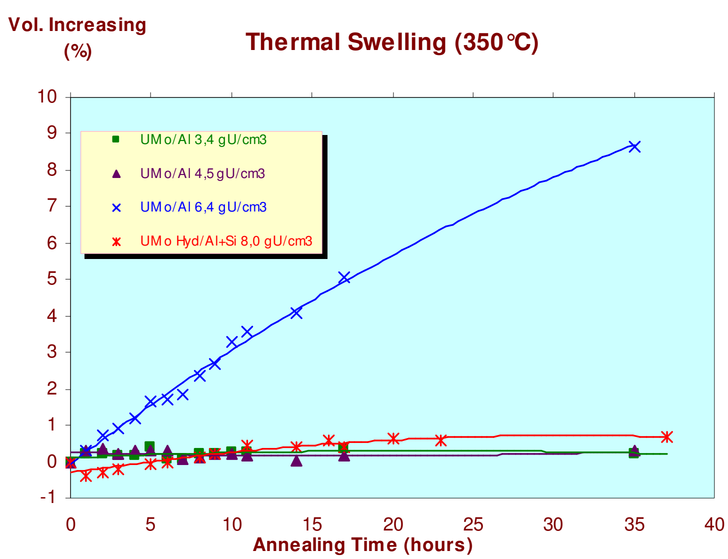


Figure 11. Out of pile thermal swelling applied to UMo-52 NU miniplate (red line)

Related to results of out-of-pile swelling test applied to UMo-52 NU miniplate, the behavior were so different compared with a group of miniplates previously tested at similar conditions (UMo-12 NU, ground powder – not SI added to matrix). The miniplate based on hydrided powder shows a trend to stabilization in terms of volume increasing and, if at the beginning a volume growth certainly occurs, after 20 hours of annealing, the volume variation, when compared with the 9% for UMo-12 miniplate, still remain below 1%.

With the analytical tools currently available in CCHEN it can be postulated that the use of hydrided powder dispersed in Al-Si matrix were not completely effective to avoid or delay the interaction phenomena, but an interesting consequence of this study could be the stabilization of volume increment during the propagation of fuel/matrix interaction. In order to complete this study, the integrity of cubic phase after all the transformations occurring during the fabrication process, and and even after approval of miniplates, must be clarified.

4. Conclusions

The use of hydrided UMo powder in dispersion fuel was not effective for avoid or delay the interaction phenomena. Nevertheless some beneficial effects in mitigation of volume increasing during processing were detected.

Considering the characteristics of powders, the manufacture parameters obtained in CCHEN, and the conditions and criteria for inspection and approval, is possible, in general terms, to meet the technical specifications for miniplates irradiation test using UMo hydrided powder.

The integrity of cubic gamma phase after fabrication processes is an issue still open for the use of hydrided UMo powders

5. Acknowledgements

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6. References

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