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THERMAL DIFFUSIVITY OF URANIUM DIOXIDE FUEL PELLETS WITH ADDITION OF BERYLLIUM OXIDE BY THE LASER FLASH METHOD

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Abstract – A research program is in progress at CDTN – *Centro de Desenvolvimento da Tecnologia Nuclear*, aiming the development of fuel pellets made from a mixture of uranium dioxide microspheres or powder and beryllium oxide powder for increasing the thermal conductivity of the nuclear fuel. This type of fuel promises to be safer than current fuels, improving the performance of the reactor. In previous studies conducted at CDTN, fuel pellets were manufactured and tested considering the following weight percentages: 1, 2, 3, 5, 7 and 14wt.% of beryllium oxide (BeO) in mixture with microspheres of UO_2. Continuing this project, new uranium dioxide fuel pellets with addition of 2wt.% of BeO were manufactured and tested. The Laser Flash Method was employed to investigate the variation of thermal diffusivity at room temperature on those new pellets. The results obtained showed an increase of 12% in the thermal diffusivity with addition of 2wt.% of BeO in the uranium dioxide fuel pellets.

Keywords**:** Nuclear fuel pellets, beryllium oxide, thermal diffusivity, laser flash method.

1. INTRODUCTION

The uranium dioxide is the most used substance as nuclear reactor fuel due to its advantages such as: high stability, even when eventually comes into contact with water at high temperatures, high melting point and high retention capacity of fission products. The conventional fuel is made with ceramic sintered pellets of uranium dioxide stacked inside fuel rods. Its low thermal conductivity is a disadvantage causing premature degradation of the fuel due to the large pellet temperature gradient. Besides, the thermal conductivity decreases further as the fuel burns, limiting the fuel operational lifetime. A research program is in progress at CDTN to develop a new alloy for fuel pellets using uranium dioxide kernels or powder and beryllium oxide [1-3]. This fuel has as great advantage: a higher thermal conductivity in relation to the conventional fuel, leading an increasing of fuel lifetime and an interesting gain on economic aspects [4-7].

Thus, an assess of improvements can be performed by comparison of the thermal diffusivity of pure uranium dioxide samples, and samples of uranium dioxide (UO_2) with addition of 2wt.% of beryllium oxide, produced with the same batch of kernels and powder. This paper complements the previous experiments and presents the obtained results of thermal diffusivity at room temperature of these fuel pellets.

2. METHODOLOGY

2.1. Fuel Pellets Fabrication with Uranium Dioxide Kernels

The uranium dioxide kernels were produced by the sol-gel process developed by Nukem/Germany that was absorbed, transferred and implemented at the CDTN nuclear fuel laboratory [8]. The process was developed to fabricate fuel elements for high temperature gas cooled reactors and adapted in order to fabricate fuel pellets for pressurized water reactors too [3,9]. By this process, a uranium nitrate solution is transformed into spherical droplets that become hard in reaction with ammonium gas and are collected in an ammonium hydroxide solution (Fig. 1). In the subsequent steps, the kernels are washed, dried, calcined and sintered. To extract the ammonium nitrate, several washings are made in equipment that revolves the kernels intensely for one hour each washing, up to verify that there is no more ammonium nitrate in the served water, because the presence of ammonium nitrate in the kernels cause its destruction in the subsequent thermal treatment. The drying step was performed at 160 °C for 16 h and the calcination, at 800 °C for 3 h, followed by reduction at 650 °C for 4 h in hydrogen atmosphere, and by passivation under CO_2 atmosphere during the oven cooling down.

Beryllium oxide furnish by Sigma-Aldrich was mixed with the uranium dioxide kernels with a content of 2wt.%. To revolve the mixture for 2 h, it was used the same equipment that is used to wash the gel kernels after gelation. The kernels were pressed in pellets with a compaction pressure of 300 MPa using an especial model of hydraulic press developed at CDTN [3].

The green pellets height and diameter were measured by a micrometer (resolution: 10^{-3} mm) and the mass was obtained with an analytical balance (resolution: 10^{-4} g) in order to determine geometrically its green density. The pellets were sinterized at 1750 °C per 2 h (samples BE 010, BE 011 e BE 032) or 3 hours (samples BE 050, BE 051, BE 052 and BE 053) in an atmosphere of argon/hydrogen.

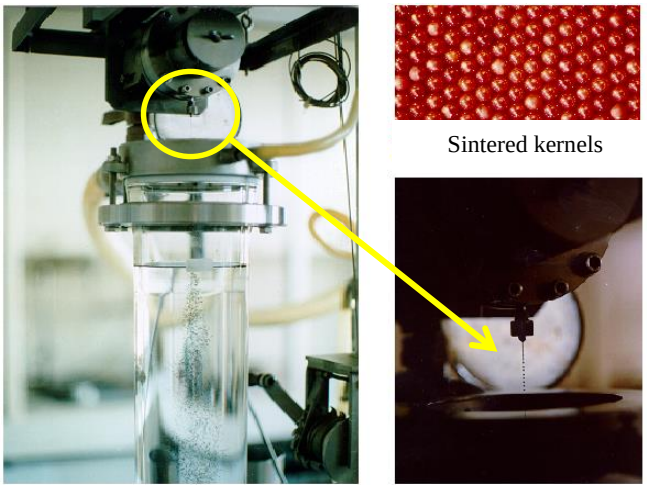

Fig. 1. Equipment for kernel production.

2.2. *Fuel Pellets Fabrication with Uranium Dioxide Powder*

Pellets of BeO in mixture with powder of UO_2 were produced by uniaxial pressing of the UO_2 powder homogeneously mixed with BeO powder at the same hydraulic press described in 2.1, but with a compaction pressure of 400 MPa. The green pellets height and diameter were also measured by a micrometer and the mass was obtained with an analytical balance in order to determine geometrically its green density. The sinterization process was the same as previously described: 1750 °C per 3 hours (samples BE 063, BE 064, BE 066 and BE 067) in an atmosphere of argon/hydrogen.

2.3. *Density determination*

The density ρ [g·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:

$$\rho = \frac{m}{\frac{\pi \cdot D^2 \cdot L}{4}} \qquad (1)$$

2.4. *Determination of thermal diffusivity*

For thermal diffusivity measurements, the laser flash method was adopted using a bench developed by CDTN researchers, and applied according to ASTM-E-1461-11 [10]. Fig. 2 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:

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

where:
α is the sample thermal diffusivity [m^2·s^{-1}];
L is the sample thickness [m];
$t_{1/2}$ is the half time [s].

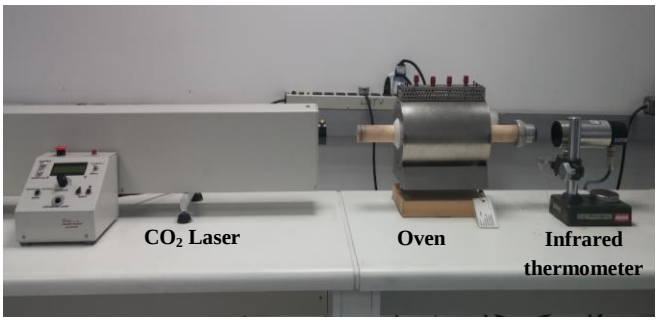

Fig. 2. Apparatus for thermophysical properties measurements.

The used sealed CO_2 laser (10.6 µm wave length), model BL 80i provided by Bioluz Company/Campinas/ São Paulo-SP, 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 ºC, but the diffusivities measurements were made at room temperature.

3. RESULTS

The table 1 shows the data (mass, dimensions, green density and sintered density) of the uranium dioxide fuel pellets and of the uranium dioxide pellets with addition of BeO. The results show some variations in the sintered density of the samples. These variations can be explained by changes on the fabrication procedure.

Table 2 shows the results of the thermal diffusivity measurements of the samples at room temperature. These values are an average of the obtained values.

When compared to standard UO_2 fuel, the uranium dioxide fuel pellets manufactured with a compaction pressure of 300 MPa and with addition of 2wt.% of BeO provides an increase in the thermal diffusivity of 12%. In the case of pellets manufactured with a compaction pressure of 400 MPa, the thermal diffusivity increased by 23%.

This was in fact the reason why this type of nuclear fuel was investigated again, considering the potential economic benefits. This increase is consistent with previous results. It can be seen that the pellets manufactured with powder of uranium dioxide and with a compaction pressure of 400 MPa exhibit better results of thermal diffusivity. These results can be explained by the higher compaction of the samples manufactured with powder instead of kernels.

Table 1. Data of the uranium dioxide pellets and uranium dioxide pellets with addition of BeO.

Sample	Mass g	Thickness cm	Diameter cm	Green density $g \cdot cm^{-3}$	Sintered Density $g \cdot cm^{-3}$
BE 010 300 MPa UO_2 kernels	1.87	0.36	1.11	5.37	9.88
BE 011 300 MPa UO_2 kernels	1.83	0.35	1.11	5.40	9.85
BE 032 300 MPa UO_2 kernels	1.69	0.26	1.11	5.35	9.56
BE 050 300 MPa UO_2 kernels	1.63	0.25	1.12	5.32	9.31
BE 051 300 MPa UO_2 kernels	1.57	0.24	1.12	5.22	9.41
BE 052 300 MPa UO_2 kernels +2wt.% BeO	1.49	0.24	1.13	4.95	8.80
BE 053 300 MPa UO_2 kernels +2wt.% BeO	1.57	0.26	1.13	4.90	8.66
BE 063 400 MPa UO_2 Powder	1.75	0.25	0.94	6.03	10.17
BE 064 400 MPa UO_2 Powder	1.65	0.23	0.94	5.77	10.22
BE 066 400 MPa UO_2 Powder +2wt.% BeO	1.75	0.26	0.94	5.59	9.66
BE 067 400 MPa UO_2 Powder +2wt.% BeO	1.74	0.26	0.94	5.67	9.73

Table 2. Thermal diffusivity of uranium dioxide pellet with addition of BeO, measured at room temperature.

Sample	Compaction pressure MPa	Thermal Diffusivity $x10^{-6}m^2 \cdot s^{-1}$
UO_2 Kernel	300	2.43
UO_2+2wt.%BeO Kernel+Powder	300	2.72
UO_2 Powder	400	2.94
UO_2+2wt.%BeO Powder+Powder	400	3.61

4. CONCLUSIONS

It was possible to obtain fuel pellets of UO_2 and fuel pellets of UO_2 with 2%wt. of beryllium oxide (BeO), and determine its thermal diffusivity using the laser flash method at the LMPT - *Laboratório de Medição de Propriedades Termofísicas* of CDTN. The obtained results show an increase of 12% on the thermal diffusivity with the addition of 2wt.% of BeO in the pellets manufactured with UO_2 kernels. This small increase in thermal diffusivity has a significant impact on the performance of commercial nuclear fuels due to increase in the thermal conductivity. Also, an increase of 23% on the thermal diffusivity was observed in the fuel pellets manufactured with UO_2 powder instead of kernels. These values are in good agreement with data existing in the literature.

In a next step it is planned to determine thermal diffusivity and thermal conductivity of these samples at higher temperatures and investigate the effect of the temperature in the thermal conductivity of these pellets.

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