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POWDER FABRICATION OF U-MO ALLOYS FOR NUCLEAR DISPERSION FUELS

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ABSTRACT

For the last 30 years high uranium density dispersion fuels have been developed in order to accomplish the low enrichment goals of the Reduced Enrichment for Research and Test Reactors (RERTR) Program. Gamma U-Mo alloys, particularly with 7 to 10 wt% Mo, as a fuel phase dispersed in aluminum matrix, have shown good results concerning its performance under irradiation tests. That's why this fissile phase is considered to be used in the nuclear fuel of the Brazilian Multipurpose Research Reactor (RMB), currently being designed. Powder production from these ductile alloys has been attained by atomization, mechanical (machining, grinding, cryogenic milling) and chemical (hydriding-dehydriding) methods. This work is a part of the efforts presently under way at IPEN to investigate the feasibility of these methods. Results on alloy fabrication by induction melting and gamma-stabilization of U-10Mo alloys are presented. Some results on powder production and characterization are also discussed.

1. INTRODUCTION

This research is part of the efforts under way at National Commission for Nuclear Energy (CNEN), and particularly at IPEN (Nuclear and Energetic Research Institute) for the construction of the Brazilian Multipurpose Research Reactor (RMB), currently been designed.

Gamma U-Mo alloys have been for long considered as fuel phase in research and test reactors using dispersion fuel in aluminum matrix [1]. Promising results concerning performance under irradiation tests of U-Mo alloys, especially with molybdenum content ranging from 6 to 10 wt.% Mo [2], have encouraged us to consider this fuel phase for the second stage of the reactor operation, since uranium silicide compound, U_3Si_2, already produced here, will be used at first.

As a fuel based on dispersion concept U-Mo alloys must be used in powder form. At least three main fabrication routes for U-Mo powders could be listed: atomization (mainly centrifugal atomization by rotating disk method [3] or even rotating electrode process [4]), mechanical comminution, i.e., machining or grinding [5,6], and chemical comminution, i.e. hydride-dehydride process, also known by its acronym HDH [7-10]. HDH of gamma U-Mo alloys can be accomplished by heating the alloy at temperatures where it decomposes in two

phases, i.e. alpha and gamma' (U_2Mo). Since alpha uranium is easily hydrated, generating a very fine powder, its content must be carefully controlled (changing soaking time) in order to obtain particles within the desired size range. A variation of the last route, named HMD, combines hydriding-dehydriding with milling process [11]. According to the authors, gamma U-Mo alloys can form a U-Mo hydride (A-15 structure) that embrittles the alloy, but not intensively, so powder could only be produced by interposing a milling operation, before dehydriding.

Each one of the previous routes are held by commercial or potential suppliers based on features like particle size range yield of the powder, costs, and the more important one, irradiation behavior. Since there are some controversial arguments, an investigation at local process conditions is necessary to find the best technological solution.

The focus of this preliminary work is to compare the characteristics of U-Mo alloy powders (10 wt% Mo) fabricated by two routes: mechanical grinding and HMDH. In this particular case, HMDH stands for hydrogenation-milling-dehydrogenation, since a hydride phase was not formed. It was reported before that gamma U-Mo alloys (particularly U-10wt%Mo) suffered a loss of ductility when submitted to a hydrogen atmosphere, by incorporating hydrogen interstitially [12]. This fact was used here to provide enough brittleness to the alloy to allow comminution by high-energy ball milling. Details of both procedures are given elsewhere.

2. EXPERIMENTAL

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 through magnesiothermic reduction [13] and the metallic molybdenum was supplied with 99.95% purity, as small cylinders with 3 mm in diameter and 3 mm in height. Both materials were charged inside the zirconia crucible and heated by induction under high-purity argon atmosphere up to melting. Melting temperature was maintained for 3 minutes providing homogenization, than the furnace was turned off allowing the alloy to be solidified inside the crucible. The solid material was a cylindrical piece with near 40 mm in diameter and 50 mm in height, weighting around 1200 g with a density of 16,87 g/cm^3. The ingot was treated at 1000°C for 72 hours under pure argon and quickly cooled for retention of the gamma phase. It was cut in pieces for studding the two routes for powder preparation, namely mechanical grinding (MG) and hydrogenation-milling-dehydrogenation (HMDH).

For the mechanical grinding route, the powder was produced by using high-speed grinding (15000 rpm) with diamond abrasive wheel. The abrasive wheel was 4 mm in diameter, having impregnated diamond particles with mean diameter of about 100 μm. Grinding was accomplished inside a glove-box under protective argon atmosphere.

For the HMDH route, small pieces were taken from the U-Mo ingot (with approximately 10x50x5 mm in thickness) and were individually heated at 400°C for 3 hours under high purity hydrogen (99.9999%) at 3 bar. A Sievert type apparatus were used and no measurable hydrogen intake could be noticed, with the pressure gauge used (precision of 0.5 bar). At this temperature and time, alpha phase is not supposed to be formed, since about 40 hours would be necessary to start the gamma decomposition according to published TTT diagram [14].

Next the pieces were manually crushed in a stainless steel mortar. The resulting granules were 3 mm in length. For comparison, crushing of pieces not hydrogenated treated was carried out but not succeeded due to ductile behaviour of the parts. This was taken as an indication that some hydrogen intake must be occurred in former pieces. U-Mo granules were milled in a planetary ball mill at 400 rpm for 10 hours, with ball-to-powder weight ratio of 20:1. The vial and the balls were made from hardened steel. Loading and opening of the vial occurred inside a glove-box with protective argon atmosphere. After milling the powder was heat treated under vacuum at 400°C for dehydrogenization.

X-ray diffraction was conducted on pieces of the heat treated ingot using Cu-Kα radiation. Powder particles were characterized by SEM analysis and also optical microscopy using image analysis for size distribution. For optical microscopy powder samples were dispersed over a glass thin plate. The images were taken by transmitted light which allows enough contrast for visualization. For each type of powder, measurements were carried out in 3 samples, accomplishing 24 image fields and at least 6000 particles. The smallest dimension between two tangents of the particles projection was considered for measurement (minimum Feret's diameter). This was done in order to reproduce the results that would be obtained by sieving analysis, since the powder mass produced was not enough to perform sieving in a reliable manner. By sieving, the second higher dimension of each particle is actually measured (which is able to pass through the sieve openings), corresponding in this particular case to the minimum Feret's diameter (on doing this it is assumed that the thickness of the particles was not measured in projected images).

3. RESULTS AND DISCUSSION

Fig. 1 shows the diffraction pattern from U-Mo ingot after heat treatment. BCC gamma phase was the major phase with some unidentified peaks of minor phases. The relative intensity of the (110) reflection from gamma phase should be the highest one, which indicates that some texture is still present. In spite of that, the heat treatment was considered suitable for the purpose of this preliminary investigation.

Fig. 2 shows the morphology of the powders prepared by both routes, mechanical grinding (MG) and hydrogenation-milling-dehydrogenation (HMDH). It was observed that the powder prepared by MG route (left column in Fig. 2) presents particles with acicular and flake shapes, while the particles from HMDH route (right column in Fig. 2) are more regular and equiaxial. Particle size distribution curves along with optical micrographs from both powder production routes are shown in Fig. 3. The powder prepared by MG route presented particles sensibly larger than the ones prepared by the HMDH route. The mean particle size (50 wt%) was about 100 μm for MG powder and 50 μm for HMDH powder. Furthermore HMDH powder particles fits very well the size requirements of dispersion fuels, with practically 100 wt% below 150 μm and about 30 wt% below 45 μm, while MG powder particles were shown to be larger, with more than 20 wt% above 150 μm. Other important difference was the aspect ratio of the particles (maximum to minimum Feret's diameters). The aspect ratio reached 10 for the MG powder, much higher than the maximum ratio measured for HMDH powder, close to 4.

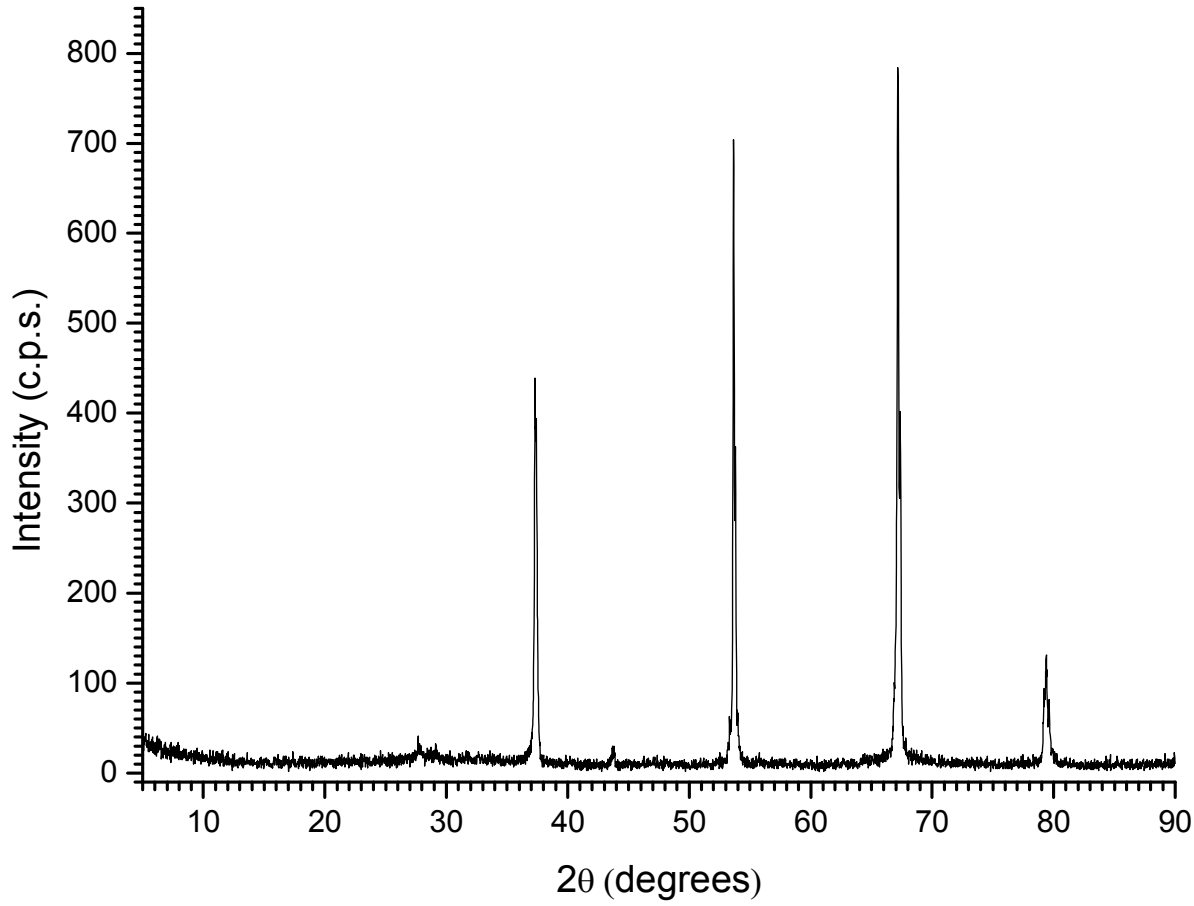

Figure 1. Diffraction pattern of the U-10wt%Mo alloy showing BCC gamma phase.

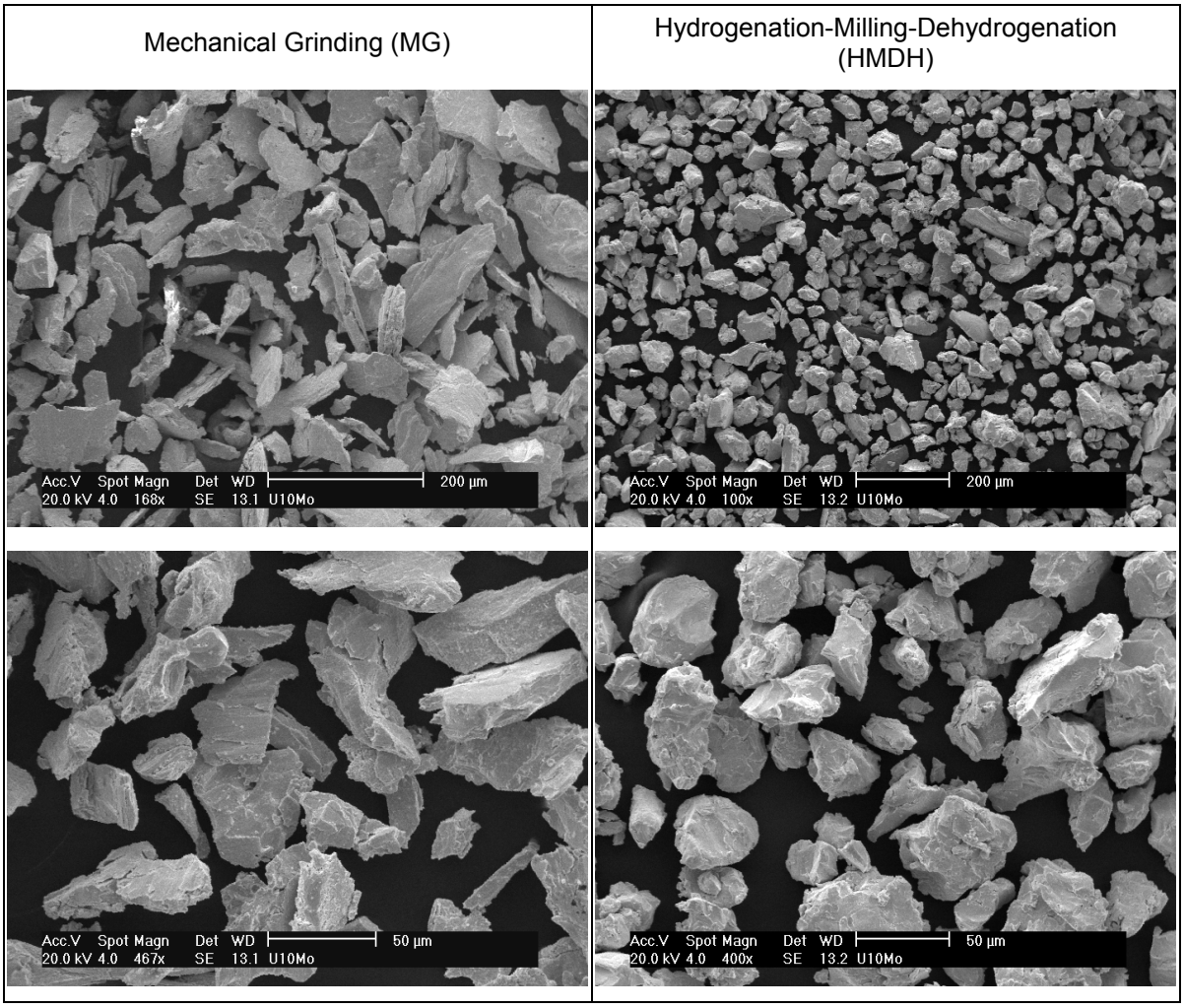

Figure 2. Scanning electron micrographs of powder particles from both investigated routes.

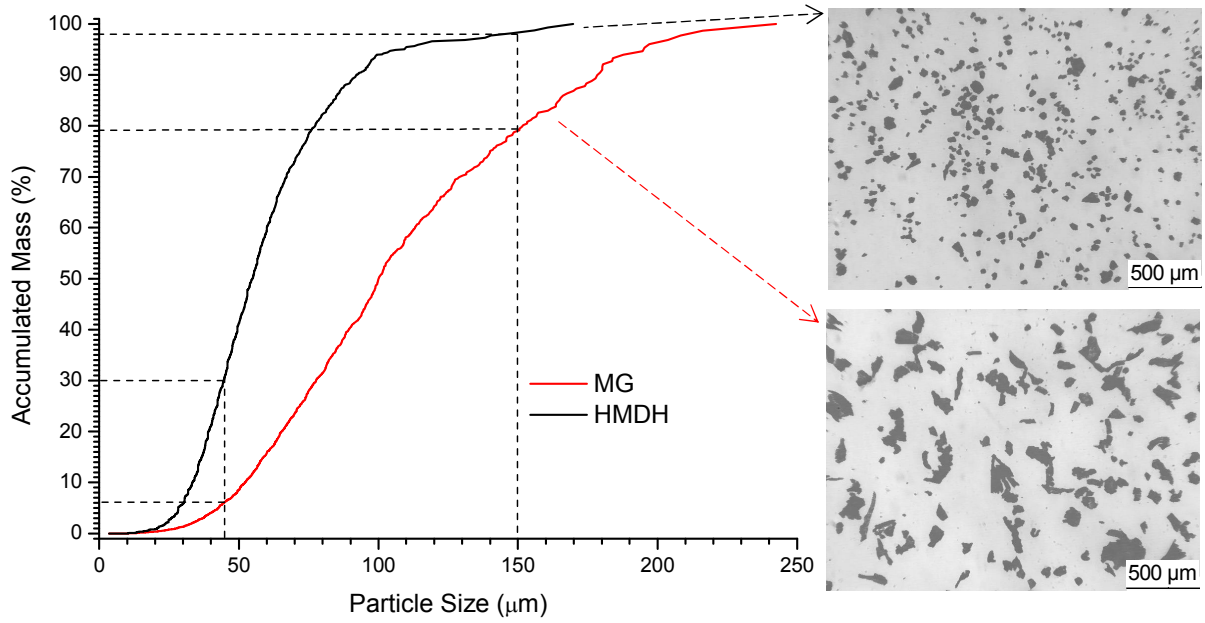

Figure 3. Particle size distribution curves for the powders prepared by mechanical grinding route (MG – red curve) and hydrogenation-milling-dehydrogenation route (HMDH – black curve).

4. CONCLUSIONS

Despite the preliminary character of this work, the results indicate the technical feasibility for producing powders from both investigated routes. The control of variables of the MG route, as the size of the diamond granules of the abrasive wheel, the pressure of the tool under the alloy surface and the rotation of the grinding wheel machine, should promote the necessary adjustment in the particle size distribution. Powder produced by the HMDH present a particle size distribution compatible to be used as a dispersion fuel. Further work is necessary to increase the yields in order to evaluate both process routes as real technological alternatives for nuclear fuel powder production to research reactors.

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