Effects of heat treatments on the thermal diffusivity of Uranium-Molybdenum alloy

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Abstract: U-Mo alloys are the most investigated nuclear fuel material to be used in research reactors. The addition of molybdenum stabilize the gamma phase of uranium and increase its melting point. A research program under development at CDTN aims the obtaining of uranium-molybdenum alloys to enable the high enriched uranium (HEU) to low enriched uranium (LEU) conversions. U-Mo ingots with 10 wt.% were induction melted and heat treated at 300 °C for 72 h, 102 h and 240 h. Thermal diffusivity was determined by the laser flash method and thermal quadrupole method, from room temperature to 300 °C and 400 °C.

Keywords: Uranium-molybdenum; laser flash method; thermal quadrupole method; thermal diffusivity.

1. INTRODUCTION

A research program is in progress at CDTN - *Centro de Desenvolvimento da Tecnologia Nuclear*, to develop a fuel consisting of a metallic alloy of uranium and molybdenum, that may eventually in future be used in the RMB-Multipurpose Brazilian Reactor, and which will enable the use of low enriched uranium (LEU). The use of metallic uranium is not feasible due to the occurrence of intense swelling related to the presence of the α-phase with orthorhombic crystal structure [1], the uranium stable phase at room temperature.

The uranium-molybdenum system was initially investigated in 1945 by Ahmann, Snow and Wilson [2] and have been one of the most investigated due to the retention capacity of the γ-phase, which rises as the Mo content is increased [3], and their higher densities. The aim of the present work is to investigate the thermal diffusivity of U-Mo with Mo content of 10% by weight, at room temperature, 300 °C and 400 °C.

2. MATERIALS AND METHODS

2.1. Production of the alloys

U-10 wt% Mo alloys were melted in an induction furnace under protective atmosphere of argon. Pieces of uranium and molybdenum were placed in a graphite crucible and, after purging under vacuum, the protective atmosphere of argon was established and the furnace was heated up to a temperature of about 1500 °C. This temperature was maintained for 12 min to provide the complete fusion and homogenization of the two elements. This melted alloy was poured into a cylindrical mold of copper cooled in air, resulting on U-Mo ingots with diameter of 25 mm and length of 150 mm [4-6].

Discoid samples with 8 mm diameter and 2,5 mm thickness were prepared from these ingots and its thermal diffusivity were determined by a laser flash method at room temperature and thermal quadrupole method at 300 °C and 400 °C.

2.2 Density determination of the alloys

The density ρ ($g \cdot cm^{-3}$) of each sample was determined geometrically from measurements of its diameter *D* (cm), its thickness *L* (cm) and its mass *m* (g) using a micrometer and a calibrated analytical balance, by means of the following equation (1):

$$\rho = \frac{m}{\frac{\pi \cdot D^2 \cdot L}{4}}$$

2.3 Thermal diffusivity determination of the alloys

2.3.1 Laser flash method

For thermal diffusivity measurements at room temperature, the laser flash method [7] was adopted using a bench developed by CDTN researchers, and applied according to ASTM-E-1461-11 [8]. Figure 1 presents schematically the experimental apparatus of CDTN for thermophysical properties measurements based on the Laser Flash Method.

By this method, the front face of a small disk-shaped sample is subjected to a very short burst of radiant energy. The source of the radiant energy is a CO_2 laser and the irradiation times are of the order of milliseconds. The resulting temperature rise of the rear surface of the sample is registered and from the obtained thermogram, the sample thermal diffusivity is calculated by the following equation (2):

$$\alpha = \frac{1 \cdot 37 \cdot L^2}{\pi^2 \cdot t_{1/2}}$$

Where α is the sample thermal diffusivity ($m^2 \cdot s^{-1}$), *L* is the sample thickness (m) and $t_{1/2}$ is the half time (s).

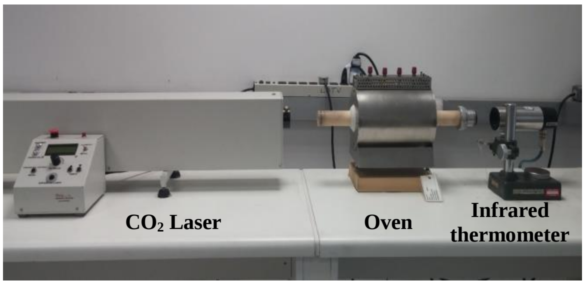

Figure 1. Apparatus for thermophysical properties measurements.

The used sealed CO_2 laser (10,6 µm wave length), model BL 80i provided by Bioluz Company has a maximal power output of 100 W and it is usually set to limit the sample rear face temperature rise in a maximum of 3 °C. The temperature of the sample rear face is measured by an infrared thermometer, model MMLTSSF1L provided by Raytek. For the processing of the thermometer signal, the data acquisition and calculations it is used the LabVIEW plataform and a 16 bits A/D card. The in house made sample oven has a heat element of Platinum/ 30 % Rodium that can be heated up to 1700 °C, but the diffusivities measurements were made at room temperature.

2.3.2 Thermal quadrupole method

The thermal quadrupole method was developed by Degiovanni [9] and can be seen as an extension of the concept of thermal resistance at steady state to transient conditions. It is an analytical method whose resolution is approximated by the least squares method, leading to identification of the thermal diffusivity. The sample gets a pulse of light on one side, raising its temperature. From the temperature field over the distance, it is possible to compare the theoretical curve and the experimental curve by the method of least squares. The modeling for the method is illustrated in figure 2 [9,10].

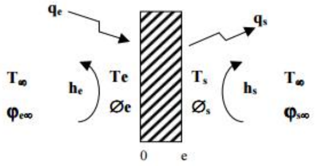

Figure 2. Heat transfer through a unidirectional plane wall.

The corresponding equations are (2):

$$\varphi_e = -\lambda \cdot \frac{\partial T}{\partial x}|_0 = q_e - h_e \cdot S \cdot (T_e - T_\infty)$$

$$\varphi_s = -\lambda \cdot \frac{\partial T}{\partial x}|_0 = q_s + h_s \cdot S \cdot (T_s - T_\infty)$$

Where: φ corresponds to the density of heat flow per unit of time, λ is the thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$), T is the temperature (K), S is the surface (m), h is the heat exchange coefficient ($W \cdot m^{-2} \cdot K^{-1}$) and q is the heat flow ($W \cdot m^{-2}$).

To determine the thermal diffusivity, it was used the diffusivimeter Protolab model QuadruFlash 1200 (figure 3) of Thermophysical Properties Measurement Laboratory (LMPT) of CDTN. The thermal diffusivity was measured 10 times for each sample, and the data averaged.

This diffusivimeter, manufactured in Brazil, is constituted by a xenon lamp (1200 J), responsible for the energy pulse, three K-type thermocouples of special class, an InSb infrared detector, an oven for heating the sample and a signal processing unit. A short thermal excitation is generated by the xenon lamp. The flash is directed inside a resistive furnace, allowing heating of the tested specimen from room temperature to 1200 ^oC and the temperature rise of the specimen is measured by three K-type thermocouples.

Figure 3. Diffusivimeter QuadruFlash 1200.

3. RESULTS AND DISCUSSION

Table 1 shows the results of the density and thermal diffusivity of the U-10 wt.% Mo alloys.

Table 1. Thermal diffusivity and density of the samples.

U-10%Mo	Density /$g \cdot cm^{-3}$	Thermal Diffusivity $x10^{-6} m^2 \cdot s^{-1}$ Temperature /^oC		
		25	300	400
As cast	16.38	3.90	7.72	8.22
300 ^oC 72h	16.45	3.83	7.74	8.17
300 ^oC 120h	16.26	4.18	7.27	8.29
300 ^oC 240h	16.19	4.17	7.43	8.39

There was no significant variation in the densities of the specimens, remaining around 16 $g \cdot cm^{-3}$. From room temperature to 300 ^oC and 400 ^oC, the thermal diffusivity increased significantly, but from 300 ^oC to 400 ^oC it increases modestly for all the samples. The results obtained in this study are similar to the work of Burkes et al. [11] at both temperatures evaluated.

It can be seen that the heat treatment at 300 ^oC for 72 h, 120 h and 240 h do not have major influence at the results. Calculated average thermal diffusivity as a function of temperature for the U-10 wt.% Mo alloy is provided in figure 4. The maximum standard deviation obtained was 9 %.

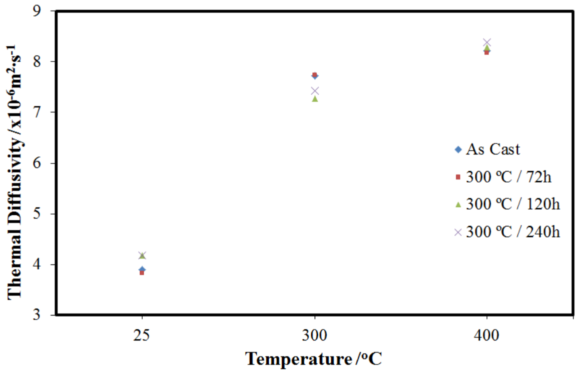

Figure 4. Average thermal diffusivity as a function of temperature of U-10 wt.% Mo alloys.

It is clearly seen that the increase of temperature increases the thermal diffusivity of the U-10 wt.% Mo alloy.

4. CONCLUSIONS

Thermophysical properties are an important role of any fuel development campaign. The present work investigated the thermal diffusivity of U-Mo with Mo content of 10% by weight, at room temperature, 300 °C and 400 °C.

It can be seen that the thermal diffusivity tends to increase with increasing temperature. Further studies may be done to evaluate different temperatures as well as heat treatments, to improve the thermophysical properties of U-10 wt.% Mo alloys.

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