RERTR 2012 — 34th International Meeting on Reduced Enrichment for Research and Test Reactors

October 14-17, 2012 Warsaw Marriott Hotel Warsaw, Poland

Dispersion fuel miniplates based on UMo powder produced by centrifugal atomization

Luis Olivares, Jorge Marin, Mario Barrera, Carlos Gutierrez and Jaime Lisboa Nuclear Materials Department Chilean Commission for Nuclear Energy – CCHEN 95 Amunategui Street, Postal Code 6500687 Santiago — Chile

ABSTRACT

This paper describes results related to the manufacturing of dispersion type fuel miniplates using UMo powder obtained by means of centrifugal atomization using Rotating Electrode Process - REP. Atomization tests were performed at CCHEN with cylindrical pins made of U-7wt% Mo. The morphology of the particles, predominantly spherical, was revealed through Scanning Electron Microscopy. A batch of six miniplates with uranium densities of 6.0-7.0 and $8.0~{\rm gU/cm^3}$, was fabricated and inspected according to international specifications. Industrial radiography was applied to evaluate defects in the fuel core (meat), besides metrology and homogeneity in the distribution of UMo particles. Defects such as fishtail and stray particles, and thickening of the meat in the edges were observed in the highest density miniplates. Blister and bend tests were used to evaluate the meat/cladding bonding. Meat and cladding thickness was measured by metallographic inspections and SEM, and also for the inspection of the meat/cladding bonding and fuel/matrix IL formation.

1. Introduction

The International Fuel Development Group, integrated by Nuclear Institutes of about 10 countries, has been working in the development and qualification of high density fuel based on U-Mo alloys. CCHEN joined this group in 2009 and its research efforts have being focused in the development of dispersion type fuel up to 8.5 gU/cm³.

Centrifugal atomization has being implemented at CCHEN since 2010, based on the conceptual design of REP (Rotating Electrode Process), patented in USA by Starmet Corporation (formerly Nuclear Metal, Inc.) [1].

In this process, a pin of UMo alloy is melted and the liquid is projected by centrifugal forces in micro droplets, which solidify with solidification rates lower than gas or water atomization and in the order of 10³ to 10⁴ °C/s [2], and as a consequence, the particles produced are mostly spheroidals. Due to the aerodynamic forces involved, the movement of atomized droplets is radial, and they move and solidify separately, reducing the contact, collision and coalescence of two or more droplets in attached irregular forms [3]. Since in the REP process, the atomization is produced mainly by centrifugal ejecting forces instead of aerodynamic carrying, the powder obtained, when compared with gas atomized powder, is essentially free of porosity. Also, as this process does not use crucibles, the contamination level of the products is very low [3]. According to the open bibliography and the atomization tests carried out at CCHEN, some mechanical constraints can limit the rotational speed, and in consequence, can affect the medium size of particles [4].

2. Experimental Set-Up

2.1 Powder production by means of Rotating Electrode Process

The details of the centrifugal atomization system and its process were presented in a previous paper of this same group [5].

2.2 Characterization of atomized UMo powder

Granulometry and size distribution were obtained by means of conventional sieving, using four meshes of the Tyler series with aperture sizes of 150, 90, 63 and 45 µm. The classification and storing of the particles was done inside glove boxes under nitrogen atmosphere. Some UMo particles were properly conditioned for metallographic examination in order to study by SEM its morphology, micro constituents, the composition profile, the surface condition and the oxide layer formation, including punctual and linescan EDS(Energy-dispersive X-ray spectroscopy) analyses.

2.3 Miniplates Manufacturing

The miniplates were manufactured including blending of UMo + Al-Si powders, compacting, assembling (compact, covers and frame), welding, hot rolling, blister test, cold rolling and QA inspections. In table 1 is included a summary of fabrication parameters for the complete batch of 6 miniplates.

Table 1. Manufacturing parameters for a batch of six dispersion fuel type miniplates based on atomized UMo powder

Miniplate Identification		UMo-84	UMo-85	UMo-86	UMo-87	UMo-88	UMo-89
Uranium Type		Natural	Natural	Natural	Natural	Natural	Natural
Uranium density gU/cm ³		6,0	6,0	7,0	7,0	8,0	8,0
Fuel UMo Weight [g]		5,93	5,93	6,18	6,18	6,40	6,4
Matrix material weight (Al+4 wt% Si) [g]		1,52	1,52	1,27	1,27	1,05	1,05
Compacting pressure [Tons.]		22	22	22	22	22	22
Volume of meat [cm ³]		1,12	1,13	1,05	1,05	0,99	1,00
Fuel volume fraction UMo		0,42	0,42	0,47	0,47	0,52	0,51
Matrix volume fraction Al-Si		0,50	0,50	0,45	0,45	0,39	0,39
Meat porosity (calculated) [%]		8,0	8,0	8,0	8,0	9,1	10,0
Miniplates	Length (mm)	129,49	130,22	130,92	131,04	130,33	129,64
measurements	Wide (mm)	50,29	50,00	50,03	50,39	50,13	50,42
(preliminary cutting)	Thickness (mm)	1,45	1,44	1,41	1,43	1,39	1,41
Starting thickness of assembly [mm]		5,8	5,84	5,67	5,64	5,48	5,52
Total Reduction (%)		75,0	75,3	75,1	74,6	74,6	74,5
Reduction rate		1:4,00	1:4,06	1:4,02	1:3,94	1:3,94	1:3,91

2.4 Miniplates characterization

After completing the hot rolling process, the miniplates were visually inspected after blister test annealing. Meat metrology, inspection of manufacturing defects (stray particles, white points) and homogeneity were done using radiography. The meat/cladding bonding quality was verified by means of bend tests and the cladding thickness was measured by Eddy current technique. Finally, cross sections taken from miniplate UMo-88 NU were used for metallographic inspection and direct measurements of cladding – meat thickness.

3. Results and Discussions

3.1 Production and characterization of atomized UMo powder.

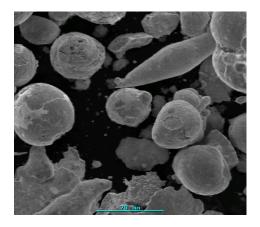
Details of the U-7% Mo pins preparation, atomization and characterization of powder were presented in a previous paper [5].

Table 2 is a summary of the results for atomization tests carried out on 5 UMo pins with different pin diameter. Rotating speed was fixed and according to the results obtained, the amount and granulometry of powders were most favourable for pins 3 and 4, both with higher fractions of finer particles. Starting from a total amount of 185.9 g of UMo alloy (5 pins), the weight of UMo powder produced was 140,3 g.

	Pin N°	Pin Weight [g]	Uranium Type	Pin diameter [mm]	Rotational Speed [RPM]	Intensity [A]	Powder produced [g]		Material <150 μm [%]
	1	35.7	LEU	6	36000	80	25.6	10.7	75.1
	2	39.6	NU	9	36000	60	30.9	11.1	65,2
	3	50.7	NU	10	36000	80	43.8	26.7	57,4
	4	36.8	NU	8	36000	80	26.7	19.2	48,9
ĺ	5	23.1	NILI	Q	36000	40	13.3	11.6	61.6

Table 2.- Summary of results of centrifugal atomization of UMo pins

Figure 1 shows UMo particles with spherical and irregular morphology. In this micrograph is possible to observe the surface layer, composed mainly by uranium oxide, brittle and weakly adhered to particle (some areas are broken). Figure 2 presents the cross section of an UMo particle. EDS analysis revealed a profile in the Mo content, very low amount of UMo was detected at the surface and practically the nominal composition at the center of the particle.



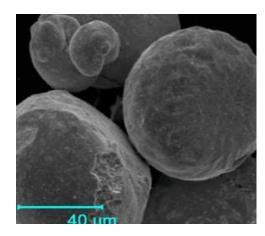


Figure 1. SEM micrographs of atomized UMo particles (pin N° 4).

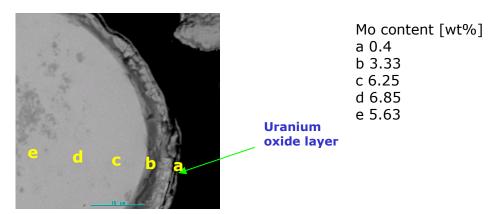


Figure 2. Details of uranium oxide layer formed at the surface of a particle. This layer is very thin, with a thickness of about $0.3 \mu m$, and brittle, i.e. cracks are present.

Previous to the miniplates fabrication, the atomized powder was sieved and its size distribution is shown in table 3.

Table 3.- Granulometry of UMo atomized powder proper for fuel miniplates manufacturing

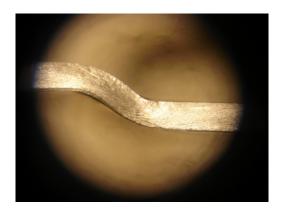
Mesh (μm)	Material below (g)	Material below (%)	
150	12,03	100,00	
90	12,73	78,41	
63	19,85	55,56	
45	11,10	19,92	
Total	55,71	_	

3.2 Miniplates Manufacturing

According to data included in table 1, the reduction rate was about 1:4, less than values usually applied to U_3Si_2 plates (1:6 -1:7). Thereby, the total reduction applied to UMo miniplates was in the order of 75%, lower than U_3Si_2 dispersion fuel plates made of, in which the total reduction is approximately 85%. Compared with miniplates made of hydrided powder, the reduction rate and total reduction are slightly higher than those used for atomized powder miniplates [4].

3.3 Miniplates Characterization

Blister test after hot rolling and bend test applied to samples taken after preliminary cutting, and also after final cutting of the miniplates, resulted in the approval of these controls for the six miniplates. An example is shown in Figure 3. Figure 4 shows some radiographs in which the meat measurements are according to specifications. Visual inspection reveals stray particles at the ends of miniplates with higher uranium density. For this reason, UMo-88 and UMo-89 miniplates were rejected after this inspection. The main cause for this defect may be related with the movement of spherical particles from the center to the ends of the meat during the rolling process. Nevertheless, the homogeneity inspection resulted according to specifications [6], either using visual inspection or using densitometry of radiographies where, in any point of meat, the readings were lower than 35% of nominal value for all miniplates.



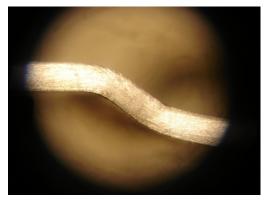


Figure 3. Bend test qualitative results corresponding to miniplates UMo-88 left; UMo-89 right.

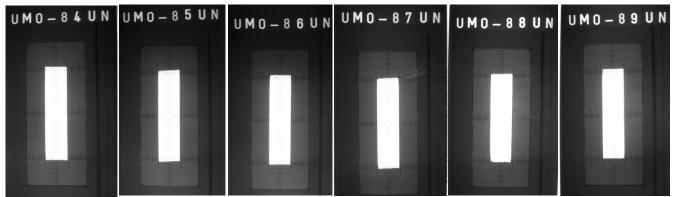


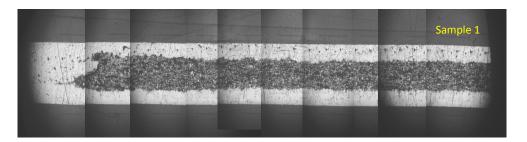
Figure 4.- Radiographies of miniplates manufactured with atomized UMo powder

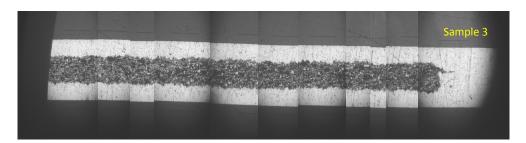
Table 4 contains cladding and meat thickness values measured directly on metallographic cross sections taken from UMo-88 miniplate (see Figure 5). Metallographic inspection revealed a slight thickening of the meat (dog-bone) in the central area of the miniplate (sample 2), where the thinning of cladding is not significant. The minimum cladding occurs at the ends of miniplates (0.25 mm for sample 1 and 0.34 mm for sample 3), both values are greater than the specified min clad [11]. Tail fish defect measures 1.60 and 0.62 mm for samples 1 and 3 respectively, usual values for UMo dispersion type fuel miniplates.

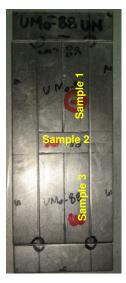
Table 4.- Meat and cladding thickness measured by metallography.

Sample	Minimum Cladding Upper [mm]	Min. Cladding thickness Lower [mm]	Average Meat [mm]	Tail Fish [mm]
1	0.30	0.25	0.73	1.60
2	0.32	0.328	0.68	
3	0.34	0.34	0.64	0.62

Optical micrographs of samples taken from UMo-88 NU miniplate included in the figure 5 revealed typical defects of dispersion type fuel, as fish tail at the ends (samples 1 and 3) and a slight thickening of meat (dog bone) at the edges of the central region of the miniplate.







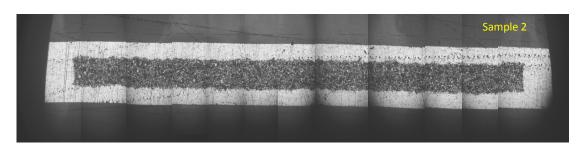
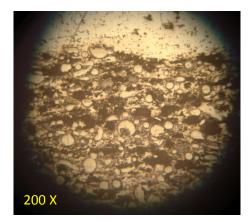


Figure 5.- Cross section and sampling photo of UMo-88 NU miniplate. Samples 1 and 3 are cross sections taken from ends of miniplate, along the meat. Sample 2 is a transverse cross section taken from the center of this miniplate. Optical micrographs - Magnification: 50X.



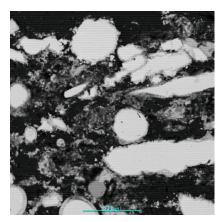


Figure 6. Micrographs of cross section of sample take from central area of UMo-88 miniplate. Low magnification optical microscopy at the left image, and SEM at the right image

The left image of figure 6 shows several UMo particles dispersed in an Al-Si matrix. The UMo particles dispersion exhibits an acceptable size distribution and a mix of irregular and spherical morphologies. All particles are dense and internal porosity is not observed. Some fuel particles exhibit evidences of IL formation, as shown in figures 6 and 7.

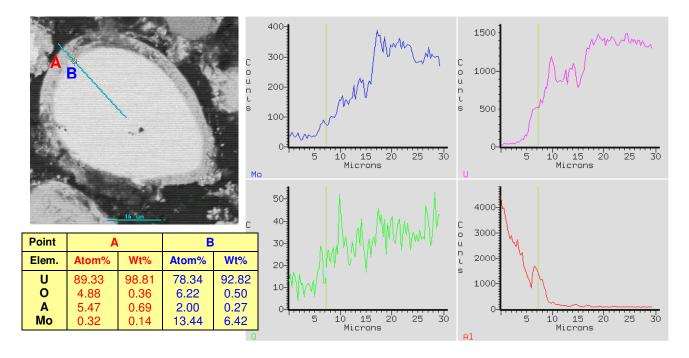


Figure 7. Energy-dispersive X-ray spectroscopy analyses. Linescan results shown evidence of migration of Al atoms from matrix towards the IL. The Mo and U content decreases from the center towards the surface of UMo particle and the oxygen content remains practically constant across whole particle. All these atomic migrations indicate that the fuel/matrix interaction was not avoided using atomized UMo powder. The UMo particle exhibits initially a very thin oxide layer, less than one micron, but after interaction with the matrix, the IL thickness increases from 5 to 10 microns. Results of point EDS showed an increment in U and Al content inside the IL, meanwhile the Mo content decreases dramatically.

4. Conclusions

In the UMo atomizing process, most of the particles produced have spherical or similar morphology. The pin diameter and intensity applied to pin were verified as process variables, besides the rotational speed and alloy properties.

According to the evaluated characteristics, such as particle size and distribution, morphology, crystalline phase and micro constituents, UMo powder produced at CCHEN by centrifugal atomization meets the specifications for dispersion type fuel fabrication.

The results of SEM and EDS analyses applied to atomized particles indicate the presence of a thin uranium oxide layer in its surface. Besides, UMo particles dispersed in Al-Si matrix reveal U and Mo concentration profiles, and that Al diffuses from the matrix towards the fuel particles. These interdiffusion mechanisms may generate IL around fuel particles. More information about

fuel/matrix interaction and IL formation and composition could be detected through SEM-EDS after out of pile swelling test. The miniplates fabricated with atomized UMo powder exhibit stray particles defects, typical for dispersion type fuel, and most likely due to the high uranium density and the movement of fuel particles towards the ends of meat, mainly those with spherical morphology. The meat of the miniplates reveals acceptable integrity and absence of cracks or excessive residual porosity.

5. Acknowledgements

The authors wish to express their thanks and acknowledgments the INL Fuel Development staff by the collaboration with the design and valuable information at the beginning of the development of CCHEN's system for centrifugal atomization.

6. References

- [1] U.S. Patent 3,099,041, July, 1963., U.S. Patent 3,802,816, April, 1974.
- [2] C. L. Tribus, J. D. Verhoeven, F. A. Schmidt and W. A. Spitzig "Use of the rotating electrode process in the fabrication of Cu-Nb Composites"; Journal of Materials Science Letters, Vol. 7 (1988), p. 532-534.
- [3] Steven A. Miller, Starmet Corporation, and Peter R. Roberts, American Superconductor From: ASM Metals Handbook, Volumen 7, Powder Metal Technologies and Applications (ASM International), Published in 1990, pages: 97-101.
- [4] Luis Olivares, Jorge Marin, Mario Barrera, Carlos Gutierrez and Jaime Lisboa, "Status and Progress of R&D in High Density Nuclear Fuel at the CCHEN" RERTR 2011 33rd International Meeting on Reduced Enrichment for Research and Test Reactors, October 23-27, 2011, Santiago, Chile.
- [5] J. Marin L. Olivares, M. Barrera J. Espinoza y J. Lisboa, Fabricación de polvos de aleación U-7Mo por atomización centrifuga. 11º Congreso Binacional de Metalurgia y Materiales, SAM / CONAMET 2011, 18 to 21 October, 2011 Rosario, Argentina.
- [6] Specification for Experimental Plates for the RERTR-11A and RERTR-ALT Campaigns. Document ID: SPC-1308, Revision ID: 1, Effective Date: 05/09/2012.