

# U-Mo alloy powder obtained by a hydride-dehydride process

**Balart S., Bruzzoni P., Granovsky M., Gribaudo L., Hermida J.,  
Ovejero J., Rubiolo G., Vicente E.**

Departamento Materiales – CAC  
Comisión Nacional de Energía Atómica  
Avda. Gral. Paz 1499 – 1650 – San Martín - ARGENTINA –

## Abstract

The use of uranium-molybdenum alloys as a component in high-density LEU dispersion fuels is very promising. However, these alloys need to be transformed to powder due to the manufacturing requirements of the fuels.

One of the possible methods to accomplish this is the hydride-dehydride process. It is based on the ability of the U- $\alpha$  phase to transform to  $\text{UH}_3$ : a brittle and relatively low-density compound.

A powder was obtained applying this method which is described in this work.

U-Mo alloys around 7 wt % Mo were heat treated at different temperature ranges in order to partially convert  $\gamma$ -phase to  $\alpha$ -phase. Subsequent hydriding transforms this  $\alpha$ -phase to  $\text{UH}_3$ . The volume change associated to the hydride formation embrittles the material which ends up as a powdered alloy.

Results of the optical metallography, scanning electron microscopy, X-ray diffraction during different steps of the process are shown.

## Introduction

New technologies are been developing in order to obtain compounds with higher uranium concentrations for fuel components used in experimental and radioisotope production nuclear reactors [1].

The use of uranium-molybdenum alloys, as a component in high-density low enriched uranium (LEU) dispersion fuels is very promising.

Addition of Mo to pure U allows the metastable retention of the high temperature stable  $\gamma$ -phase (body centered cubic) which offers good properties in fuel elements.

These alloys need to be transformed to powder due to the manufacturing requirements of the fuels.

The  $\gamma$ -phase U-Mo alloys exhibit typical metallic ductility and cannot be easily reduced to powder by grinding, as is the case of ceramic compounds or many intermetallic compounds.

Different processes were suggested in order to obtain U-Mo powder [2]. The hydride-dehydride operation is one of them and it is based on the property of pure U in the  $\alpha$  (orthorhombic) structure to form  $\text{UH}_3$  under hydrogen atmosphere. The volume change associated to the hydride formation embrittles the material which ends up as a powdered alloy.

Taking into account this behavior it was proposed [3] that alloys between 5 and 8 wt % Mo were heat-treated at convenient temperatures in order to obtain a two-phase material including the  $\alpha$ -phase. After the hydride process this phase would be transformed to  $\text{UH}_3$  and the powder could be obtained directly as a result of the cracked structure. Moreover if the obtained  $\alpha$ -phase is only present at grain boundaries, that is a 'decorated' zone (rimmed microstructure), the hydriding

process could lead to the intergranular cracking of the material. After a dehydride process, a new heat treatment will be necessary to transform the remaining microstructure back to the original  $\gamma$ -phase.

This work is related to the description of a 'laboratory scale' process, which allows obtaining powder of the U-Mo (7 and 8 wt % Mo) alloy in the  $\gamma$  structure by the hydride-dehydride method.

## Experimental

Uranium 99.6% pure and Mo 99.9% pure were used as starting materials.

The component metals, in batches of approximately 70 g, were melted in an arc furnace under argon pressure, using a non-consumable tungsten electrode and water-cooled copper hearth.

The button-shaped ingots were turned over and remelted up to four times to ensure homogeneity.

Thermal treatments were performed in sealed evacuated silica tubes with the samples individually wrapped in tantalum foils.

The gas-phase hydriding technique was used in the experiments. Hydrogen was obtained from an electrolytic generator with a maximum of 10 ppm impurities and argon was high purity type.

The hydriding reactor (chamber), as shown in Figure 1, is a closed vertical silica tube. The samples are placed at the bottom of this tube in a sample holder. Gases enter the chamber by a central tube with the opening close to the sample and leave it by the upper end. The chamber can be lowered into a furnace.

Once the sample is set in place the chamber is filled with  $H_2$  at about 1 atm, temperature is raised up to the working range, typically around 250 °C. The amount of hydrogen absorbed by the sample is monitored. The process is stopped when absorption reaches a plateau indicating that the sample has taken up all the possible H. The chamber is then removed from the furnace and the samples cooled. Dehydride is achieved by introducing again the chamber into the furnace with a small Ar flux. Temperature is raised to 500 °C and hydrogen is released from the sample due to the decomposition of  $UH_3$ . Recovering of the  $\gamma$ -phase in the whole sample is achieved by raising the temperature at least at 750 °C, for a suitable period of time.

All of the above described steps can be performed in a single process without opening the chamber.

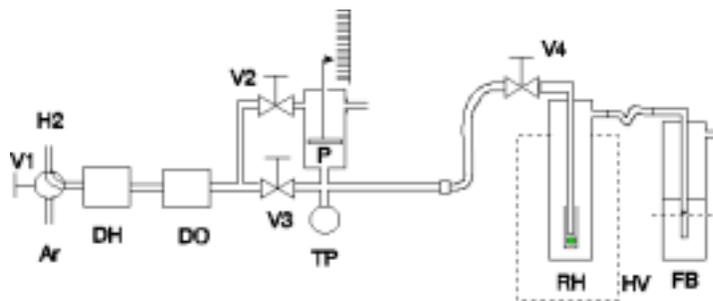


Figure 1 - Hydride apparatus - DH: dehumidifier, DO: oxygen stripper; P: piston; TP: pressure transducer; RH: hydriding chamber; HV: vertical furnace, FB: bubbling flask with silicone oil seal.

Microstructural characterization of the samples was performed by X-ray diffraction (XRD), optical microscopy (OM), scanning electron microscopy (SEM) and electron microprobe analysis (EPMA).

## Results

The results obtained applying the hydride-dehydride process to samples from two ingots of U-8 wt % Mo, and two of U-7 wt % Mo are presented in this work.

All the ingots have homogeneous microstructure of grains, 10 to 40  $\mu\text{m}$  in size, Figure 2. Impurities as globular precipitates are randomly distributed in the alloys. These precipitates remain without any change in the subsequent treatments. XRD patterns show only  $\gamma$ -phase retained due to the rapid cooling of the ingots, Figure 3.

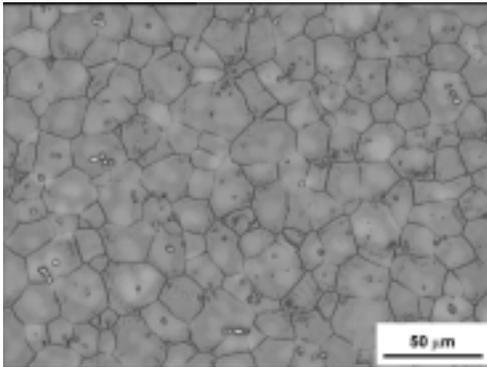


Figure 2: As-cast microstructure of U-7wt%Mo alloy

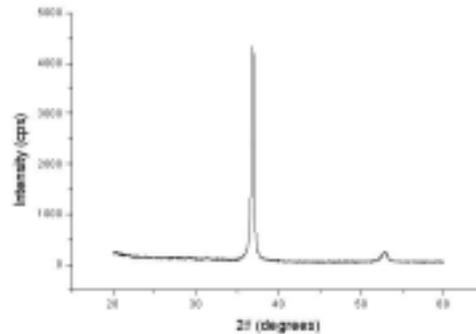


Figure3: XRD pattern of as-cast alloy

All the samples were heat treated at 1000 °C in order to homogenize the composition. Grain growth of  $\gamma$ -phase occurred in the longer anneals. As a result of heat treatments performed between 560 and 565 °C (with or without intermediate quenching after homogenization), the  $\gamma$ -phase transforms by a cellular decomposition composed by lamellae of  $\alpha$ -phase and enriched  $\gamma$ -phase [4]. This transformation starts at grain boundaries and proceeds toward the interior of the grains. Times between 10 to 24 hours, depending on temperature and the composition of the sample, are necessary in order to obtain the convenient rimmed structure as is shown in Figure 4. The XRD pattern confirms the presence of the two phases, Figure 5.

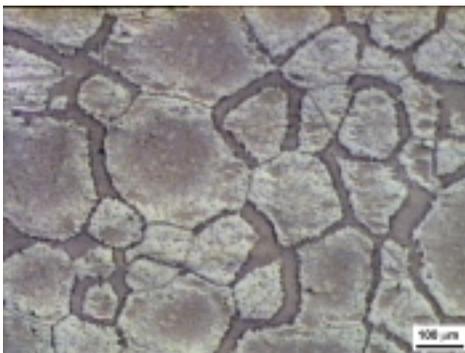


Figure 4 - U-7wt%Mo 24 h at 1000 °C and 24 h at 565 °C

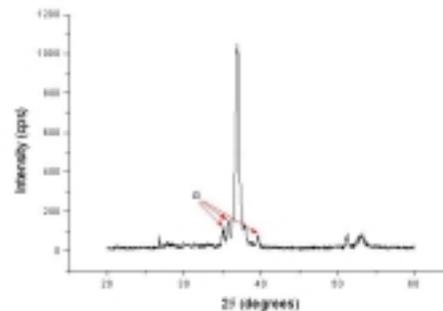


Figure 5 - XRD pattern of sample in Fig.4.  $\alpha$  and  $\gamma$  phase

Samples in this two-phase structure were treated in the hydriding reactor. Typical absorption curves are presented in Figure 6.

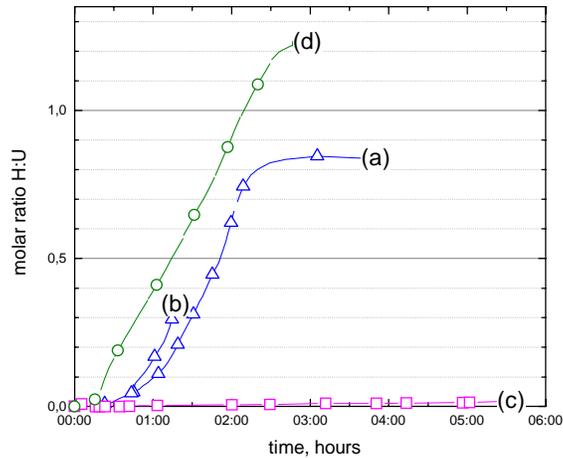


Figure 6 - Kinetics of gas-phase hydriding ( $P_{H_2} = 1$  atm) of U-Mo alloys with microstructures resulting from thermal aging of the  $\gamma$ -phase in the metastable field: (a) 8 wt % Mo, discontinuous cellular transformation at grain boundaries; (b) idem (a), partial hydriding; (c) 8 wt % Mo, without cellular structure; (d) 7 wt % Mo, increased amount of cellular transformation compare to sample of curve (a). Hydriding temperature: 250 °C.

It was observed that when the uptake of hydrogen reaches the saturation, the sample is converted to powder, Figure 7.

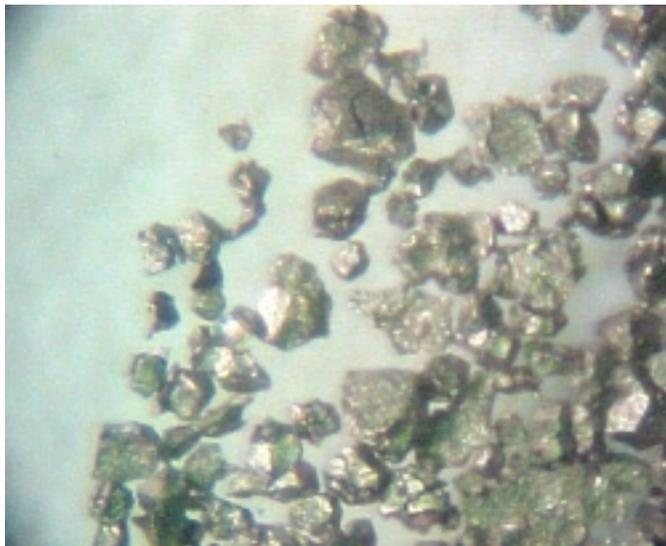


Figure 7: Powder from U-7 wt % Mo alloy. Each particle correspond to a single grain in the original sample

Once the powders were dehydrided and annealed 3 h at 750 °C, they were included in epoxy and polished in order to observe a transversal section of the particles. Optical microscopy shows a

single-phase structure, and the XRD patterns indicate that  $\gamma$ -phase has been recovered in the whole sample, Figure 8 and Figure 9.



Figure 8 - Particles included and polished.

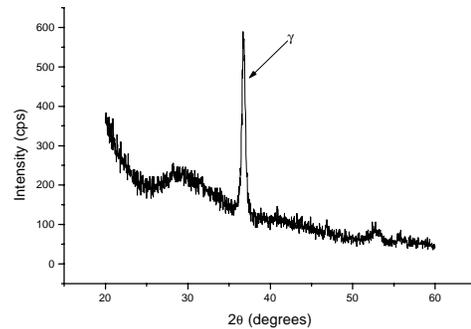
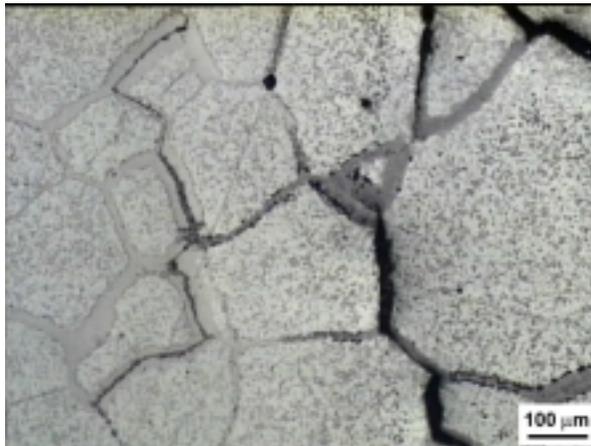


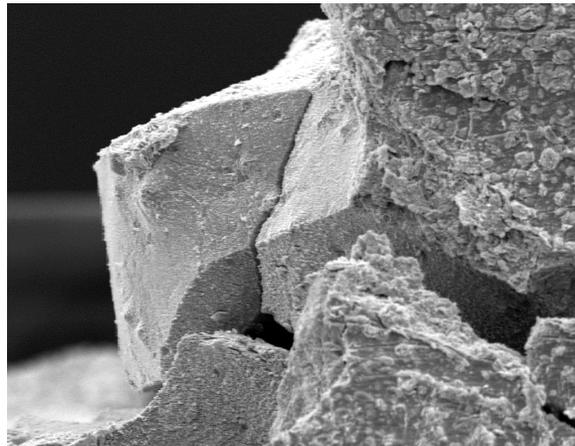
Figure 9 – DRX pattern of dehydrated powder

Some experiments were interrupted in order to study the evolution of the process. It was seen that before the maximum uptake of hydrogen is reached (curve *b* in Figure 6) the sample has already started to crack and decrepitate, Figure 10.

It is clearly observed that intergranular cracking is the main mechanism of the decrepitation. It starts at the surface and proceeds towards the interior of the sample..



a



b

Figure 10 –a: Transversal section of a partially hydrated sample. b: SEM micrograph showing a detail of an edge.

The amount of cellular transformed zone is closely related to the initial composition of the alloy, including the impurities. In U- 7 wt % Mo the transformation starts at shorter times than predicted elsewhere [5]. Besides, an alloy accidentally contaminated with copper (> 50 ppm), oxygen ( $\approx$  500 ppm) and nitrogen ( $\approx$  240 ppm) did not show the transformation at the same time than a pure one. The quantitative effects of the impurities have not been estimated yet.

Also for the longest anneals in the range of 560 to 565 °C some samples showed the presence of the intermetallic  $U_2Mo$ , they also can be converted to powder.

## Conclusions

This work shows that it is possible to obtain powder from the  $\gamma$ -phase of U-Mo (7 and 8 wt % Mo) alloy by the hydride-dehydride process.

The uptake of hydrogen and the further hydride precipitation is induced when the  $\alpha$ -phase is part of the cellular zone formed at grain boundaries.

Heat treatments between 560 and 565 °C from 10 to 24 h allow to obtain the optimal microstructure in these alloys to be subject to hydriding.

The time required for producing cracking and the subsequent production of powder is dependent on the fraction of the cellular structure in the alloys. Because the decrepitation starts at the surface of the samples, the time to pulverize is very dependent of the size and shape of the specimen.

It must be pointed out that the grain size after the treatment at 1000 °C determines the size of the particles constituting the powder.

## Acknowledgements

The authors want to thank P. Adelfang, E. Pasqualini and A. Denis for their support through the communication of the fundamental idea of the work and the provision of the starting materials.

They also thank G. Hofman and J. Snelgrove (ANL-USA) for the useful discussions and the encouragement to present this paper.

The Technical Staff of the Departamento Materiales (CAC-CNEA) had an important participation in the fulfillment of this work. This is sincerely recognized.

## References

[1] Snelgrove J., Hofman G., Meyer M., Trybus C., and Wiencek R., Nucl. Eng. Design, 178 (1997) 119.

[2] Clark, C.R., Meyer M.K. and Strauss J.T., 'Fuel powder production form ductile uranium alloys', 1998 International Reduced Enrichment for Test Reactor Conference, Sao Pablo, Brazil (1998)

[3] Pasqualini E., Personal communication, Technical meeting, Dto.Combustible and Dto.Materiales, CAC, CNEA, October 27<sup>th</sup> 1999.

[4] Blake D. and Hehemann R., 'Transformations in Uranium Base Alloys' in Physical Metallurgy of Uranium Alloys, Proceedings of the Third Materials Technology Conference, Editors: Burke J., Collin D., Gorum A., Greenspan J., Brook Hill Publishing Co., Chestnut Hill, Massachusetts (1976) 189.

[5] Hofman G., Meyer M., Ray A., 'Design of high gamma-phase uranium alloys for LEU dispersion fuel applications', 1998 International Reduced Enrichment for Test Reactor Conference, Sao Pablo, Brazil (1998)