

## STUDY OF THE MANUFACTURING PROCESS OF $\gamma$ -U7%wtMo DISPERSION FUEL PLATES

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### ABSTRACT

The search for new materials for nuclear fuels has been developed over the last 50 years, with the main aim of increasing the fuel efficiency during the operation of the reactors. The need to increase the uranium density in fuels to compensate the reduction of enrichment proposes that the UMo alloy is one of the materials that presents better characteristics to be used as fuel: molybdenum is a material that retains the gamma phase of the uranium in low concentrations, which is the only stable phase of uranium under the irradiation conditions, besides having low thermal neutron absorption. Although more advanced studies already provide information on the interaction between UMo and the Al matrix, we still need to study how this material behaves during all processing steps for fuel fabrication. The present work has the objective of to deepen the technological knowledge about the stages of production of dispersion type nuclear fuel, including the comminution process of the UMo alloy. The alloy pulverization made by the hydriding-grinding-dehydriding technique still reveals a large number of unknowns in the process variables. Knowing some parameters already existent in the literature, it is possible to discuss the behavior of the hydriding process and envision improvements to optimize it as well as make it reproducible. Subsequent manufacturing steps for briquette and rolling were performed according to IPEN's expertise and the results indicate that the UMo alloy is mechanically doable and may prove to be a substitute fuel for the current  $U_3Si_2$  with a higher uranium density.

### 1. INTRODUCTION

The need to reduce the uranium enrichment to the limit of 20% around the world in the late 1970s, started the development of fuels with higher uranium mass in the fuel element. Although the uranium increase outweighed the enrichment gap, the physical and mechanical properties of the materials became limiting to development.

The major problem encountered in compounds with high uranium content in aluminum matrix dispersion type fuels plate is due to their dimensional instability under irradiation during reactor operation. This instability causes swelling of the plates, damaging the thermal exchange, thus compromising the safety of the system [1].

In the middle of 1988, the results of irradiation tests qualified the fuel based on the  $U_3Si_2$ -Al dispersion by the US Nuclear Regulatory Commission with uranium densities up to  $4.8 \text{ gU/cm}^3$  [2, 3, 4].

Although the  $U_3Si_2$  has been qualified, some research reactors with high neutron flux have not yet been able to be converted for low enrichment operation. For these reactors, a uranium

density of 6 to 9 gU/cm<sup>3</sup> is required. U-Mo alloys are possible candidates for replacement of uranium silicide. U-Mo alloys can provide up to 9gU/cm<sup>3</sup> with a volumetric fraction of 45% of fissile material in the dispersion.

The focus of this work is to study the entire manufacturing process of a U-7%wtMo mini-fuel plate from the alloy melting stage to the final plate preparation, in order to identify how the manufacturing parameters of the fuel plate with uranium silicide are impacted by differences in the physical and mechanical properties of U7Mo.

## 2. EXPERIMENTAL

The entire process was developed based on IPEN's expertise in the fabrication of uranium silicide dispersion fuel type with the adaptations for the new uranium-molybdenum fuel. The fuel plate production steps are described below.

### 2.1. U7Mo alloy fusion

Alloy fusion was carried out in the induction furnace model Indutherm VTC200VTi. The pure uranium metal was previously cut into cylinders and weighed to define the molybdenum charge at a concentration of 7% of weight in the alloy. Molybdenum metal was in the form of small cylinders and after assembly, the charge was loaded in a zirconia crucible.

The oxygen in the furnace atmosphere was removed under vacuum. The system was heated to 1800 °C for about 10 minutes for homogenization of the alloy under argon atmosphere. The casting temperature was close to the 1800 °C. The ingot in cylindrical format was removed from the furnace after 4 hours.

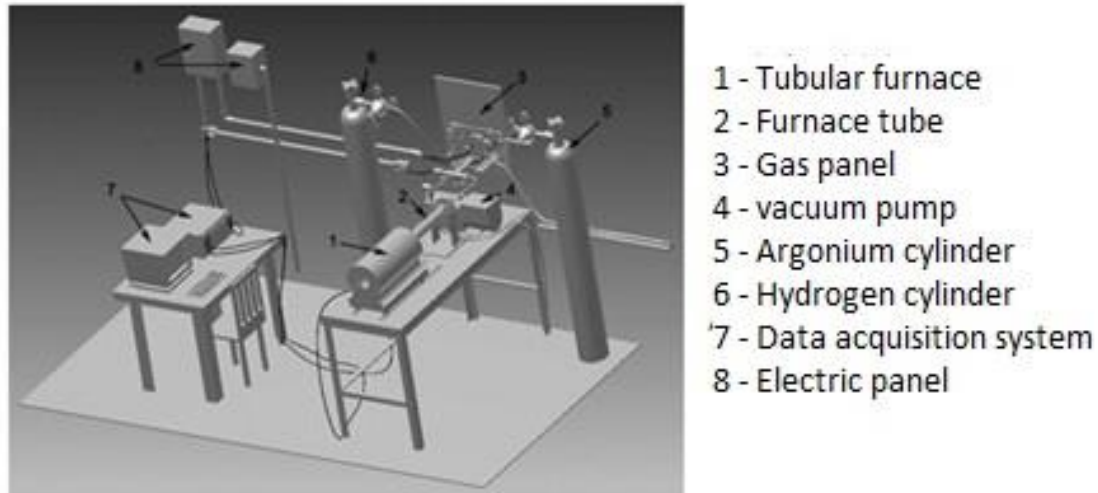
### 2.2. U7Mo powder production

The ingot was cut into two smaller cylinders and pickled with nitric acid to remove the oxidized layer on the surface. The comminution process chosen was the hydriding-grinding-dehydriding (HMD), once the alloy can't be powdering directly due to its ductility. Figure 1 shows the hydriding system.

The samples were weighed inside a glove box, which was hermetically sealed, and placed inside an alumina crucible which was then inserted into a tubular furnace. Three purges were performed with argon, after vacuum of 10<sup>-3</sup> mbar. The samples were hydrided heating at 800 °C (rate of 15 °C/min) under pressurized hydrogen at 2 bar for 1 hour. The reactor was removed from the furnace for cooling until the room temperature. [5, 6, 7, 8, 9, 10, 11, 12].

After cooling, the furnace was opened inside the glove box in an inert atmosphere. The powder was ground manually using a mortar and pestle and sieved manually, originating two particle granulometric size fractions. The granulometry was defined as powder at the appropriate size (44 to 125 µm) and fines below than 44 µm. At the end of this process, the mass of the sample above 125 µm returned to the furnace to carry out a new hydriding cycle. The hydriding cycles were carried out until all the pulverized material size was less than 125 µm.

The dehydrating of the both powders size fractions was performed separately. The vacuum pump was connected to the tubular furnace to obtain a vacuum of  $4 \times 10^{-2}$  mbar. Once the vacuum was obtained, the system was heated to 700 °C at a rate of 5 °C/min and then slowly cooled to room temperature. After finishing the operation, the system was pressurized to 3 bar of argon to avoid presence of hydrogen atmosphere.



**Figure 1: Hydriding system**

### 2.3. Briquettes fabrication

The U7Mo-Al briquettes were prepared according to the traditional procedures adopted by the Nuclear Fuel Center of IPEN, only adapting its dimensional characteristics to obtain miniplates [13].

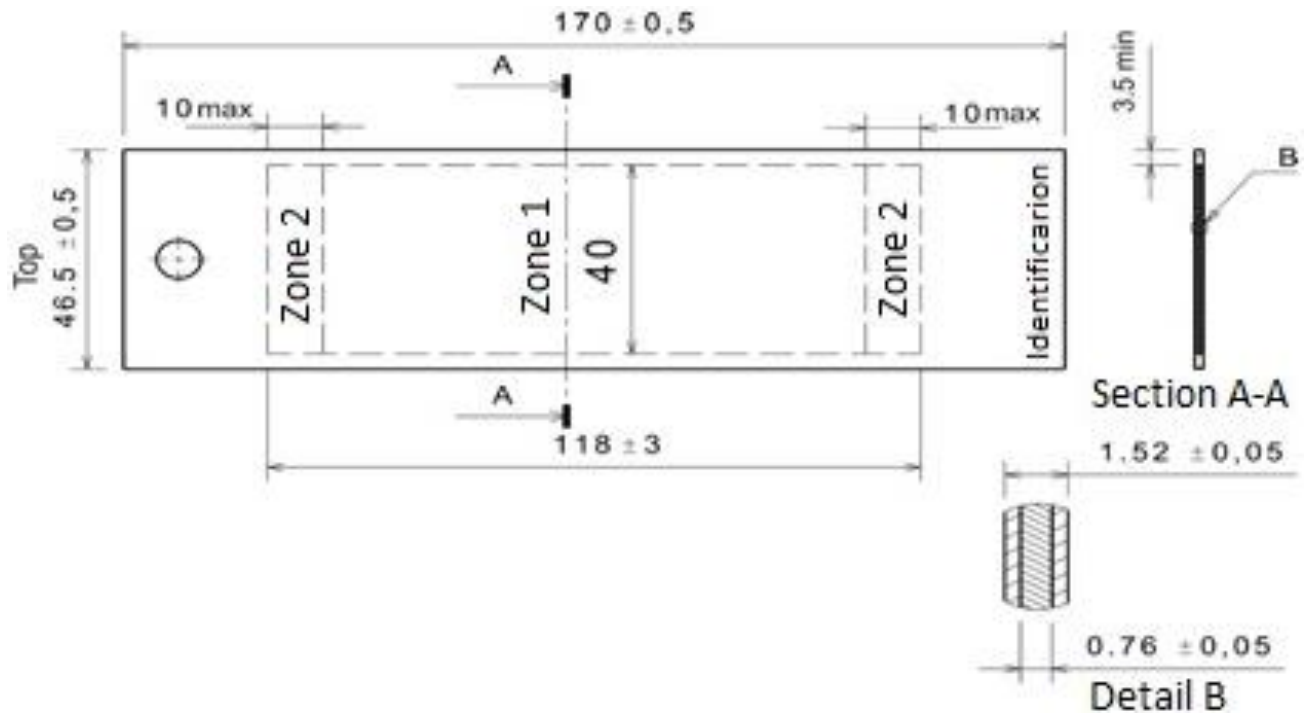
The aluminum and U7Mo powders were weighed separately on a 0.01 mg precision scale inside a glove box. They were then mixed in a glass container with a sealed cap. The pulverized U7Mo was charged first, followed by the Al powder. The containers were then coupled to a blender. The mixture was homogenized for 2 hours in rotary equipment developed by IPEN.

After mixing, the powders were carefully transferred to a matrix for the briquette pressing system. The cavity of the matrix was manually fed, taking care to avoid segregation of the U7Mo powder, which occurs due to the large density difference of the powders. For the compaction, the equipment used was a press of 100 tonnes. The compaction pressure was 4 ton/cm<sup>2</sup>.

The compacted was subjected to a vacuum degassing treatment of  $10^{-6}$  bar at 250 °C for removal of moisture, volatiles and entrained gases.

### 2.3. Miniplates fabrication

The U7Mo-Al fuel miniplates were specified according to the specifications used to manufacture  $UAl_x$ -Al dispersion miniplates used as irradiation targets. The dimensions are shown in figure 2.



**Figure 2: Miniplate dimensions**

The manufacturing technique was the same used in the routine production of fuel plates in the CCN, which is the picture frame technique [13].

The cladding and frame plates were pickled in a 10% NaOH solution, followed by washing and neutralizing in a 40% solution of  $HNO_3$ . The frame was preheated at 480 °C for 5 minutes in order to produce expansion for briquette engagement and fitting the briquette in the frame cavity.

The cladding and frame were welded on the borders by TIG (Tungsten Inert Gas) process. The set was heated to 440 °C for 1 hour before starting hot rolling. Six rolling passes were performed, and in the interval of each passage the set was again heated for 15 minutes at 440 °C. At each pass the miniplates were rotated transversely and longitudinally in order to minimize manufacturing defects. After the visual inspection to verify the existence of blisters, cold rolling was performed with 2 passes at room temperature to achieve the final dimension within specified tolerance.

After the plates were manufactured, they were previously radiographed for fuel meat location for final cutting, as well as for measuring core dimensions (width and length). The miniplates were cut by a hydraulic guillotine.

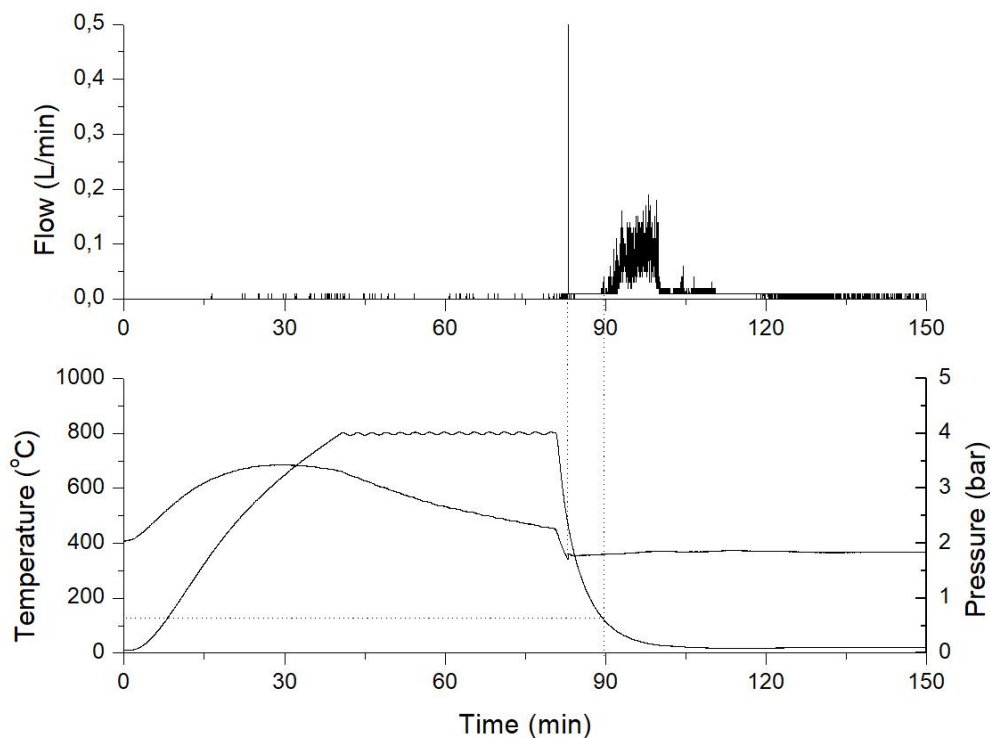
### 3. RESULTS AND DISCUSSION

The ingot of U7Mo was cut in 2 pieces of mass 188.85 g and 183.68 g. Alloy comminution was performed by the HMD technique. In the first hydriding cycle, the two pieces from the ingot were conditioned in alumina crucibles separately, totaling 372.53 g.

During the heating of the first cycle there was a pressure increase in the furnace reactor due to the temperature increase, reaching a maximum of 3.43 bar at 800 °C. This temperature value was maintained for 1 hour and it is noticed a decrease in pressure up to 2.5 bar, possibly due to system leaks. When the furnace was cooled, a sudden drop in pressure to 1.7 bar was observed near 470 °C and consequently an H<sub>2</sub> flow peak to adjust the pressure in the system. When the temperature was close to 125 °C, variation in H<sub>2</sub> flow was observed indicating some level of hydriding in the material. As the furnace remains under constant pressure, the hydrogen consumption in the hydride reaction promotes a replacement of H<sub>2</sub> to maintain the pressure, which is detected by the flow sensor. The conditions described can be seen in the graph of figure 3.

Studies on hydriding indicate that the temperature at which the reaction occurs is in the range of 120 °C [8], which indicates that the oscillation of the H<sub>2</sub> flow presented in the figure 3 is related to the alloy hydriding.

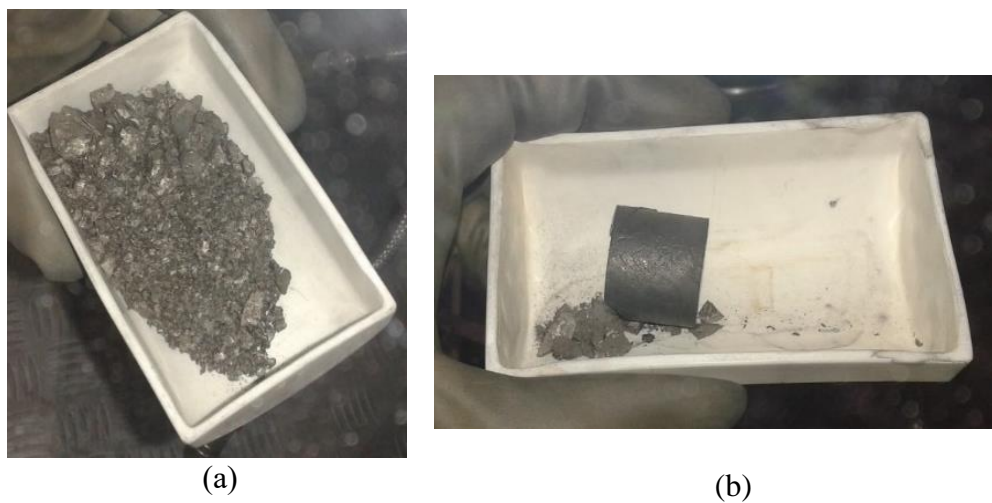
The piece with 188.85 g, called D2, was slightly damaged at the ends, while the piece with 183.68 g, called D4, fragmented, with about 30% of its mass in the desired granulometry. Figure 4 shows the result of the hydriding of the 2 pieces after the first cycle.



**Figure 3: Hydriding**

From the second cycle the parts were hydrided separately. D4 required two more cycles to completely fragment, while D2 required another 8 cycles for complete comminution. The curves of the subsequent cycles were similar to those shown in figure 3, except for the H<sub>2</sub> flow that displayed no oscillation during the hydriding. Although hydriding occurred, the system was not sensitive to the H<sub>2</sub> flow variation, probably due to the decrease in the amount of mass in further cycles which consequently decreased the volume of H<sub>2</sub> reacted.

It is suggested that the difference between the behavior in the hydriding of the two parts is due to the cooling rates of the alloy during casting causing microstructure differences in the ingot. After casting, the top has greater chances of presenting more defects in its microstructure. It was observed that the piece from the top of the ingot was hydrided easier than the piece from the bottom. As observed in [14], the presence of stress in the piece helps to promote hydride reaction.



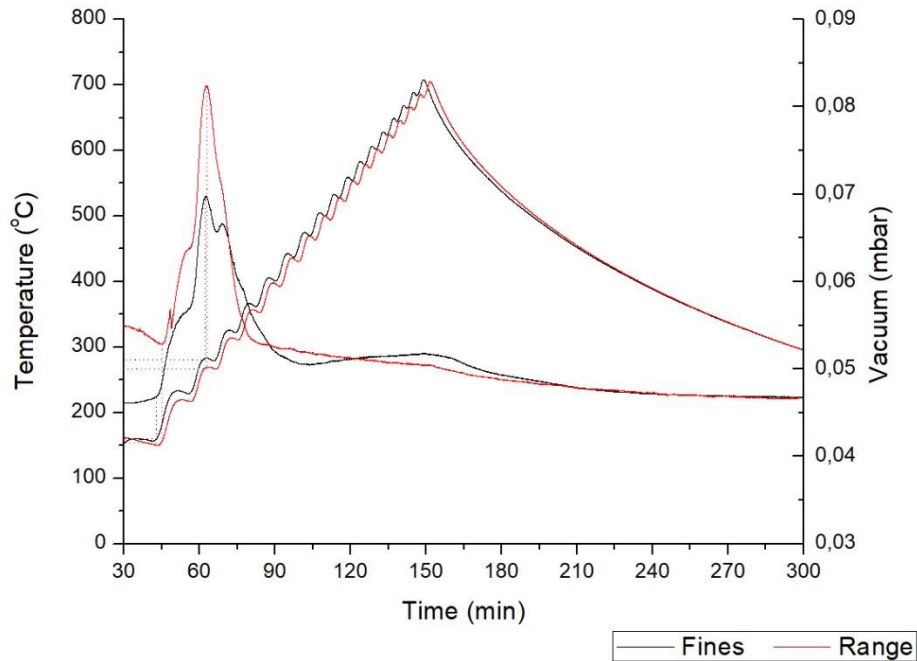
**Figure 4: Ingots after first hydriding cycle D4 (a), D2 (b)**

The dehydriding was performed separately between the two fractions size. The system was heated again at 700 °C under vacuum of  $4.2 \times 10^{-2}$  mbar. The dehydriding curves of the fine and range powders were similar as can be seen in figure 5.

It was observed that in both sizes, the significant increase of pressure begins at about 150 °C. This increase is due to the release of hydrogen gas present in the metal. The steps present in the vacuum curve are consequences of the temperature increase until the maximum rate of H<sub>2</sub> release.

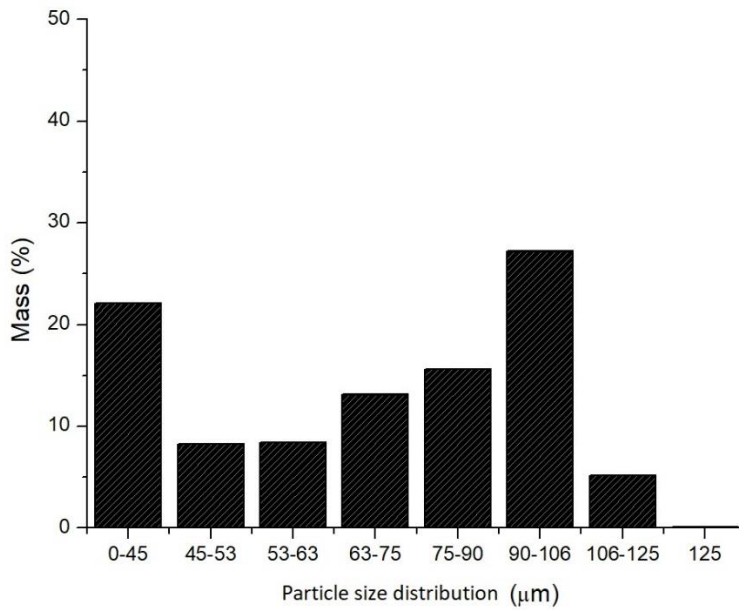
Although the peaks of pressure increase in the system occur in the same temperature range, it is noted that its intensity is related to the mass of hydrided powder: the higher the amount of powder, in the case of powders manufactured in the range, the greater the amount of hydrogen gas released and then the higher the peak intensity. It is interesting to note that even though this variation of mass and intensity occurs, the curves occur in the same temperature range up to approximately 400 °C. After this value, the small pressure oscillation occurs due to the heating of the system.

Although the granulometry of the powders after the hydriding and milling cycles were divided into fines and in the range, the actual value of the granulometric bands was determined after the dehydriding. Figure 6 shows the granulometric distribution of the powders after the dehydriding process.



**Figure 5: Dehydriding**

A pycnometric analysis of the powder was performed to obtain the specific mass. The value was  $16.46 \text{ g/cm}^3$ . Based on this and the dimensions of the plate's meat, it was possible to calculate the amount of U7Mo required for the manufacture of the miniplates. The total mass weighed was 300.61 g, being 240.59 g of powder with adequate granulometry and 20% of fine powders. The total is equivalent to 80% of mass in the beginning of the process.



**Figure 6: Particle size distribution**

In order to reach the specification of the miniplates, the dimensions of the briquettes were calculated according to the previous experience in terms of elongation and enlargement of the meat after rolling. The briquettes used in the manufacture of the miniplates had dimensions of 4.2 mm of thickness by 4 cm of width and 2 cm of length (dimension in the direction of lamination). The briquette mass was calculated according to the volume of the aluminum matrix and the desired uranium density in the fuel, totalizing a mass of 21.01 g, being 6.69 g referring to the Al matrix and 14.32 g of the U7Mo alloy. Figure 7 shows the briquettes.



**Figure 7: Briquettes**



The plates were assembled as explained in item 2.3. After 6 passes of hot rolling, the plates obtained a reduction of 82.5% of the thickness on the set of claddings and frame. Then, the cold rolling was carried out for dimensional adjustment of the same. The thickness of the core was measured by metallography, and the values obtained are shown in Table 1.

**Table 1: Miniplates thickness measures**

| POSITION                          | CLADDINGS<br>(mm) |      | MEAT<br>(mm) |
|-----------------------------------|-------------------|------|--------------|
|                                   | A                 | B    |              |
| Upper Longitudinal Terminal (ULT) | 0.27              | 0.27 | 0.89         |
| Central Longitudinal (CL)         | 0.33              | 0.35 | 0.76         |
| Central Transversal (CT)          | 0.35              | 0.35 | 0.73         |
| Lower Longitudinal Terminal (LLT) | 0.27              | 0.28 | 0.89         |

It is observed that at the ends of the miniplates, the meats are thicker than in the central region, which reduces the thickness of the claddings (ULT, LLT). This behavior comes from one of the most common defects in the production of fuel plates which is the dog boning. Figures 8a and 8b show images of the meat obtained by optical microscopy.

Note a homogeneous distribution of the fuel material throughout the core. These images are typical metallography for a dispersion fuel plate with a continuous matrix in their meat. However, it is observed the presence of another common behavior in the end that is the drag of the meat between the frame and the claddings, which is known as “fish tail” defect.

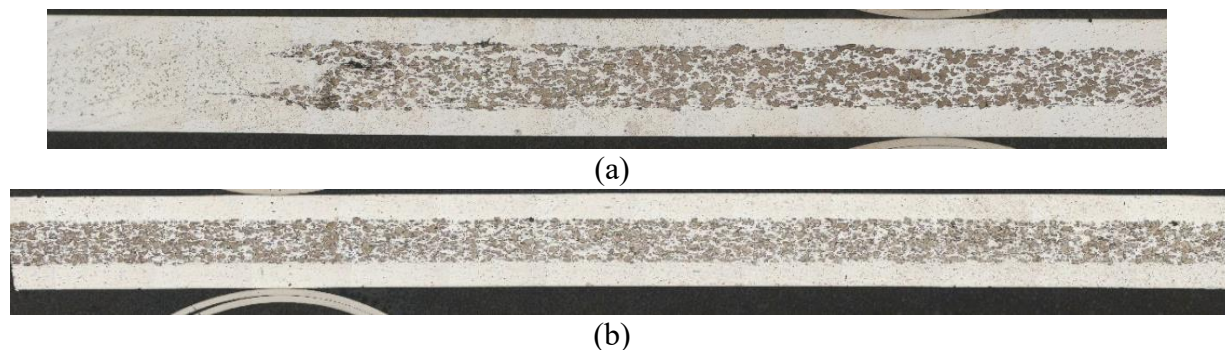
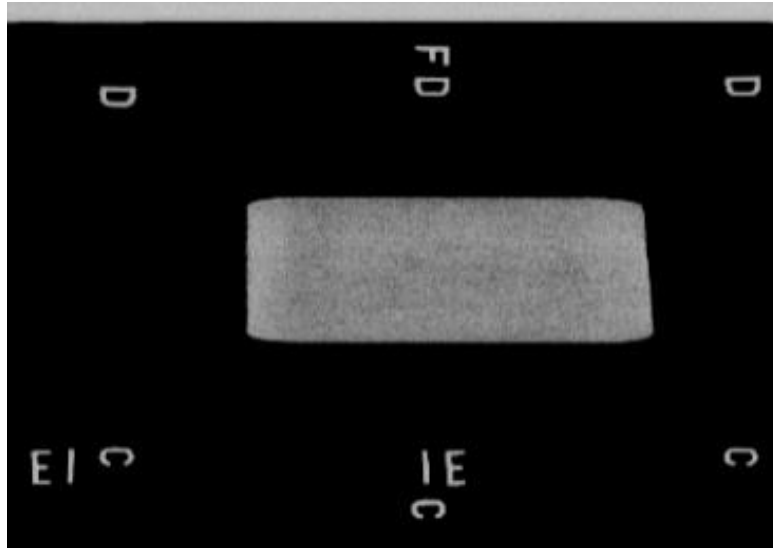


Figura 8 – Miniplates metallographic images: ULT (a); CL (b)

The plates were radiographed before final cutting, both for meat location and for verification of possible internal defects in it, such as cracks and lack of homogeneity in the uranium distribution. A typical image of a radiography can be seen in figure 9. The image have both whitish longitudinal ends, consequence of the dog bonning defect that increases the thickness of the edges.

The plates had the following average dimensions after the final cut: 1.50 mm thick, 46.31 mm wide and 170.58 mm long. The meat being 41.49 mm wide and 118.24 mm long, which within the values specified in figure 2.



**Figure 9: Miniplates radiograph before cut**

Finally, figure 10 presents the final set of miniplates produced through the whole process described in this work.



**Figure 10: Miniplates**

#### **4. CONCLUSIONS**

The production process for the manufacturing of uranium silicide fuel plate developed by the Nuclear Fuel Center of IPEN is applicable to the production of new fuels based on molybdenum uranium alloy.

The difference between the hardness of the silicide and the U7Mo alloy is responsible for the major changes in the production process of the fuel. Opting for the HMD route for material powdering, IPEN still needs to define parameters process for qualification and reproducibility.

The fusion step of the U7Mo alloy itself may be decisive for the hydriding step. As it was observed, the behavior of two pieces of the same ingot obtained very different results during

the hydriding process. It is suggested a study of the influence of microstructural and superficial characteristics of the ingot in the hydriding process.

Although studies show that pure U7Mo is not stable under irradiation, the UMo alloys are the most promising materials for obtaining a fuel with uranium density above 6%. So, defining conditions for its process is an important step for all new fuel development.

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