RERTR 2014 – 35TH INTERNATIONAL MEETING ON REDUCED ENRICHMENT FOR RESEARCH AND TEST REACTORS

OCTOBER 12-16, 2014
IAEA VIENNA INTERNATIONAL CENTER
VIENNA, AUSTRIA

Fabrication Procedures for Manufacturing UMo-Al Dispersion Fuel at IPEN

M. Durazzo, R. M. Leal Neto and J. R. O. Marques
Nuclear Fuel Center
Nuclear and Energy Research Institute IPEN-CNEN/SP, Av. Prof. Lineu Prestes 2242
05508-000 São Paulo – Brazil

ABSTRACT

This work forms part of the studies presently ongoing at IPEN investigating the feasibility of fabricating U10wt%Mo-Al dispersion fuel. Miniplates were fabricated with UMo alloy powder prepared by hydriding-milling-dehydriding of the gamma phase (HMD). Hydrided pieces were fragile enough to be hand milled to the desired particle size range. Hydrogen was removed by heating the samples under vacuum. Based on IPEN previous experience in developing and manufacturing U_3Si_2-Al dispersion type fuel, the objective of this work was starting the studies to promote an adjustment of the current fuel manufacturing procedures, allowing the incorporation of higher uranium concentrations to the fuel. The goal is to increase the uranium concentration up to around 7 gU/cm^3 by using the UMo-Al dispersion. This paper describes the main procedures used for manufacturing UMo-Al miniplates.

1. Introduction

For the last 30 years, high uranium density dispersion fuels have been developed in order to achieve the low enrichment goals of the Reduced Enrichment for Research and Test Reactors (RERTR) Program. Gamma U-Mo alloys, particularly those with 7 to 10 wt% Mo, as a fuel phase dispersed within an aluminum matrix have shown good results regarding their performance in irradiation tests [1,2]. These results have encouraged us to consider the use of this alloy in the nuclear fuel for future research reactors in Brazil.

Powdering U-Mo alloys is a major concern when fuel fabrication is based on the dispersion technology. There are at least three main process routes by which ductile (and tough) gamma U-Mo alloys can be powdered: atomization (mainly centrifugal atomization) by the rotating disk method [3] or the rotating electrode process [4]; mechanical comminution, i.e. machining or grinding [5,6]; and chemical comminution, i.e. the hydriding-dehydriding process, also known by its acronym HDH or HD [7-12], or as HMD when a milling operation is

performed between these treatments.

HMD combines hydriding-dehydriding with a milling process. Gamma U-Mo alloys are directly hydrided and powder can be produced by interposing a milling operation before dehydriding. Pasqualini *et al.* [10] reported that after heating the material to 700°C (1 h), it was able to absorb hydrogen during cooling at a temperature range between 190 and 50°C. The authors argued that beta U-Mo hydride (A-15 structure) is formed causing embrittlement of the alloy. Chen *et al*. verified that after a heating to 600°C under vacuum, alloy samples showed increased absorption of hydrogen at room temperature during successive hydriding-dehydriding cycles, until saturation was achieved after 8 cycles of hydriding at room temperature and 0.5 MPa dehydriding at 600°C under vacuum [11]. Moreover, Yi-Fu *et al*. demonstrated that activation could be obtained by careful chemical cleaning of the alloy surface followed by heating to only 250°C under vacuum, after which samples could be hydrided at room temperature [12].

In this investigation, we used powdered U-10wt%Mo alloy (lab scale) by hydriding the gamma phase after a thermal activation process. Once activated, the samples were able to absorb hydrogen. The resulting material was fragile enough to be manually milled. The powder was used to manufacture UMo-Al dispersion miniplates with uranium loading of around 6.7 gU/cm^3. Details of the miniplates manufacturing are provided elsewhere.

2. Powder Preparation

Ingots of U-Mo alloy with 10 wt% Mo were induction melted into a magnesia-stabilized zirconia crucible. Metallic uranium and metallic molybdenum were used as raw materials. Metallic uranium was home-produced by magnesiothermic reduction [13,14] and the metallic molybdenum cylinders produced had 99.95% purity and measured 3 mm in diameter and 3 mm in height. Both materials were charged inside the zirconia crucible and heated by induction under a high-purity argon atmosphere up to 1700^{o}C. The melting temperature was maintained for 2 min providing homogenization, after which the liquid alloy was poured in a high purity graphite mold. The solid material was a cylindrical piece measuring close to 20 mm in diameter by 150 mm in height, weighing around 800 g with a density of 16.87 g/cm^3.

Pieces taken from the U-Mo ingot were cleaned in a solution of nitric acid diluted in water, followed by washing in water and then in anhydrous ethanol. As-cleaned samples were immediately mounted inside the hydrogen reactor and evacuated at 5 x 10^{-3} mbar. Hydrogen flow was monitored by a digital flowmeter, together with sample temperature and time. The pressure inside the hydrogen reactor was monitored by a digital pressure gauge. Data were stored in a computer. After connecting the reactor to the apparatus, it was purged with argon and hydrogen in sequence. Sample temperature was monitored by a thermocouple inside the reactor, located just above the sample holder.

Activation consisted of heating the sample under pressurized hydrogen (2 bar) at 15°C/min up to 700°C for 1 h. Cooling was achieved by removing the reactor from the furnace and assisted with forced air cooling (fan). After reaching 300^{o}C the hydrogen pressure was increased to 8 bar and cooling continued to room temperature under hydrogen pressure. The hydrogen pressure was maintained for 24 hours.

The hydrogen reactor was transferred to a glove-box and opened under argon atmosphere. Several intermediary sieving operations help to control the fines generation. The

sieve set comprises a sieve with 150 μm opening, a fine sieve with 44 μm opening and a background compartment. The material was sieved to be classified in the size ranges between 150 μm and 44 μm and less than 44 μm. The fines content was 30 wt%.

After grinding, the hydride powder was charged in the reactor inside the glove-box and the reactor was connected to the apparatus. The reactor was evacuated (5 x 10^{-3} mbar), heated up to 700 ^{o}C at 15 ^{o}C/min and maintained at this temperature for 1 h, which was enough to complete the hydrogen desorption.

The hydriding process was not completed after a cycle of 24 hours under 8 bar hydrogen pressure. Only about half of the material could be ground after the first hydriding cycle. Three hydriding cycles were necessary for complete grinding of the hydrided material. Figure 1 shows the result obtained in the first hydriding cycle. After increasing the hydrogen pressure to 8 bar, it was noted that abrupt hydrogen absorption peaks occurred in which hydrogen absorption was recorded over 20 L/min in few seconds. In some cases, abrupt hydrogen absorption peaks occurred after 10 hours. This suggests that cracks occur in the solid piece randomly during hydriding, which causes the absorption peaks recorded. It was also observed that two absorption peaks occur in sequence (doublet). Additional studies are needed to better understand the hydriding mechanism and the variables that influence the process in order to develop a reproducible hydring process.

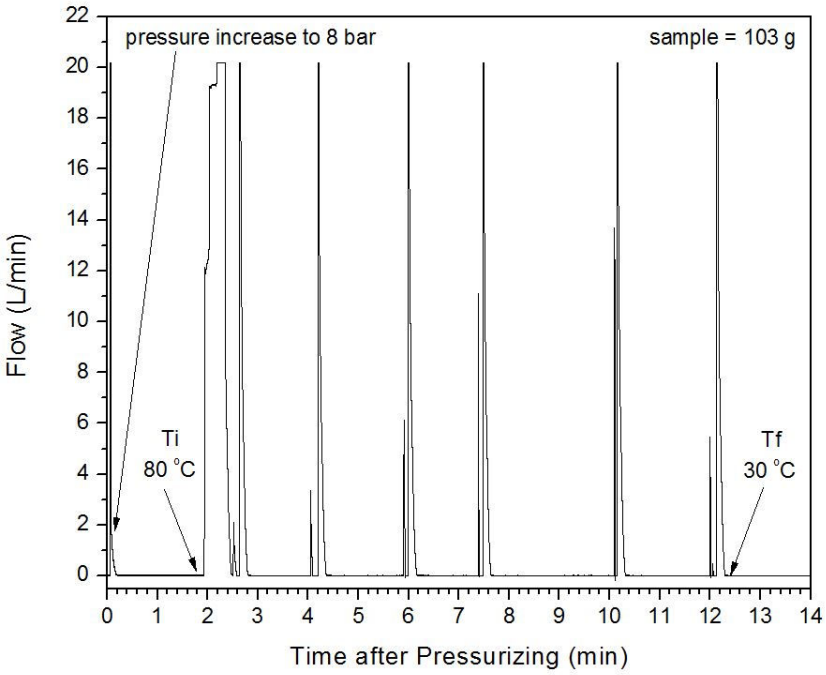

Figure 1. Flow of hydrogen recorded during the hydriding process.

X-ray diffraction was conducted on hydrided and dehydrided particles using Cu Kα radiation. Powder particles were also characterized by SEM analysis.

X-ray diffraction patterns from hydrided and dehydrided samples are presented in Figure 2. After hydriding, an amorphous-like pattern was observed, as well as the reflection of UO oxide phase. Gamma U-10wt%Mo phase reflections were not detected after hydriding, which

indicates a complete transformation following hydrogen absorption. After dehydriding, the gamma phase structure was observed. Oxide phases were also observed (UO_2 and UO). These observations confirm previous results, where a hypothesis for the amorphization of the hydrided alloy was proposed. The hypothesis is based on high strains developed during hydriding [15].

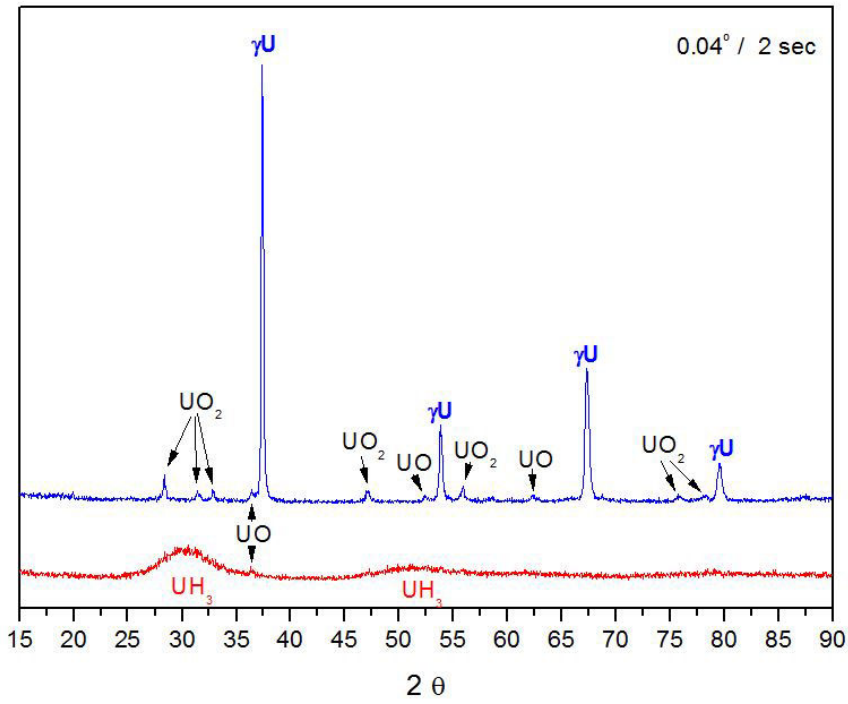

Figure 2. X-ray diffraction patterns of hydrided (red) and dehydrided (blue) samples.

Figure 3 shows some SEM micrographs took from hydrided and dehydrided powder particles (-100+150 mesh size range). Particle shape was very similar in both cases. Most particles are regular and equiaxial, but some of them exhibit a platelet shape. This could be the result of a preferential crack plan.

3. Miniplate Fabrication

A total of about 100 g UMo powder with particle size in the range between 150 and 44 μm was produced. This powder was used to manufacture 4 briquettes having a volume fraction of 45% UMo powder. The briquettes were produced using a pressing die with dimensions of 20 x 40 mm with rounded corners (radius 3 mm). The compaction pressure used was 5 tf/cm^2. Table 1 shows the characteristics of the briquettes produced. Figure 4 shows photographs of the pressing die and briquettes.

The briquettes were degassed at 250 °C for two hours under dynamic vacuum of 4 x 10^{-3} mbar. The miniplates were manufactured according to the procedures routinely adopted by IPEN in the fabrication of fuel plates. Aluminum alloy 6061 was used in the preparation of frames and covers. Two briquettes were assembled simultaneously on each frame plate, as illustrated in Figure 5 (left). The assemblies were welded and rolled according the typical rolling schedule used at IPEN (Table 2). Figure 5 (right) shows the procedure for rolling the UMo-Al dispersion miniplates.

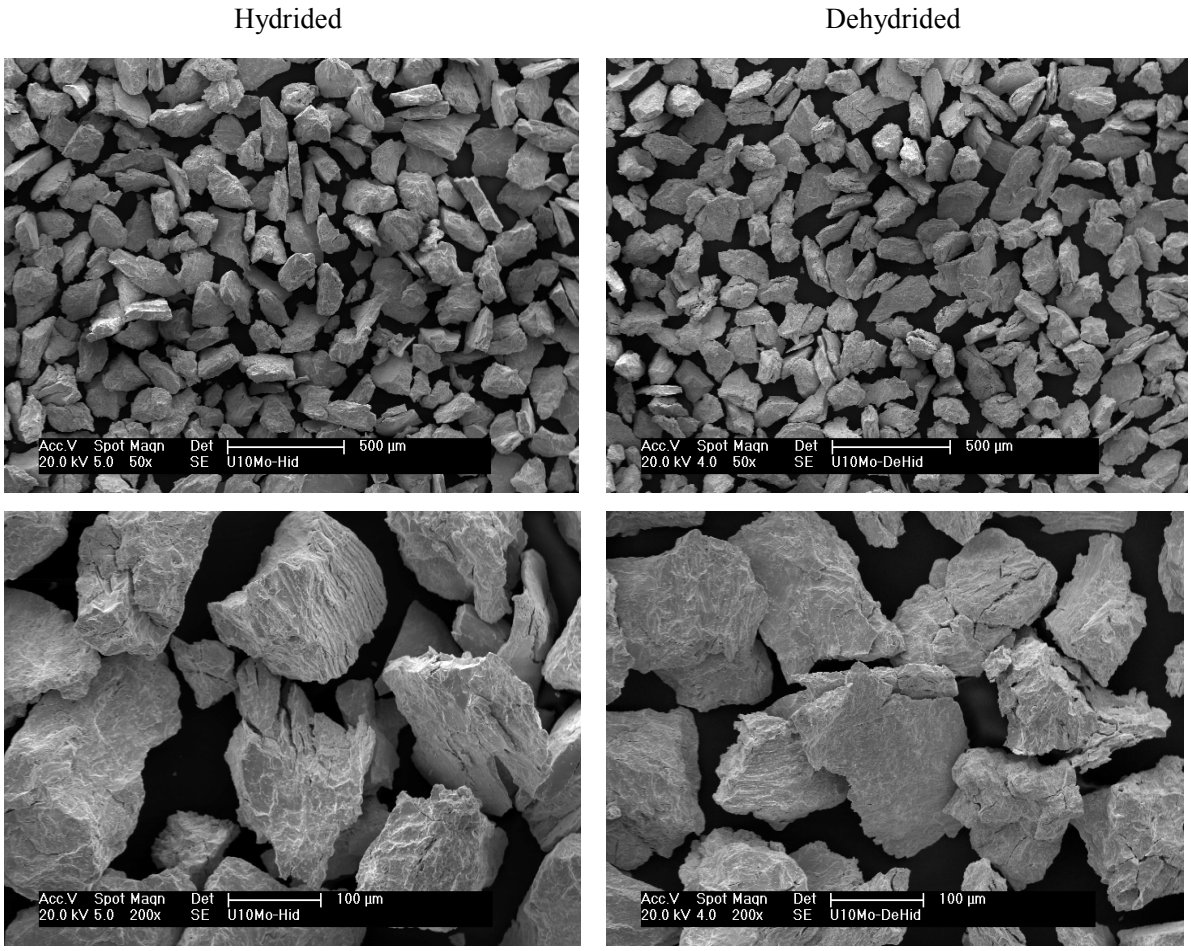

Figure 3. SEM micrographs (secondary electrons images) of hydrided and dehydrided U10wt%Mo powder particles.

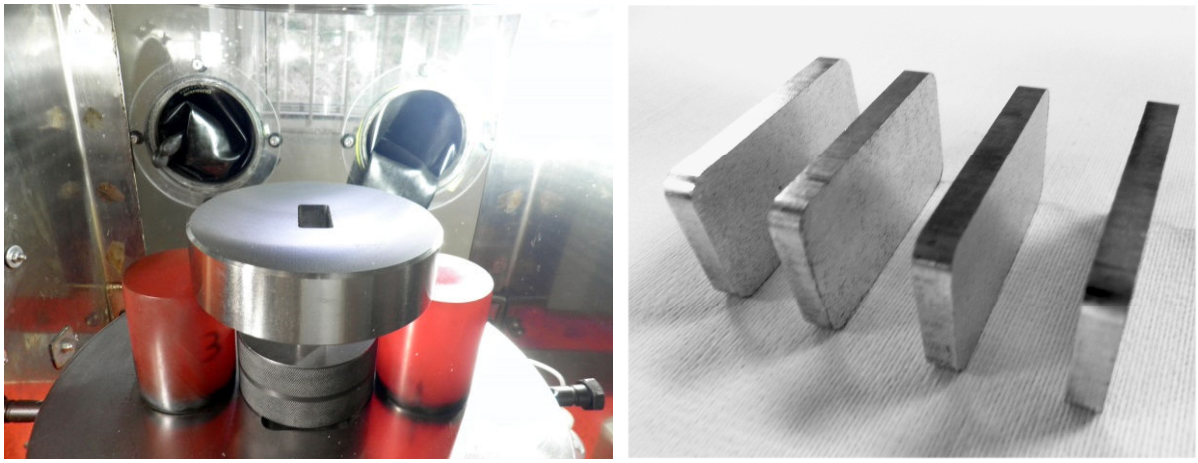

Figure 4. Photographs illustrating the pressing die (left) and the briquettes produced (right).

Table 1. Main characteristics of the briquettes produced.

	M_{briq}	M_{UMo}	M_{Al}	*Dimensions* (mm)			V_{briq}	V_{UM}	V_{Al}	VF_{UMo}	*Pores*	*UD*
	(g)	(g)	(g)	*L*	*W*	*T*	(cm^3)	(cm^3)	(cm^3)	(%)	(%)	(gU/cm^3)
UMo-01	28.79	24.65	4.24	20.05	40.11	4.06	3.265	1.456	1.565	44.59	7.48	6.80
UMo-02	28.84	24.64	4.24	20.05	40.09	4.15	3.336	1.455	1.565	43.62	9.47	6.65
UMo-03	28.80	24.64	4.23	20.04	40.06	4.10	3.291	1.455	1.565	44.21	8.36	6.74
UMo-04	28.80	24.64	4.23	20.04	40.06	4.13	3.316	1.455	1.565	43.88	9.04	6.69

U10Mo powder density=16.93 g/cm^3 (helium pycnometry)

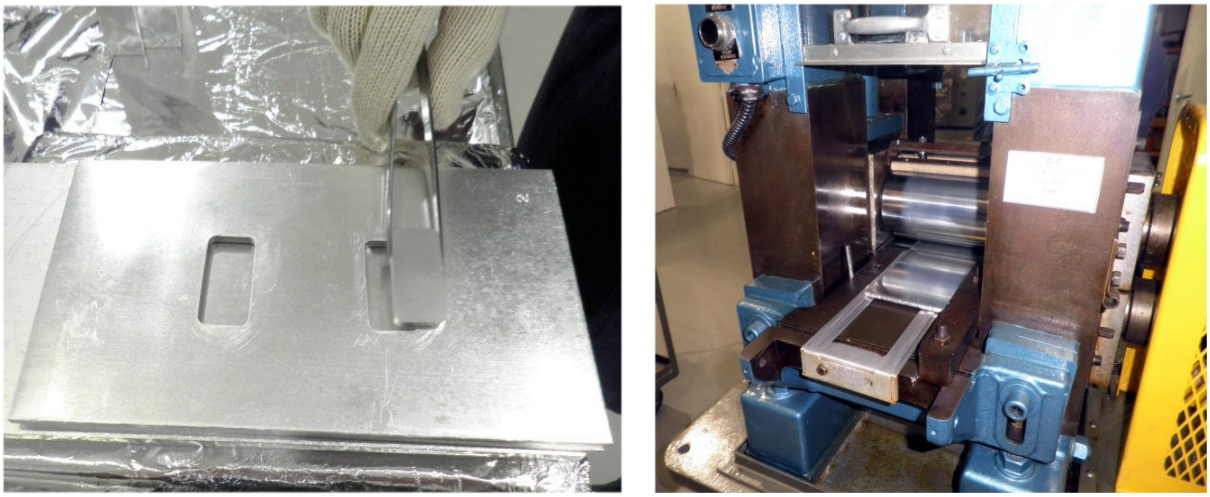

Figure 5. Photographs illustrating the assembling (left) and the rolling (right).

Table 2 – Typical rolling schedule.
hot-rolling (450 ^oC)

Pass	Reduction (%)	Gage (mm)	Heating Time (min)
0	0	9.20	preheating (60 min)
1	25	6.93	15
2	25	5.20	15
3	15	4.42	15
4	15	3.76	15
5	15	3.20	15
6	15	2.72	15
7	15	2.31	15
8	13	2.01	15
9	13	1.75	blistering test (60 min)

cold-rolling

Pass	Reduction (%)	Gage (mm)	Heating Time (min)
0	0	1.75	from hot-rolling
1	7	1.63	no heating
2	6-7	1.53 – 1.51	no heating
final	0	1.53 – 1.51	final adjustment

The rolled miniplates were pre-cut and X-rayed. Two radiographs were obtained at this stage, the first one to locate the miniplate meat and the other one to check the uranium distribution in the meat and the presence of cracks. Based on the location radiographic the final cut was performed. The resulting pieces of pre-cut were extensively inspected for bonding by bending test. Bonding was also inspected by blistering test. This test consisted in heating the miniplates to 450 °C for 1 hour before the final cold-rolling passes. After the test a visual inspection was performed to check for blistering. Table 3 presents the main characteristics of the meats of the fabricated miniplates. Figure 6 shows a typical radiography of the meat.

Table 3. Main characteristics of the meats.

	Thickness (mm)	***Mass of Uranium*** (g)	***Meat Dimensions*** (mm)		***Uranium Density*** (gU/cm^3)
			Length	***Width***	
UMo-01	1.54	22.19	117	42	6.65
UMo-02	1.54	22.18	116	42	6.72
UMo-03	1.54	22.18	117	42	6.68
UMo-04	1.54	22.18	116	42	6.70

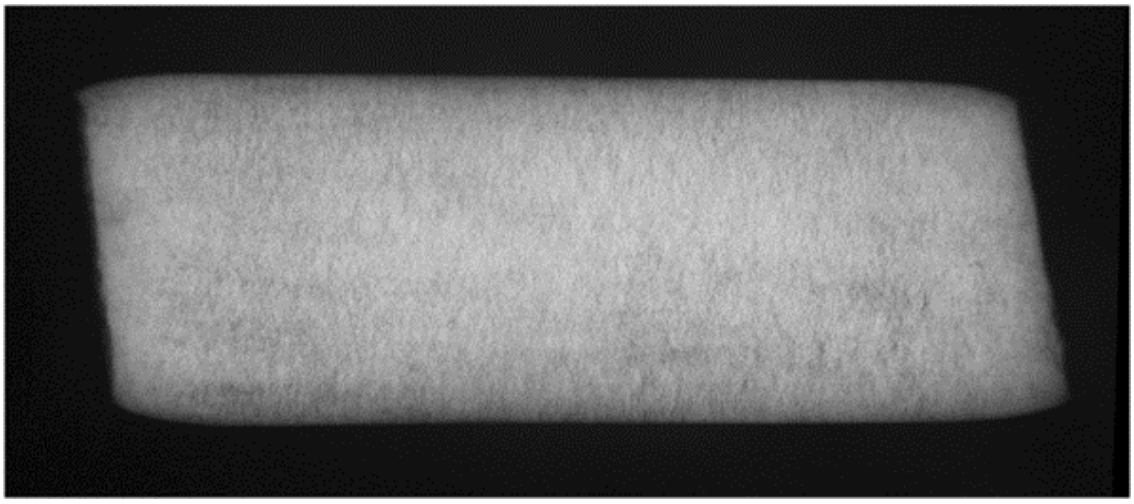

Figure 6. Radiography of the meat of the UMo-04 miniplate.

All manufactured miniplates showed good metallurgical bond. Blisters were not observed and no problems were detected in the bending tests. The integrity and the uranium homogeneity of the meat were considered satisfactory.

The microstructural appearance of the dispersion after rolling was also considered satisfactory, as illustrated in Figure 7. The minimum cladding thicknesses at the dog-boning were determined by metallography as 0.25 mm (top cladding) and 0.29 mm (under cladding). The cladding thickness at the center of the meat varied from 0.34 mm to 0.37 mm. Figure 8 shows a photograph of the fabricated UMo-Al miniplates.

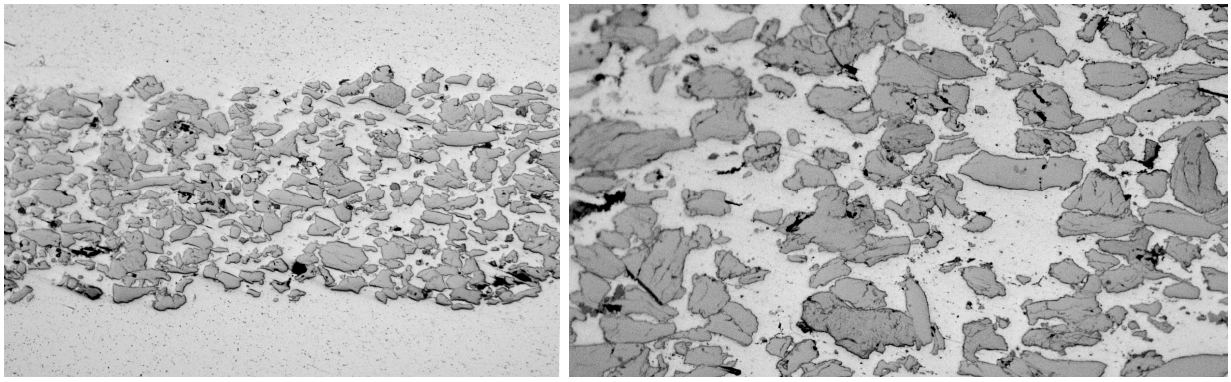

Figure 7. Typical meat microstructure of the UMo-Al dispersion miniplates.

Figure 8. Photography of the fabricated miniplates.

4. Conclusion

At this stage it was found that it is feasible to manufacture the fuel based in the UMo-Al dispersion at IPEN. Miniplates made from natural uranium demonstrated the feasibility of making miniplates UMo-Al to give the desired uranium concentration. Theoretically, the maximum uranium concentration that can be obtained with UMo alloy with 10 wt% of molybdenum is 6.9 gU/cm^3, assuming the theoretical density for the powder of 17 g/cm^3. The

uranium concentrations obtained in the miniplates developed in this paper were slightly lower, around 6.7 gU/cm^3, mainly due to the density of the powder produced in this work, which was determined to be 16.93 g/cm^3. Due to the very low UMo particle fragmentation, the particle size could be reduced to 100-90 μm, which probably would improve the microstructure of the dispersion. New manufacturing tests should be conducted to obtain a more suitable microstructure.

In this work the hydriding process did not showed to be reproducible and controllable for large charges. Additional studies are needed to better understand the hydriding mechanism and the variables that influence the process in order to develop a reproducible hydring procedure to process increased UMo alloy quantities (~1000g).

5. References

1. J.L. Snelgrove, G.L Hofman, M.K Meyer, C.L Trybus, T.C Wiencek, *Nuclear Engineering and Design* **178**(1) (1997) 119-126.
2. G.L. Hofman, J.L. Snelgrove, S.L. Hayes, M.K. Meyer, Progress in development of low-enriched U-Mo dispersion fuels: in 6th International Topical Meeting on Research Reactor Fuel Management (RRFM 2002), European Nuclear Society, Ghent, Belgium, April 21-25. pp. 50-58.
3. K.H. Kim, D.B. Lee, C.K. Kim, G.E. Hofman, K.W. Paik, *Journal of Nuclear Materials*, **245**(2-3) (1997) 179-184.
4. C.R. Clark, B.R. Muntifering, J.F. Jue, Production and characterization of atomized U-Mo powder by the rotating electrode process: in Proceedings of the 2007 International Meeting on Reduced Enrichment fot Research and Test Reactors, Prague, Czech Republic, September 23-27, 2007.
5. C.R. Clark, M.K. Meyer, Fuel powder production from ductile uranium alloys: in Proceedings of the 1998 International Meeting on Reduced Enrichment for Research and Test Reactors, São Paulo, Brazil, October 18-23,1998.
6. H.Vacelet, P. Sacristan, A. Languille,Y. Lavastre, M. Grasse, Irradiation of full size UMo plates: in Proceedings of the 1999 International Meeting on Reduced Enrichment for Research and Test Reactors, Budapest, Hungary, October 3-8, 1999. (available at http://www.rertr.anl.gov/Web1999/PDF/07vacelet99.pdf)
7. S. Balart, P. Bruzzoni, M. Granovsky, L. Gribaudo, J. Hermida, J. Ovejero, G. Rubiolo, E. Vicente, U-Mo alloy powder obtained by a hydride-dehydride process: in Proceedings of the 2000 International Meeting on Reduced Enrichment for Research and Test Reactors, Las Vegas, Nevada, October 1-6, 2000.
8. M. Solonin, A.V. Vatulin, Y.A. Stetsky, Y.I. Trifonov, B.D. Rogozkin, Development of the method of high density fuel comminution by hydride-dehydride processing: in Proceedings of the 2000 International Meeting on Reduced Enrichment for Research and Test Reactors, Las Vegas, Nevada, October 1-6, 2000.

9. L. Olivares, J. Marin, J. Lisboa, H. Pesenti. Powder production of uranium-molybdenum-metal alloys applying hydride-dehydride methodology: in Proceedings of the 2008 International Meeting on Reduced Enrichment for Research and Test Reactors, Washington, D.C.,October 5-9, 2008.
10. E.E. Pasqualini, J.H. Garcia, M. Lopez, E. Cabanillas, P. Adelfang, Powder Production of U-Mo Alloy by HMD Process.(Hydriding-Milling-Dehydriding): in 6th International Topical Meeting on Research Reactor Fuel Management (RRFM 2002), European Nuclear Society, Ghent, Belgium, April 21-25. pp. 183-187. (available at http://www.euronuclear.org/meetings/rrfm/pdf/RRFM%202002.pdf)
11. M. Chen, X. Yi-fu, W. Jing, J. Jia, P. Zhang, *Journal of Nuclear Materials*, **400**(1) (2010) 69-72.
12. X. Yi-Fu, M. Chen, W. Jing, C. Chen, *Fusion Engineering and Design*, **85**(7-9) (2010) 1492-1495.
13. M. Durazzo, A. Silva, J. de Souza, E. de Carvalho, H. Riella, Current status of U_3Si_2 fuel elements fabrication in Brazil: in Proceedings of the 2007 International Meeting on Reduced Enrichment fot Research and Test Reactors, Prague, Czech Republic, September 23-27, 2007.
14. A.M. Saliba Silva, I.C. Martins, L.R. dos Santos, D.G. da Silva, E.F. Urano de Carvalho, H.G. Riella, M. Durazzo, Metallic Uranium Production for Irradiation Targets Alloys: in 15th International Topical Meeting on Research Reactor Fuel Management (RRFM 2011), European Nuclear Society, Rome, Italy, March 20-24, 2011. pp. 232-240.
15. R.M. Leal Neto, C.J. Rocha, E.F.Urano de Carvalho, H.G. Riella, M. Durazzo, *Journal of Nuclear Materials*, **445** (2014) 218-223.