MANUFACTURING AND CHARACTERIZATION OF LEU DISPERSION MINIPLATES BASED ON HYDRIDED UMo POWDERS

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

ABSTRACT

As result of the local UMo Fuel Development Program, CCHEN has been producing U-Mo powders applying two hydriding methodologies. Starting from LEU alloys, the hydriding processes conducted at different temperature and duration produce particles with irregular morphology and real density values than differ in about 30%. This paper presents results of powder characterization and manufacture of high density dispersion fuel miniplates. Both powder types, porous and dense, were characterized using SEM for morphology and compositional micro analysis, particle size and distribution, phase composition (XRD), real density by helium pycnometry and residual contents of oxygen. For characterization of UMo/Al-Si miniplates made with natural uranium, were included dimensional control, blister and bending tests, homogeneity, ultrasonic scanning and dog bone, stray particles, minimum cladding evaluated by metallographic inspection. Finally, manufacturing processes details were defined for dispersion miniplates manufactured, inspected and evaluated according to RERTR technical specifications as part of Chilean collaborations to international qualification efforts.

1. Introduction

Recent publications related to post irradiation results for UMo dispersion type fuel [1] are in concordance with the information generated by the CCHEN as results of their program of development in this type of fuels [2]. These results confirm the hypothesis that small amounts of silicon added to the aluminum matrix allow, in some way, to stabilize the interaction layer between fuel particles and the Al matrix that surrounds them [3,7]. Since this fuel is in route to be qualified internationally [4], it is opportune the optimization of some processes of production. It is the case of the powder production technologies, where nevertheless the atomization methodology has shown comparative advantages [5], opportunities are visualized to try with alternative technologies as it could be the hydriding.

A brief bibliographical revision of atomization applied to UMo alloys reveals some details for comment; Due to the spherical morphology, the movement and segregation of the particles
of fuel in the Al matrix could occur during blending and rolling processes, limiting even more the rolling reduction rates, which already have been reduced as a way of controlling the fuel/matrix interaction activated by the rolling temperature [6]. On the other hand, a dispersion of UMo spherical particles in the aluminum matrix generates as-fabricated porosity levels up to five times lower than the values obtained using irregular morphologies [8], in consequence, it could limit the available space to contain fission gaseous products produced during the burn-up of the fuel in the reactor and, finally, to promote the occurrence of swelling of fuel plates. [8]

The hydriding methodology has been considered by several authors as an interesting alternative considering that it is not a complex process, besides the lower cost of the equipment [9], the simplicity [5] and the lower power consumption of systems used [10]. The authors consider that this process is an interesting option to produce powders of U-Mo with morphologies, particles size, crystalline phases and chemical composition capable to be used in nuclear fuels.

Taken into account all these antecedents, CCHEN has developed hydriding methodologies for production of powders of U-Mo alloys. The hydriding process considers two variants in the first stage, in which the dissolution of hydrogen into the alloy is promoted. In a first method, called in this paper HMD-Dense, the hydrogen dissolution take place in the region of stability of the cubic phase γ-U, over the γ-U → α-U + γ-U (800°C) transformation line in the U-Mo phase diagram, and then the formation of U-H compound is promoted through low temperature and long lasting thermal treatment. The other methodology tested consider the dissolution of hydrogen under the most favorable conditions for the γ-U → α-U + γ'-U phase transformation, according to the TTT diagram for this compositional range, due to this reason this process is considered as hydriding in α-U phase.

In this paper are reported and discussed the results obtained for production of powders using low enriched uranium-LEU, characterized through powders density measurements, crystalline phases identification by XRD, SEM for morphology and constitutional microanalysis (EDS) and size and distribution of particles. Finally, results of dispersion type miniplates production and its characterization are reported for nominal uranium densities of 8gU/cm$^3$ using both types of powders. These miniplates was done with natural uranium-NU in order to setting up the inspection and evaluation techniques. The effect of each powder type was evaluated on the base of calculation of real uranium density in the meat and also were discussed results of non destructive analyses and metallographic characterization of the miniplates.

2. Experimental Sep-Up

The used alloy, with nominal composition of 7wt% of Mo was obtained by melting and pouring at inert atmosphere by means an induction furnace, using LEU with 19.75 wt% $^{235}$U and metallic molybdenum with 99.5% of purity. Due to the high pyrophoricity exhibited by the hydrided products, it became necessary to keep always protective atmosphere conditions (hydrogen, vacuum or argon). For this purpose a glove box was adapted to the furnace chamber in order to maintain inert atmosphere (Ar) during the opening of the chamber and
the extraction of the samples, which were stored in hermetic containers and transferred immediately to glove box with nitrogen based atmosphere and oxygen level under 5%vol.

Figure 1. Thermal cycles applied for UMo alloys treatments. Redline represents hydriding in γ-U phase and blue line corresponding to hydriding in α-U phase

Details of the hydriding - dehydriding thermal treatments are included in the Figure 1. Prior to apply the hydriding thermal treatments, the alloy pieces were cleaned and placed into double alumina crucibles to put them into the furnace chamber. The powders obtained were sieved under 150 μm and the oversize was subjected to mechanical grinding using a rotating knives mill (titanium blades). The characterization of the powders considered analysis of real density for helium pycnometry, SEM for verification of morphology of particles and constitutional microanalysis (EDS), size and distribution of particles and measuring of remaining oxygen content. The XRD analyses were carried out to samples prepared dispersing UMo powder in epoxy resin, grinding and polishing to avoid the interference of the superficial oxide layer. With other hydrided powders manufactured in a similar way but using natural uranium-NU, miniplates with nominal uranium density of 8 gU/cm³ was fabricated, using production techniques know for dispersion fuel, as blending of powders, compacting, encapsulated in 6061 aluminum cladding, TIG welding, hot and cold rolling, limiting the reduction rates to values lower than 1:4. The NU-UMo powder was dispersed in an aluminum matrix prepared with 100% < 90 μm and 80% < 45 μm, blended with 4wt% of fine silicon powder (100% < 15 μm). The behavior of each powder type was evaluated considering their effect in the final density of the meat. In the next section are presented results of blister test, meat metrology, density, homogeneity in the distribution of the fuel phase by means X Ray inspection, bending test, metallography and bonding test through ultrasonic scanning.
3. Results and Discussion

The figure 2 shown as cast microstructure for the LEU-UMo alloy. A two phase microstructure was detected; a massive phase in light blue tone constituted for γ-U and a second phase, light green, dispersed and with globular aspect, which according to EDS analyses, could be the α-U phase. Also are present some inclusions, probably oxides or other impurities and was not possible to observe important segregation of Mo. EDS microanalyses shown uranium and Mo in average contents of 6.5 wt% in light blue region and 7.6 wt% for green zone. The XRD pattern show the majority presence of the phase γ-U, while the uranium oxides are in small amount and α-U phase was not detected, probably for their very small quantity.

The transformation of small pieces into powder take place directly during the thermal treatments in atmosphere of hydrogen, in each case applied to not homogenized samples and therefore, these samples probably contents small amounts of phase α-U in its microstructure. The application of a sequence of thermal treatments of hydriding, dehydriding, homogenization and quenching allow to recover the cubic phase and to reduce at minimum the presence of orthorhombic α-U phase, being obtained powders with contents of oxygen of 1.4 wt%, several times lower than the powder on surface (6.7 wt%). According to the XRD analyses of powders (Figure 4), the presence of oxygen in these powders would be preferably in form of UO$_2$. The oxidizes layer in the surface of crucible had a characteristic color and was easily removable for analyses, meanwhile, the powder produced under this layer shown metallized dark grey tone. In relation to the hydrogen contents in the powders, the analyses revealed high content after the hydriding stage (774 ppm), while after the quenching, the remaining hydrogen was only 58 ppm, slightly higher to the 41 ppm present in the as-cast alloy.
In the figure 3 is possible to observe the differences in the morphology of the particles obtained by both hydriding methodologies. The left image corresponding to the product of hydriding in phase γ-U, these particles have an aspect compact while the micrograph at the right, product of hydriding in phase α-U, shown abundance of micro cracks and interconnected porosity.

![Figure 3 SEM micrographs of hydrided LEU-UMo powders: (a) dense and (b) porous.](image)

The differences in morphology are in accordance with the density of the powders whose results are 16.15 g/cm³ for the HMD-dense product (left) and only 11.45 g/cm³ for powders obtained by HD-porous methodology (right). Figure 5 shown particles size and distribution, where dense powder appears finer than porous powder.

![Figure 4 XRD patterns for LEU-UMo hydrided powders dense (a) and porous (b)](image)
The table 1 summarizes the results for miniplates made of NU-hydrided powders. From the radiographs of figure 7, is possible to deduce, in general terms, homogeneous dispersion of UMo particles in the Al-Si matrix. The results of blister test, carried out at 480°C/40 minutes, controlled after cold rolling were satisfactory. The volume increase before and after blister test were 1.7% for UMo-47 (dense) and 0.1 % for UMo-48 (porous). As seen on Figure 6, the ultrasonic scanning test revealed complete and homogeneous bonding in the cladding/meat interfaces with some irregularities in edge and ends of meat.
Figure 6 Ultrasonic scanning analysis for bonding test for NU-UMo/Al-Si miniplates
(a) UMo-47 and (b) UMo-48

Figure 7 Radiographic inspections of NU-UMo/Al-Si miniplates

From the figure 8, the results for bending test also showed satisfactory results for all samples extracted from outer zone surrounded the 130mm x 50 mm miniplate area. The meat thickness calculation based on the x-rays exhibit slightly smaller values that those measured by means of metallography. This difference is due, probably, to the presence of edge defects in meat which appear wider than real dimensions. These defects are shown in optical micrographs at right of Figures 9 and 10.
In general, for the UMo miniplates the reduction rate by rolling decreases until a minimum of 1: 3.66 – greatly lower than 1:5 - 1:7, usual values for silicides based fuel. Likewise, the homogeneity of the extreme area of the miniplates appears affected by the high uranium density. This fact could require modifying of the criteria for acceptance and rejection of fuel plates for these defects.
Figure 10  SEM micrographs of transverse cut for UMo-46 miniplate (porous powder). At right, optical micrographs of meat edges

The metallographic inspection allowed the measurements of the Al cladding and to verify the typical defects produced in dispersion fuel plates like dogbone, some stray particles, minimum cladding thickness near to the end of miniplates. Anyway, these min clad values were always thicker than the specified minimum acceptable. The particles of porous powder shown some evidences of fragmentation produced during rolling.

4. Conclusions

The density measured for the U-7Mo alloy was 17.45 g/cm$^3$. After convert this alloy in powders through hydriding, the real density decreased at 16.15 g/cm$^3$ and 11.45 g/cm$^3$ for dense and porous powders respectively. This last value is nearer to the uranium silicides density, then reasonable doubts appears in reaching, with hydrided powders, the values of uranium density expected for UMo. Anyway, the hydriding processes, made to low or high temperature, constitutes an alternative for UMo powder production. In the case of LEU UMo powder, it present as majority phase the cubic structure γ-U and a little, although not quantified amount of orthorhombic phase α-U.

Was possible to produce miniplates with hydrided powders until nominal density of 8gU/cm$^3$ with acceptable level of defects. The homogeneity, the min clad, the presence of remote islands, stray particles and residual porosity in the meat were defects that seem enhanced in UMo miniplates in comparison with silicides fuel, taken into account, in any case, that it doesn't seem feasible this level of uranium density using silicides.
As consequence of the results reported in this paper, the immediate following activity will be to manufacture miniplates using LEU dense and porous powders. These miniplates will be prepared and inspected with the purpose of irradiating them like part of RERTR experiments.

5. Acknowledgements

The authors are grateful for the support received from the IAEA and from the Chilean Commission for Nuclear, especially from staff members of CCHEN’s Fuel Element Plant – PEC.

6. References