



Effect of compaction pressure on the thermal conductivity of $\text{UO}_2\text{-BeO-Gd}_2\text{O}_3$ pellets

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ABSTRACT

The $(\text{U},\text{Gd})\text{O}_2$ fuels are used in pressurized water reactors (PWR) to control the neutron population in the reactor during the early life with the purpose of extended fuel cycles and higher target burnups. Nevertheless, the incorporation of Gd_2O_3 in the UO_2 fuel decreases the thermal conductivity, leading to premature fuel degradation. This is the reason for the addition of beryllium oxide (BeO), which has a high thermal conductivity and is chemically compatible with UO_2 . Pellets were obtained from powder mixtures of the UO_2 , Gd_2O_3 and BeO, being the oxide contents of the beryllium equal to 2 and 3wt%, and the gadolinium fixed at 6wt%. The pellets were compacted at 400, 500, 600, and 700 MPa and sintering under hydrogen reducing atmosphere. The purpose of this study was to investigate the effect of BeO, Gd_2O_3 and compaction pressure on the thermal conductivity of the UO_2 pellets. The thermal diffusivity and conductivity of the pellets were determined from 298 K to 773 K and the results obtained were compared to UO_2 fuel pellets. The thermal diffusivity was determined by Laser Flash and Thermal Quadrupole methods and the thermal conductivity was calculated from the product of thermal diffusivity, the specific heat capacity and density. The sintered density of the pellets was determined by the xylol penetration and immersion method. The results showed an increase in the thermal conductivity of the pellets with additions of BeO and with the compaction pressure compared to the values obtained with UO_2 pellets.

Keywords: thermal conductivity, nuclear fuel, uranium dioxide, beryllium oxide, gadolinium oxide

1. INTRODUCTION

One of the most important limiting factors in water nuclear reactor operation is the maximum temperature reached by the fuel, being a higher thermal conductivity of fuel essential for improving reactor performance under normal operation and accident conditions. The UO₂ pellet is most used fuel in LWR (light water reactor), but its low thermal conductivity in the range of 2-8 W·m⁻¹·K⁻¹ [1] has a direct influence on the thermal behavior of the fuel during reactor operation as center fuel temperature, thermal expansion, fission gas release, gaseous swelling, etc.

Since there is a great interest in improving the thermal conductivity on nuclear fuel, several researches are being carried out with the addition of oxide to increase the thermal conductivity of the UO₂ [2-7]. Researches to extent the fuel burnup are also conducted with the addition of oxide to UO₂. These processes of oxide addition are generally based on the mechanical mixing process because it is an easy step to implement in the nuclear industry conventional of UO₂ fuel.

Based on previous studies in the Nuclear Technology Development Center (CDTN) on the fuel compounds UO₂-BeO and UO₂-Gd₂O₃, the main additives employed here are BeO and Gd₂O₃, once the addition of small amounts of BeO significantly increases the fuel thermal conductivity [4,7] and Gd₂O₃ is widely used in the nuclear industry because it can compensate for the excess initial reactivity at the beginning of reactor life, promoting a higher fuel burnup [8,9].

However, the addition of Gd₂O₃ to UO₂ leads to a decrease in the thermal conductivity of the fuel, making the amount of gadolinium oxide added a limiting parameter of the fuel performance [8,9]. Beryllium and gadolinium oxides have excellent properties as a high melting point, good behavior under irradiation, chemical stability, etc.

Ishimoto *et al.* [4] showed significant improvements in the thermal conductivity of UO₂ that can be achieved with only 3.2 vol.% of BeO. Garcia *et al.* [6] demonstrated that UO₂ thermal conductivity over the temperature range of 298.15 K to 523.15 K was improved by approximately 10% for each 1 vol.% of BeO added. Our research [10,11] has shown that the addition of 2 and 3 wt% of BeO in UO₂ can lead to an increase in the thermal conductivity of 22% and 28% at 573 K, respectively.

The purpose of this paper is to investigate the effect of the compaction pressure on the thermal conductivity of the UO₂ pellets added with BeO and Gd₂O₃. All pellets were obtained based on the conventional fabrication process, such as mixing, pressing, and sintering under a reducing

atmosphere. The thermal diffusivity of the pellets was determined at 298 K by the Laser Flash method [12] and from 473 K to 773 K by the Thermal Quadrupole method [13]. The results for UO₂ and UO₂-BeO-Gd₂O₃ pellets such as density and thermophysical properties (thermal diffusivity, specific heat capacity and thermal conductivity) are reported. The expanded uncertainty was estimated according to the ISO/BIPM Guide to the Expression of Uncertainty in Measurement (GUM) [14].

2. MATERIALS AND METHODS

2.1. Obtaining and pellets characterization

The powders of BeO and Gd₂O₃ used in this work were supplied by Alfa Aesar (99.99% pure) and Sigma-Aldrich (99.98% pure), respectively, and the UO₂ powder was provided by Institute of Energy and Nuclear Research (IPEN).

The UO₂ and UO₂-BeO-Gd₂O₃ pellets were obtained by mixing of powders, pressing into green pellets and sintering under a hydrogen atmosphere. The powders of UO₂, BeO and Gd₂O₃ were mechanically homogenized for 4 h in a rotating apparatus for each content of BeO (2% and 3wt%) and 6wt% Gd₂O₃. These powders mixtures were compacted at 400, 500, 600, and 700 MPa utilizing a uniaxial hydraulic press. Then, the green pellets were sintered at 1700 °C for 3 h and presented final geometric dimensions of about 10 mm in diameter and 2-3 mm in thickness.

The density of the sintered pellets was determined by xylol penetration and immersion method [15] and the mass of the pellets was taken using a Mettler AT201 balance, which has a resolution of 0.1 mg. The dependence of the sintered density of UO₂-BeO-Gd₂O₃ pellets with temperature was determined from linear thermal expansion data of the UO₂.

2.2. Thermophysical properties

Thermal diffusivity of the pellets was determined by two methods: the Flash Laser [11] using an apparatus developed at CDTN [8] and the Thermal Quadrupole [13] employing a diffusivimeter (Protolab, QuadruFlash 1200), manufactured in Brazil. The Flash laser method was employed for measurements carried out at room temperature while the Thermal Quadrupole method was utilized in the temperature range of 473 to 773 K.

Before the measurements, the pellets were coated with a carbon film on both flat faces to improve the emissivity and uniform absorption of the laser beam. In both methods, the front surface of the pellets was subjected to a very short burst of radiant energy.

The thermal diffusivity results were normalized by the following equations [4]:

$$\alpha_{95} = \alpha_M \left[\left(\frac{1-0,05 \cdot \varepsilon}{1-\varepsilon \cdot P} \right) \cdot \left(\frac{1-P}{1-0,05} \right) \right] \quad (1)$$

$$\varepsilon = 2.6 - 5 \times 10^{-4} \cdot (T - 273.15) \quad (2)$$

where α_{95} corresponds to the thermal diffusivity normalized to 95% of theoretical density (TD), α_M to the determined thermal diffusivity, P to the pellet porosity and T to the temperature.

The specific heat capacity values were calculated by the law of mixing [4] using reported data of specific heat of the individual components [16-18]. The thermal conductivity of fuel pellets was determined by product of their thermal diffusivity, density and specific heat capacity.

3. RESULTS AND DISCUSSION

Table 1 shows the geometric dimensions and sintered densities of the pellets for each compaction pressure. It can be observed from this table that the sintered density of the pellets was between 94% TD and 96% TD, exception for only one UO₂-6wt%Gd₂O₃-3wt%BeO pellet pressed at 700 MPa. The maximum expanded relative uncertainty of sintered density pellets was estimated in 2%, for a coverage probability of approximately 95%, k=2. Furthermore, there is an increase in the theoretical density of pellets with the increase compaction pressure.

Table 1: Geometric dimensions and density of the sintered pellets related to compaction pressure.

Pellet composition	Compaction pressure (MPa)	Thickness (cm)	Diameter (cm)	Sintered Density (g·cm ⁻³)	%TD
UO ₂	400	0.25	0.93	10.32	94
		0.24	0.93	10.34	94
	500	0.24	0.94	10.36	95
		0.24	0.94	10.47	96
	600	0.23	0.94	10.55	96
		0.23	0.94	10.56	96
	700	0.23	0.96	10.54	96
		0.23	0.95	10.45	95
	400	0.26	0.94	9.50	94
		0.27	0.94	9.52	94
UO ₂ +6wt%Gd ₂ O ₃ +2wt%BeO	500	0.25	0.94	9.59	95
		0.26	0.94	9.67	96
	600	0.25	0.95	9.65	95
		0.25	0.95	9.73	96
	700	0.25	0.96	9.59	95
		0.25	0.97	9.59	95
	400	0.27	0.94	9.34	94
		0.27	0.94	9.33	94
	500	0.26	0.94	9.45	96
		0.26	0.94	9.43	95
UO ₂ +6wt%Gd ₂ O ₃ +3wt%BeO	600	0.26	0.94	9.37	95
		0.26	0.95	9.40	95
	700	0.26	0.96	9.24	93
		0.27	0.96	9.44	96

The determined specific heat capacity of the pellets is presented in Table 2, where it can see an increase of the specific heat capacity of the pellets as a function of the BeO content, as expected. The expanded uncertainty adopted for the specific heat was assumed to be 2% [1].

Table 2: Specific heat capacity of the pellets.

Pellet composition	Specific heat capacity (J·kg ⁻¹ ·K ⁻¹)				
	298 K	473 K	573 K	673 K	773 K
UO ₂	235	279	290	297	303
UO ₂ +2wt%BeO+6wt%Gd ₂ O ₃	254	306	319	328	335
UO ₂ +3wt%BeO+6wt%Gd ₂ O ₃	262	318	332	342	350

Table 3 and Table 4 show thermal diffusivities as well as the thermal conductivities, both normalized to 95% TD as a function of temperature for all compaction pressures. For these results, the expanded relative uncertainty was estimated at 7.5% for thermal diffusivity and 8.5% for thermal conductivity. It is known that the addition of gadolinia to UO₂ decreases the thermal diffusivities and thermal conductivities [8], as observed in Table 3 and Table 4, respectively.

Table 3: Normalized thermal diffusivity of the pellets.

Pellet composition	Compaction pressure (MPa)	Normalized thermal diffusivity (mm²·s⁻¹)				
		298 K	473 K	573 K	673 K	773 K
UO₂	400	2.87	2.02	1.73	1.58	1.42
		2.88	1.98	1.69	-	-
	500	2.79	1.99	1.68	1.52	1.37
		2.73	1.95	1.67	1.51	-
	600	2.79	1.91	1.65	1.51	1.34
		2.74	-	-	-	-
	700	2.74	1.98	1.78	1.61	1.44
		2.78	-	-	-	-
	400	2.14	1.68	1.46	1.39	1.28
		2.23	1.69	1.51	1.38	1.30
UO₂+2wt%BeO+6wt%Gd₂O₃	500	2.19	1.71	1.48	1.36	1.23
		2.17	-	-	-	-
	600	2.14	1.71	1.47	1.34	1.27
		2.13	-	-	-	-
	700	2.19	1.65	1.49	1.32	1.27
		2.16	-	-	-	-
	400	2.35	1.74	1.53	1.44	1.36
		2.34	1.73	1.57	1.46	1.34
	500	2.29	1.69	1.52	1.41	1.29
		2.31	-	-	-	-
UO₂+3wt%BeO+6wt%Gd₂O₃	600	2.34	1.77	1.55	1.48	1.35
		2.30	-	-	-	-
	700	2.69	2.00	1.71	1.62	1.46
		2.56	-	-	-	-

Table 4: Normalized thermal conductivity of the pellets.

Pellet composition	Compaction pressure (MPa)	Normalized thermal conductivity (W·m ⁻¹ ·K ⁻¹)				
		298 K	473 K	573 K	673 K	773 K
UO ₂	400	6.97	5.79	5.14	4.80	4.38
		7.00	5.70	5.03	-	-
	500	6.81	5.73	5.00	4.62	4.24
		6.73	5.68	5.02	4.65	-
	600	6.93	5.59	5.02	4.68	4.24
		6.80	-	-	-	-
	700	6.78	5.79	5.40	5.00	4.54
		6.82	-	-	-	-
	400	5.18	4.86	4.40	4.29	4.02
		5.41	4.88	4.55	4.26	4.10
UO ₂ +2wt%BeO+6wt%Gd ₂ O ₃	500	5.33	5.00	4.50	4.24	3.90
		5.33	-	-	-	-
	600	5.24	5.01	4.48	4.21	4.05
		5.26	-	-	-	-
	700	5.34	4.83	4.52	4.11	4.04
		5.26	-	-	-	-
	400	5.74	5.15	4.70	4.55	4.40
		5.70	5.11	4.84	4.61	4.33
	500	5.68	5.05	4.75	4.50	4.22
		5.72	-	-	-	-
UO ₂ +3wt%BeO+6wt%Gd ₂ O ₃	600	5.74	5.24	4.80	4.69	4.38
		5.66	-	-	-	-
	700	6.51	5.85	5.22	5.08	4.67
		6.33	-	-	-	-

Results of normalized thermal conductivity of the pellets pressed at 400 MPa were in the range from 7.00 up to 4.38 W·m⁻¹·K⁻¹ for UO₂; from 5.41 up to 4.02 W·m⁻¹·K⁻¹ for UO₂-2%BeO-6%Gd₂O₃ and from 5.74 up to 4.33 W·m⁻¹·K⁻¹ for UO₂-3%BeO-6%Gd₂O₃ throughout the measured temperature range. The highest percentage deviation from normalized thermal conductivity of the pellets of the same composition pressed at 400 MPa in relation to other compactions pressures was 5%. Only for pellets with 3 wt% BeO pressed at 700 MPa showed a deviation higher than 14%. An increase of only 1wt% in BeO in the case of pellets pressed at 400MPa caused an increase in normalized thermal conductivity measured at room temperature and up to 773 K by 8% and 5%, respectively. Table 4

also includes some results of the replicated pellets which indicated a good reproducibility of the process of obtaining pellets.

Characteristic graphs of thermal conductivities are shown from Fig. 1 to Fig. 3, where it can be seen there is a gradual increase of the thermal conductivity with increasing BeO content and a decrease in these values with increasing temperature that followed the same trend observed for thermal diffusivity not shown here.

Figure 1: Normalized thermal conductivity of 95% TD of UO_2 pellets in function of the compaction pressure.

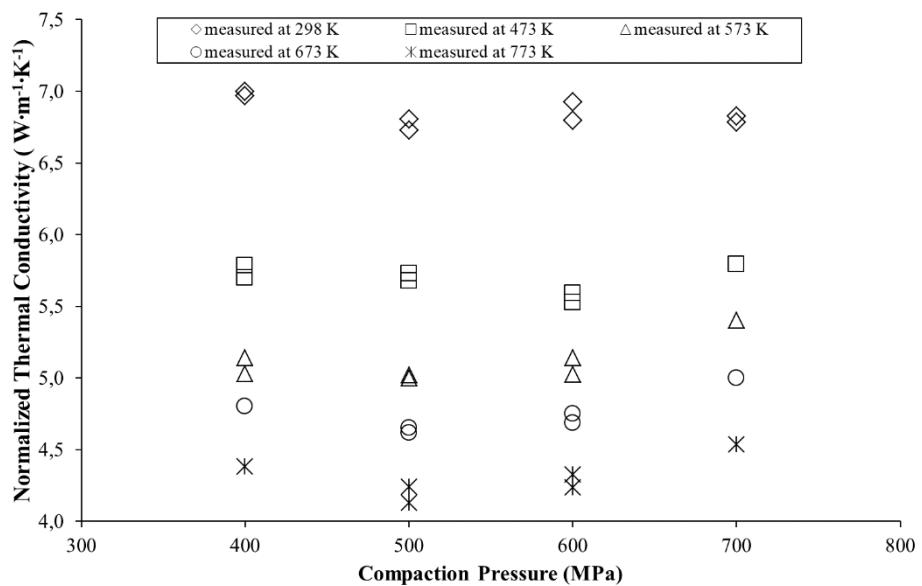


Figure 2: Normalized thermal conductivity of 95% TD of UO_2 -2%BeO-6% Gd_2O_3 pellets in function of the compaction pressure.

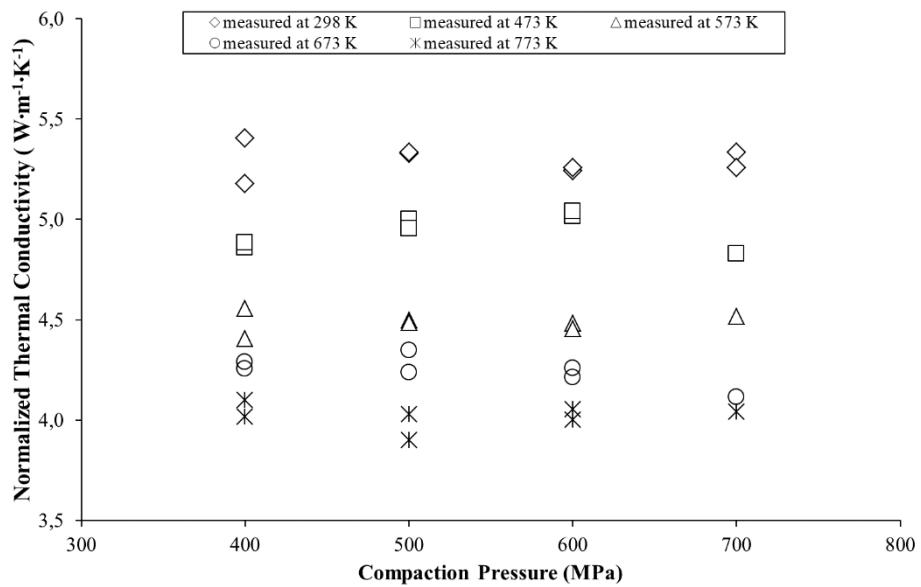
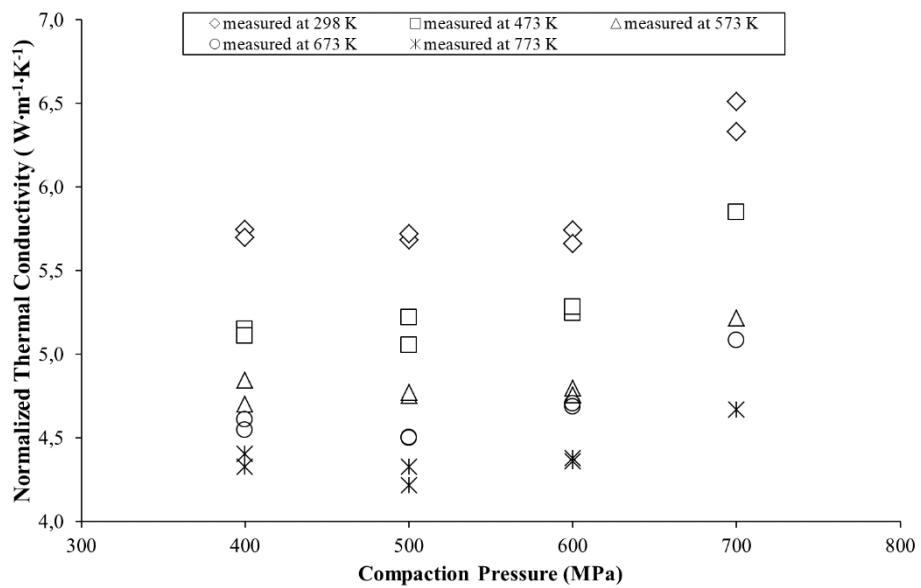


Figure 3: Normalized thermal conductivity of 95% TD of UO_2 -3%BeO-6% Gd_2O_3 pellets in function of the compaction pressure.



4. CONCLUSION

To improve the performance of nuclear fuel, the thermal conductivity of UO₂ pellets was investigated with the incorporation of additives in the form of beryllium and gadolinium oxides. In addition to the beryllium content, the compaction pressures of the UO₂ and UO₂-xBeO-6Gd₂O₃ pellets ($x = 1\text{wt\%}$ and 2wt\%) varied from 400 to 700 MPa. The additions of Gd and Be had the objective of prolonging the burnup time of UO₂ and attenuating the decrease in the thermal conductivity of UO₂ caused by the presence of gadolinium, respectively. The comparison between the values of thermal conductivity obtained in the pellets with mixed oxide to UO₂ for all compaction pressures indicated that BeO increased the thermal conductivity and between pressures of 500 and 700 MPa there was no the significative difference. There is an increase in thermal conductivity from 8% at room temperature and 5% up to 773K, for the compaction pressure of 400 MPa considering the temperature range measured when comparing insert data with 2 and 3% by weight of BeO.

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