Scaling up the Production Capacity of U-Mo Powder by HMD Process.

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Abstract

The recent discovery that uranium alloys in metastable gamma phase can be hydrided at low temperatures and pressures have allowed developing the method of comminuting bulk materials by milling the hydride to desired size and then dehydriding the powder. This process is called HMD (hydriding-milling-dehydriding) and needs an initial step of hydrogen incorporation to allow the alloy to be hydrided.

This four step process has been conveniently set up for the production of U-7Mo powder for its use in nuclear fuels. Low equipment investment and low man power are needed for this achievement.

The process is being analyzed in its scaling up for one kilogram batches and a 50 kilogram per year production capacity of U-Mo powder.

Introduction

Methods of producing U-Mo powders where thoroughly analyzed at CNEA during the last two years. Different concerns were taken in account from the point of view of equipment investment, rentability, process control, repeat- ness, final product characteristics, workmanship, automatization and scale production. Known methods were analyzed using bibliographical data (centrifugal and gas atomization, machining, etc.) and experimental results where obtained for alternative processes (cyclonic centrifugal atomization, spark erosion, wheel grinding, hydriding, etc).

One of the more important results of this field research was the discovery of the massive hydriding of uranium-molybdenum alloys in gamma phase at low temperature and pressure (1, 2). The brittleness of the hydride (300 VH) allows performing conventional milling and dehydriding for obtaining U-Mo powder (HMD process). Natural uranium in a 7% w/w Mo composition was used for this development that actually is in the industrial scaling up final step. Optimal conditioning of the alloy to be hydrided was selected testing the influence of different casting molds, fragmentation of the melted material, lump size, impurity levels, surface conditioning, residual stresses, etc. Process variables such as pressure, temperature, time and batch sizes were optimized for the following steps of hydriding, milling and dehydriding using conventional laboratory equipment.

Miniplates and full size MTR fuel element plates were fabricated without major inconvenient as prototypes to test production performance.

Actual development is focused in the optimization of the comminution of the hydride phase using low impact mills to obtain less than 30% fines (45 microns). Full optimization of the production process is in its way and a facility is being build up for performing initial practices with enriched uranium. It is planned that the facility will be shortly in production with an initial scale production of 50 kg per year.

Hydrogen Incorporation

An initial step of hydrogen solubilization and probably some hydride precipitation is needed for the hydriding of the U-Mo alloys. This initial step is carried out during one hour in a one bar pure hydrogen atmosphere at 700 °C. Temperatures from around 500 °C to melting temperature where tested with similar results. Pressure as high as two bars was tested. Alloys in which the hydrogen incorporation was performed in pressures less than 250 mbar during a couple of hours could not be hydrided. The hydrogen incorporation during this step is of the order of tens of hydrogen wppm´s. It is believed that hydrogen could also be incorporated during the melting of the alloy or by cathodic charge. What is happening in the material during this initial step that allows that the alloy can be hydrided afterwards could not be resolved and is an open question. Casting conditions are important from the point of view of residual stresses obtained. Worst results were obtained with compression surface stresses. The initial incorporation of hydrogen is a necessary condition for performing the hydriding of the gamma phase alloy.

Hydriding

The range of hydriding the U-7Mo alloy is between 50 °C and 190 °C and the maximum absorption rate is around 120 °C. The differences noticed by working at pressures below two atmospheres were fundamentally that the rate of hydrogen absorption increased as pressure was increased. No changes were noticed in the temperature range when hydriding at different pressures. Hydrogen absorption rates can reach values as high as 1 lt/min.kg.

The hydriding of the alloy is performed in the same chamber used for the initial hydrogen incorporation step. One kilogram of U-7Mo alloy lumps of approximately 50 grams are used and hydrided at 120 °C until saturation in a one atmosphere pressure of pure hydrogen during 36 hours. Pressure control is performed with a loop control of pressure and flow. A low inertia furnace is used in this step for quick control of the exothermic hydriding reaction avoiding temperature excess. A typical curve of the hydriding process is shown in figure 1.

A highly stressed one phase hydride is obtained with an A-15 structure and a stoichiometry of MH_{3-x} , were M stands for the U-7Mo alloy and x is smaller than .5. The general appearance of the hydride is of fragments of approximately 5 mm size with small transgranular cracks 30 microns apart. The reason of these fragmentations is the more than 50% volume change between the initial and the final phases.

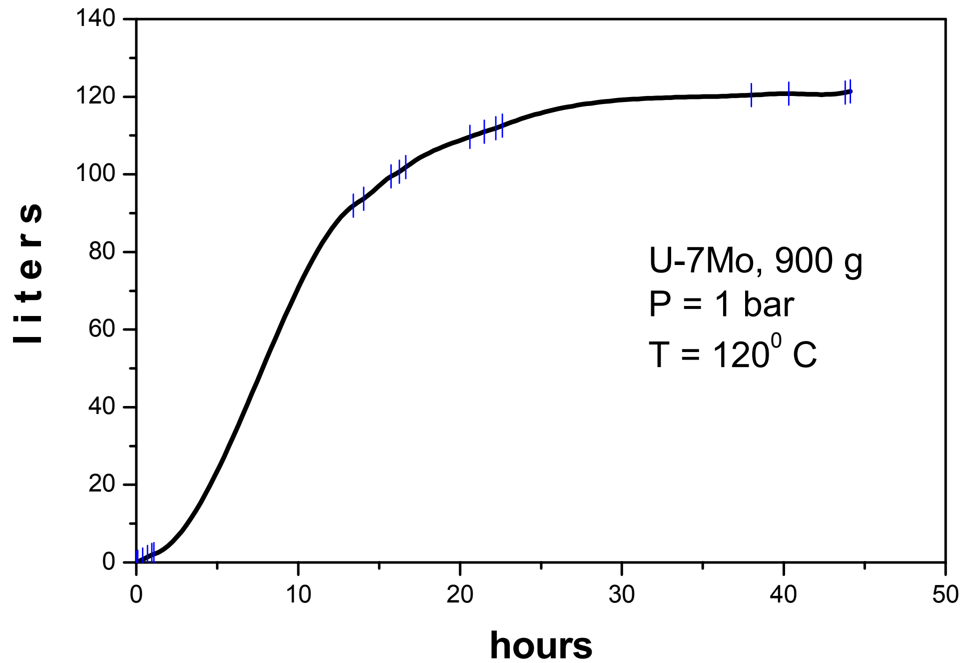


Figure 1. *Absorption of hydrogen during the hydriding of U-7Mo.*

Milling

The comminution of the hydride phase can be done with conventional milling equipment. If high impact mills are used, more than 50 % in weight fines (particles with less than 45 microns size) are obtained. Since the reaction kinetics of U-Mo with the aluminum matrix in the meat is higher than in the case of silicide fuel (3), it is convenient to reduce the presence of small particles.

Low impact mills were used in which less than 30 % fines are obtained. Specially designed two roll mill or conical vibratory crushers are being used (figure 2). Although there seems to be no technical necessity of using a low oxygen atmosphere for the milling, it is mandatory to avoid hazardous incidents because of the pyrophicity of the hydride and the hydrogen presence. A glove box with an atmosphere with air depleted in oxygen content (less than 5% oxygen) can be used.

The material is milled so as to have particles size smaller than 125 microns. Fine particles, if desired, can be refused in other melting batch in a proportion that will depend on oxygen solubility (4) and the quantity of oxide present in the particles, or directly melted in an arc furnace. Milling of the one kilogram batch is performed in less than two hours.

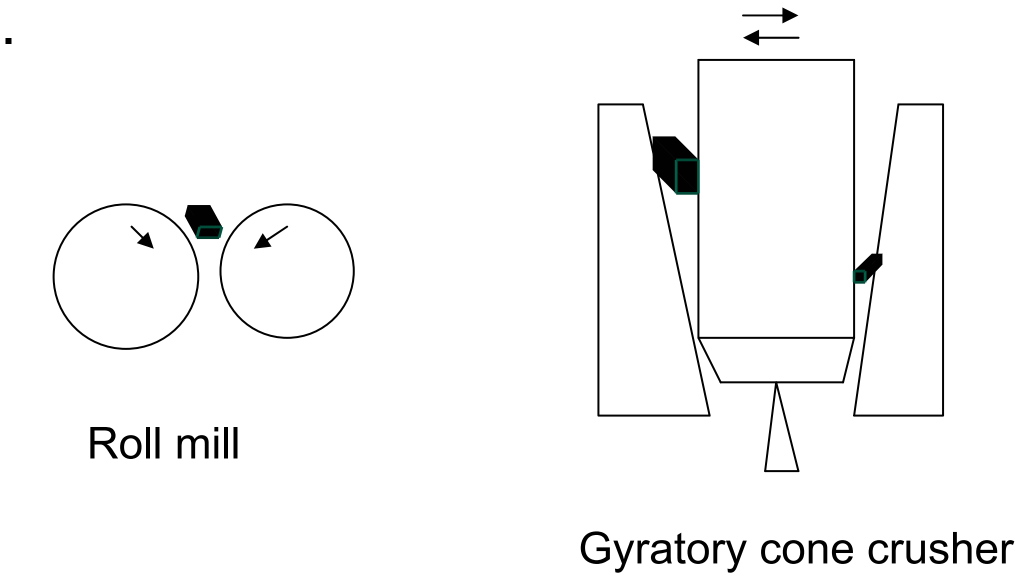


Figure 2. *Schematic view of low impact mills.*

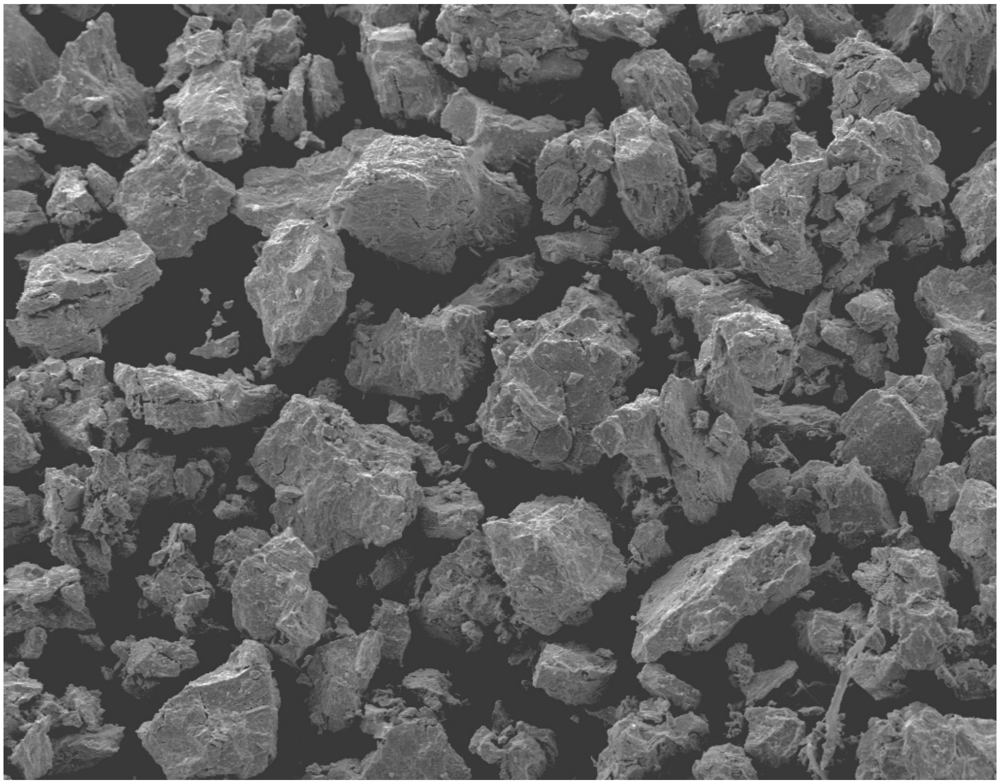
Dehydriding

The dehydriding of the milled powder is performed in the same chamber that is used for the hydriding. A maximum temperature of 700 °C is achieved using the mechanical and diffusion pumps so as to guarantee that no hydrogen remains in the alloy and the gamma phase is recovered. The process is done in a controlled way in approximately 5 hours so as to avoid the flying of the smaller particles out of the heating zone because of hydrogen flow. The densification of the material involves a reduction of slightly more than 15% in the dimension of the particles.

Figure 2 shows the appearance of the dehydrided particles. In figure 4 it is represented a metallography of the meat of a fuel plate fabricated with the HMD comminuting process. Holes between particles were fundamentally produced during polishing. UMo particles do not brake during the rolling of the plate and the final porosity is some value between 5 and 10%.

Passivation

The hydriding and dehydriding steps of the HMD process are performed in atmospheres without oxygen. Before exposing the material to air it is necessary to passivate the surface to avoid “burning” the material. One hour exposure in a mixture of 25% air and 75% argon followed by another hour exposure to an atmosphere of 50% air and 50% argon is enough to slowly oxidize the material. In the case of the powder it is important to guarantee that all of it is oxidized, for example, by moving the powder during the passivation.



During milling, particles oxidize as they crack in the low oxygen atmosphere and no further passivation is needed.

Figure 3. *U-7Mo powder. Size of big particles is 100 microns (SEM).*

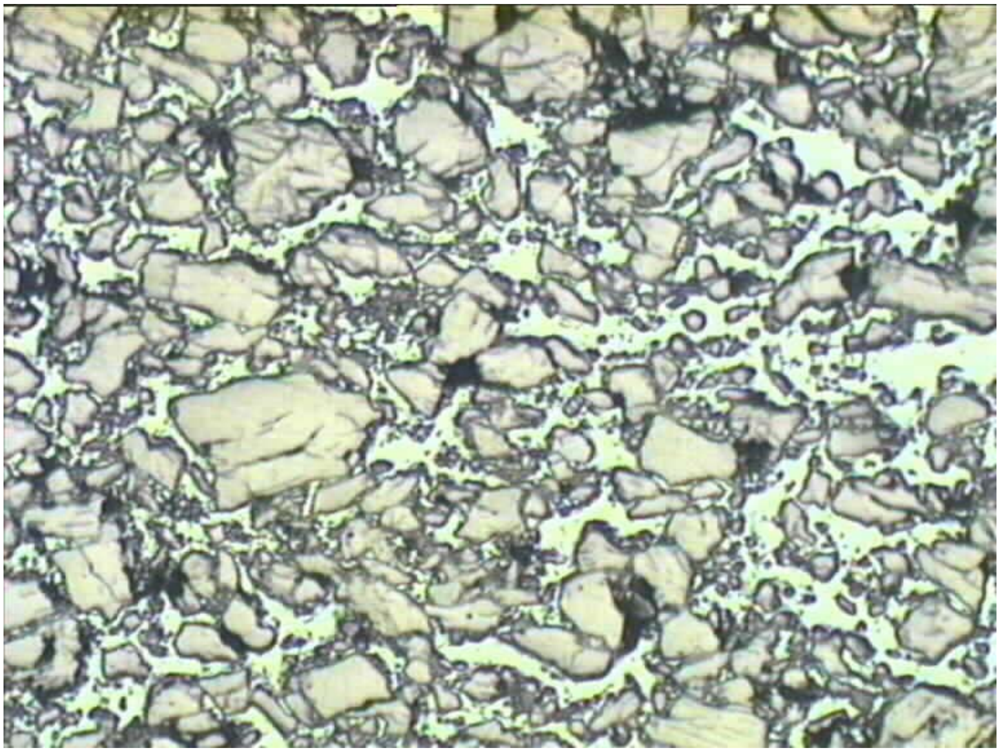


Figure 4. *Metallography of the meat of a plate obtained using HMD U-7Mo powder. The size of the big particle is approximately 100 microns.*

Conclusions

The HMD process is accomplished in four steps that have been fit in their characteristics for a scale production of 50 kg/year of U-7Mo powder in one kilogram batches. Temperature, atmosphere and time were specified for the initial hydrogen incorporation, hydriding and dehydriding steps. The milling of the hydride is performed in low impact mills. The hydriding and dehidriding have been fully automatisized.

Conventional equipment is used that requires small man-power and low investment. A facility is being build up for this capacity to work with enriched uranium.

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