

URANIUM TETRAFLUORIDE THERMOPHYSICAL PROPERTIES MEASUREMENTS BY THE LASER FLASH METHOD

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ABSTRACT

In order to increase the efficiency of the metalthermic reduction process employed by the IPEN-*Instituto de Pesquisas Energéticas e Nucleares*, to produce metallic uranium by means of uranium tetrafluoride reduction with magnesium, is planned utilize a mathematical model that numerically simulates the heat distribution in the charge of powders of uranium tetrafluoride and metallic magnesium duly blended. Because this program requires as input data the thermal conductivity of the uranium tetrafluoride powder and this data was not found in the literature, pellets were produced by uniaxially pressing the uranium tetrafluoride powder in pellets, employing the same technology developed at CDTN-*Centro de Desenvolvimento da Tecnologia Nuclear*, to produce nuclear fuel pellets. The thermophysical properties of these pellets were measured at the LMPT - *Laboratório de Medição de Propriedades Termofísicas de Combustíveis Nucleares e Materiais* of CDTN applying the laser flash method. From the measurements results, it was possible to obtain an extrapolated value of ca. $0.24 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for the thermal conductivity of the UF_4 powder sample in the loose state and at room temperature. This value will be employed as input data of the mathematical model, whose results will be subject of a future publication. The UF_4 powder pelletizing parameters and the thermophysical properties results (density, thermal diffusivity and thermal conductivity), are reported.

1. INTRODUCTION

The production of metallic uranium at IPEN-*Instituto de Pesquisas Energéticas e Nucleares* is performed by means of a metalthermic reduction process, whereby uranium tetrafluoride

powder is blended together with metallic magnesium powder and charged in a graphite crucible. Fluorite is added in order to complete the open space. The crucible is put inside a stainless steel reactor and placed in a soaking pit furnace and then heated up 640°C, when magnesiothermic ignition happens promoting the formation of molten metallic uranium. The ingot is formed in the crucible bottom, covered basically by MgF₂ slag [1,2,3].

It is planned to study the process by means of a mathematical model that numerically simulates the distribution of heat in the charge of powders of uranium tetrafluoride and metallic magnesium, aiming to increase the efficiency of the ignition process during reaction. Because this program requires as input data the thermal conductivity of the uranium tetrafluoride powder, pellets were produced by uniaxially pressing the powder in pellets, in order to measure its thermophysical properties and so to obtain the thermal conductivity of the uranium tetrafluoride powder. Thermal conductivities for UF₄, with and without Mg blendings have been presented by Beltrán [4], suggesting values from 0.6 to 1.2 W·m⁻¹·K⁻¹ for pure UF₄ and from 0.9 to 2.1 W·m⁻¹·K⁻¹ for UF₄/Mg blendings, perhaps measured in a loosely pressed material, without information about the measuring technique.

The UF₄ powder pelletizing parameters and the thermophysical properties (density, thermal diffusivity and thermal conductivity) results are reported.

2. METODOLOGY

2.1. UF₄ Pellets Fabrication

The material was pressed in pellets using an especial model of hydraulic press, whose cycle is shown in Fig. 1 [5]. The cylindrical die used has a diameter of 11.1 mm and the system works with a floating die in order to obtain double effect. To avoid lamination due to the pellet respiration during the extraction, the pellet is extracted under a reduced pressure, and the die has a conicity in its opening. The die is automatically lubricated by the lower punch, but in the present case, it was not used lubricant. Three pellets were manufactured at compaction pressures of 400, 600 and 800 MPa.

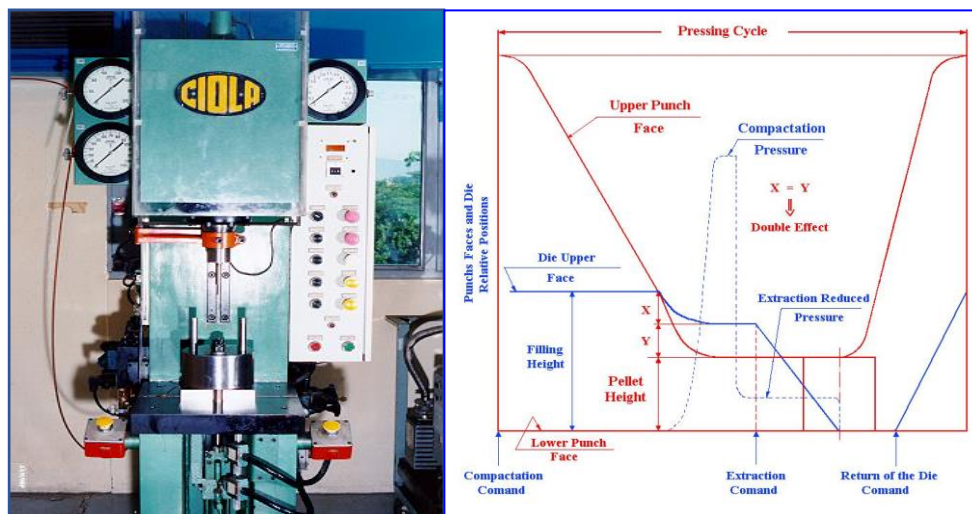


Figure 1. Hydraulic press for fuel pellet fabrication and pressing cycle.

2.2. UF₄ Pellets Green Density Measurement

The green density of each pellet was determined geometrically by using the following equation:

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

where:

ρ : green pellet density [g·cm⁻³]

M : green pellet mass [g]

D : green pellet diameter [cm]

L : green pellet thickness [cm]

2.3. Thermophysical Properties Measurements - Laser Flash Method

For the pellets thermal diffusivity measurement were utilized the laser flash method [6, 7], and the in house made bench shown in Fig. 2.



Figure 4. In house made bench for the thermophysical properties measurements.
LMPT – Laboratório de Medição de Propriedades Termofísicas de Combustíveis Nucleares e Materiais.

In 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 usually a laser or a xenon flash lamp and irradiation times are of the order of milliseconds (Figure 3).

The resulting temperature rise of the rear surface of the sample is registered and from the obtained thermogram, the thermal diffusivity is calculated by the following equation:

$$\alpha = \frac{1.37 \cdot L^2}{\pi^2 \cdot t_{1/2}} \quad (2)$$

where:

α is the sample thermal diffusivity [m²·s⁻¹].

L is the sample thickness [m].

$t_{1/2}$ is the half time [s].

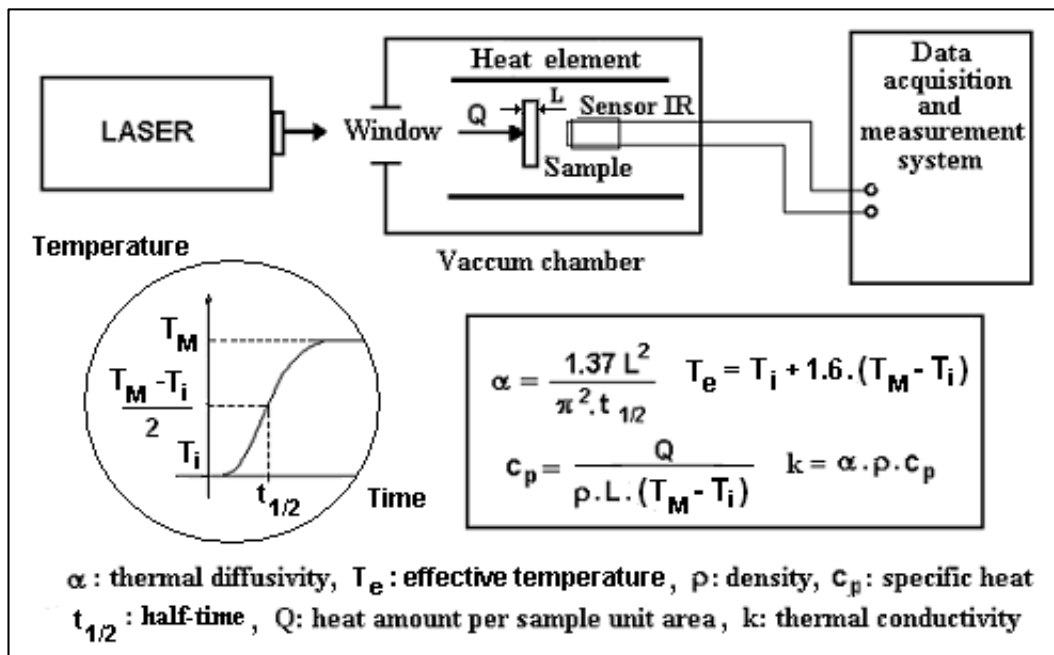


Figure 3. Principle of the Laser Flash Method.

The used CO₂ laser has a maximal power output of 100 Watts and the temperature of the sample rear face is measured by an infrared thermometer. The sample oven has a heat element of platinum / 30%Rodium that can be heated up to 1700°C, but the measurements were made from room temperature up to ca. 177°C. The thermal conductivity k [W·m⁻¹·K⁻¹] of each pellet was calculated by the following equation:

$$k = \alpha \cdot \rho \cdot C_p \quad (3)$$

where C_p is the specific heat of the material [J·kg⁻¹·K⁻¹], and ρ its density [·kg·m⁻³].

3. RESULTS

Table 1, 2, 3 and 4 show the pelletizing parameters and the obtained thermophysical properties for the three pellets.

To the thermal conductivity calculations, considering that the temperature range is narrow, for the specific heat of the material it is used a mean value of 228.8 J·kg⁻¹·K⁻¹, from a total of 45 measurements results, obtained by applying a model proposed by Grossi [8], to the laser flash method. For the sample density it was used the green pellet density ρ values.

Fig. 4 shows the pressing characteristic curve of the material. In Figure 5 it is observed the discrete decrease of the UF₄ thermal diffusivity with the increase of the effective temperature, and Figure 6 and 7, the increase of the thermal diffusivity with the increase of the compaction pressure and the increase of the pellet green density, respectively (as expected). Figures 8 and 9 show the increase of the thermal conductivity with the increase of the compaction pressure and the increase of the pellet green density, respectively, and Figure

10, the discrete decrease of the UF₄ thermal conductivity with the increase of the effective temperature.

Table 1. UF₄ pelletizing parameters and obtained pellets green densities and dimensions.

Pellet Number	Material	Compaction Pressure MPa	Green pellet mass g	Green pellet diameter cm	Green pellet thickness cm	Pellet green density g·cm ⁻³
1	UF ₄	400	1.2232	1.118	0.2688	4.64
2	UF ₄	600	1.4241	1.118	0.2894	5.01
3	UF ₄	800	1.5511	1.118	0.2998	5.27

Table 2. UF₄ number 1 pellet thermophysical properties.

Sample Initial Temperature °C	Rear Face Temperature Excursion °C	Effective Temperature °C	Thermal diffusivity x10 ⁶ m ² ·s ⁻¹	Thermal Conductivity W·m ⁻¹ ·K ⁻¹
23.2	4.2	30	0.3225	0.342
95.7	3.7	102	0.3116	0.331
170.5	3.8	177	0.3050	0.324

Table 3. UF₄ number 2 pellet thermophysical properties.

Sample Initial Temperature °C	Rear Face Temperature Excursion °C	Effective Temperature °C	Thermal diffusivity x10 ⁶ m ² ·s ⁻¹	Thermal Conductivity W·m ⁻¹ ·K ⁻¹
18.2	3.6	24	0.3437	0.394
93.5	3.5	99	0.3381	0.387
172.5	3.0	177	0.3318	0.380

Table 4. UF₄ number 3 pellet thermophysical properties.

Sample Initial Temperature °C	Rear Face Temperature Excursion °C	Effective Temperature °C	Thermal diffusivity x10 ⁶ m ² ·s ⁻¹	Thermal Conductivity W·m ⁻¹ ·K ⁻¹
20.4	3.0	25	0.3644	0.439
95.9	2.8	100	0.3485	0.420
171.5	2.7	176	0.3480	0.419

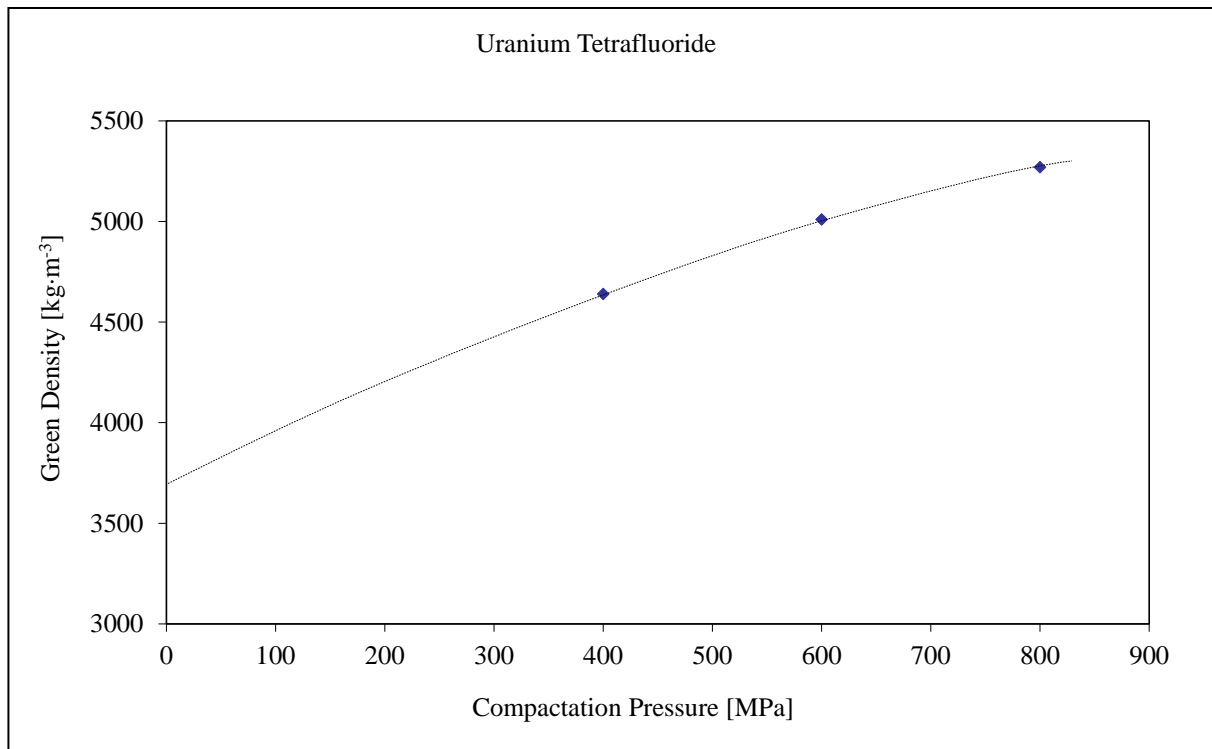


Figure 4. Pressing characteristic curve of the UF₄ powder.

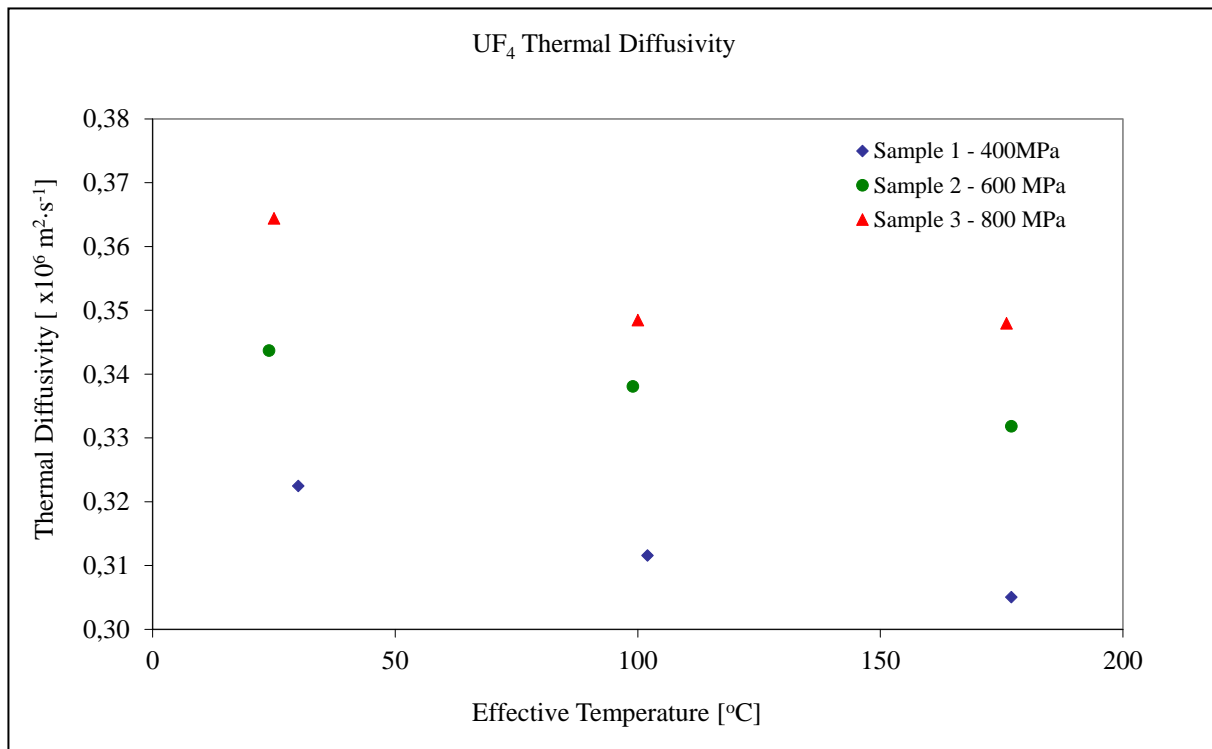


Figure 5. Discrete decrease of the UF₄ thermal diffusivity with the increase of the effective temperature.

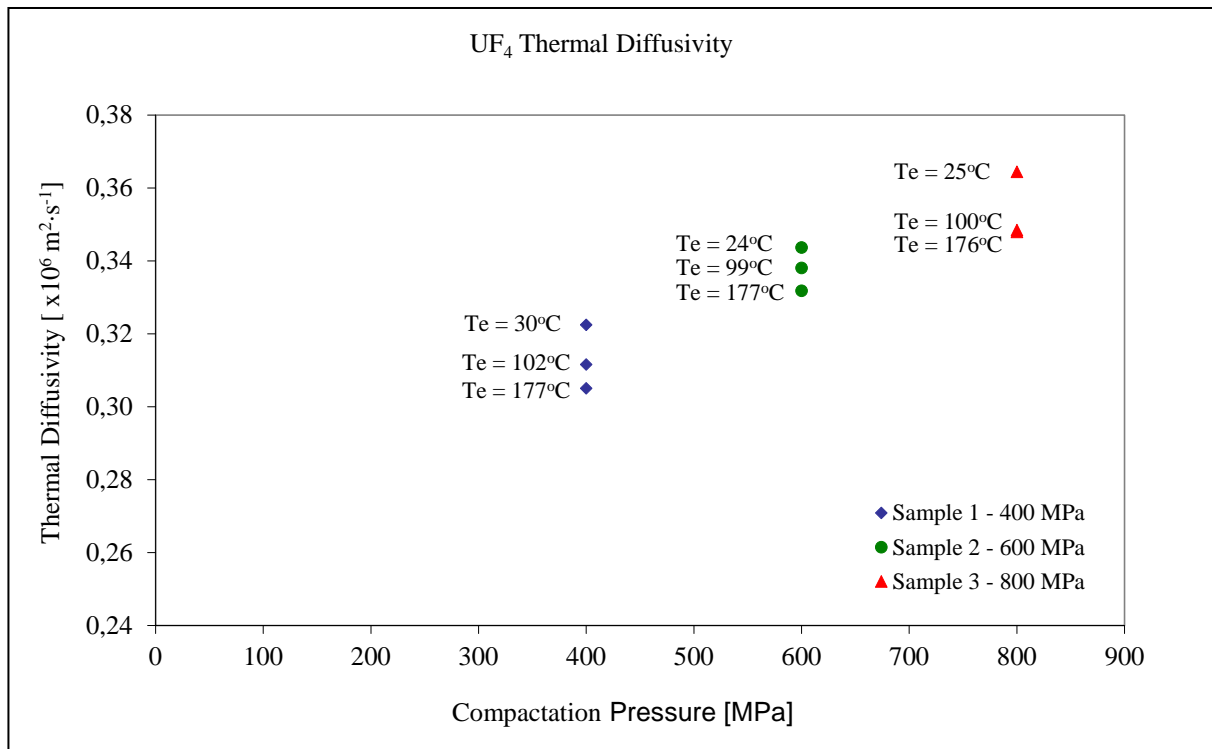


Figure 6. Increase of the UF₄ pellets thermal diffusivity with the increase of the compaction pressure.

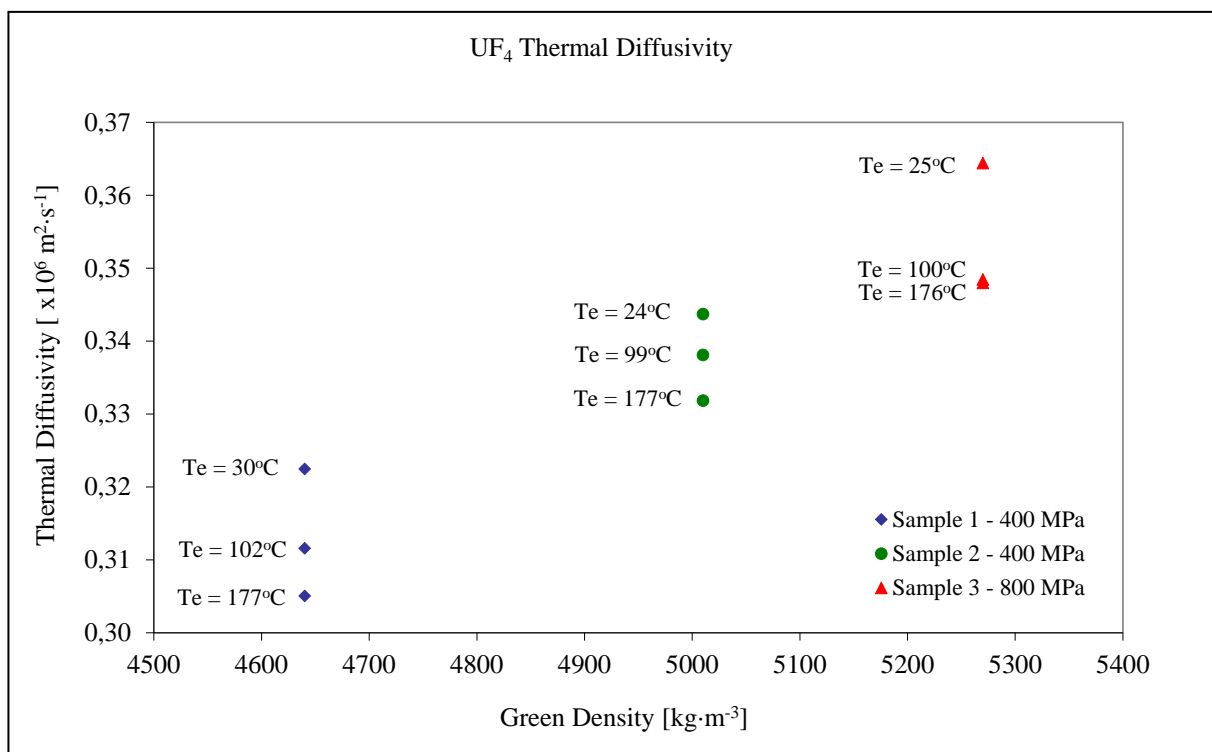


Figure 7. Increase of the UF₄ pellets thermal diffusivity with the increase of the pellet green density.

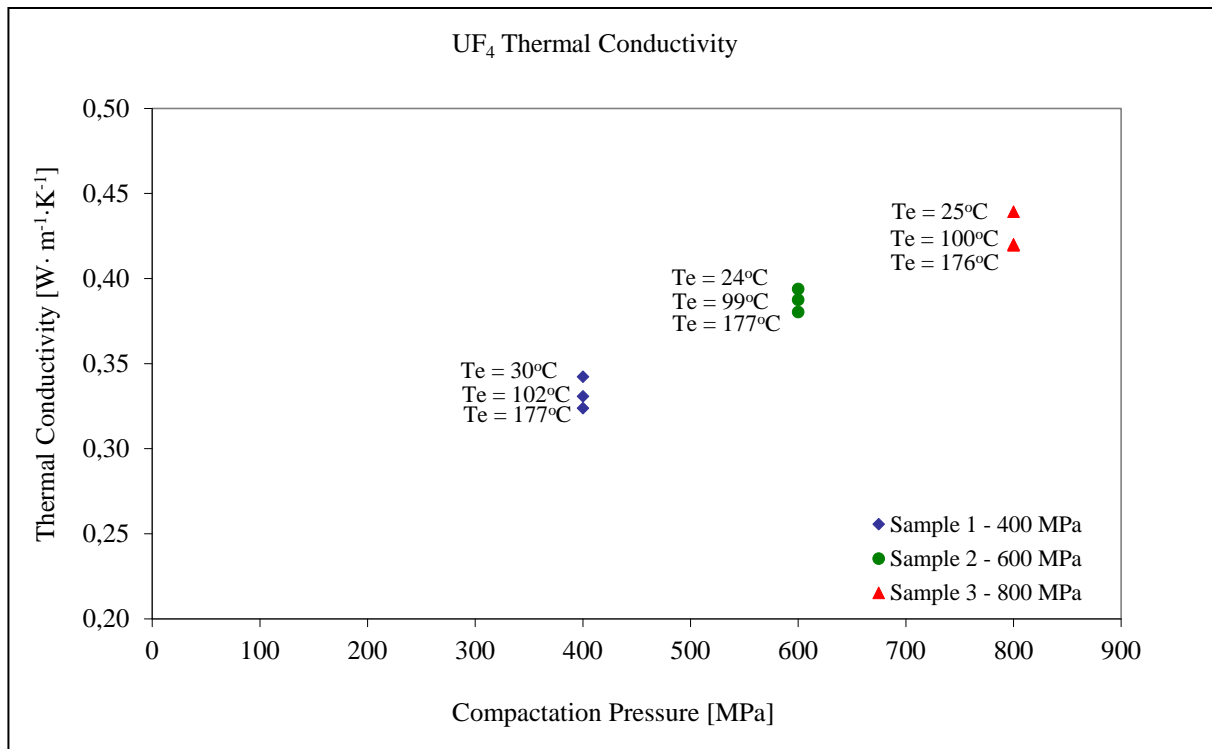


Figure 8. Increase of the UF₄ pellets thermal conductivity with the increase of the compaction pressure.

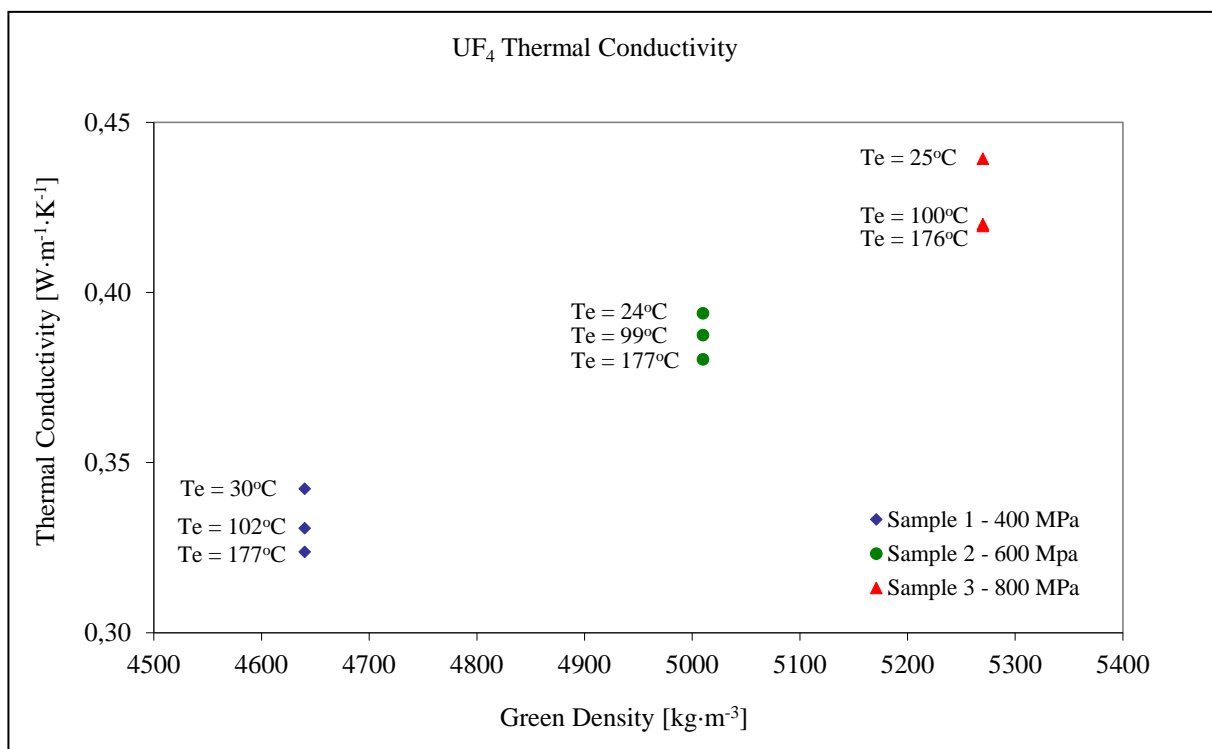


Figure 9. Increase of the UF₄ pellets thermal conductivity with the increase of the pellet green density.

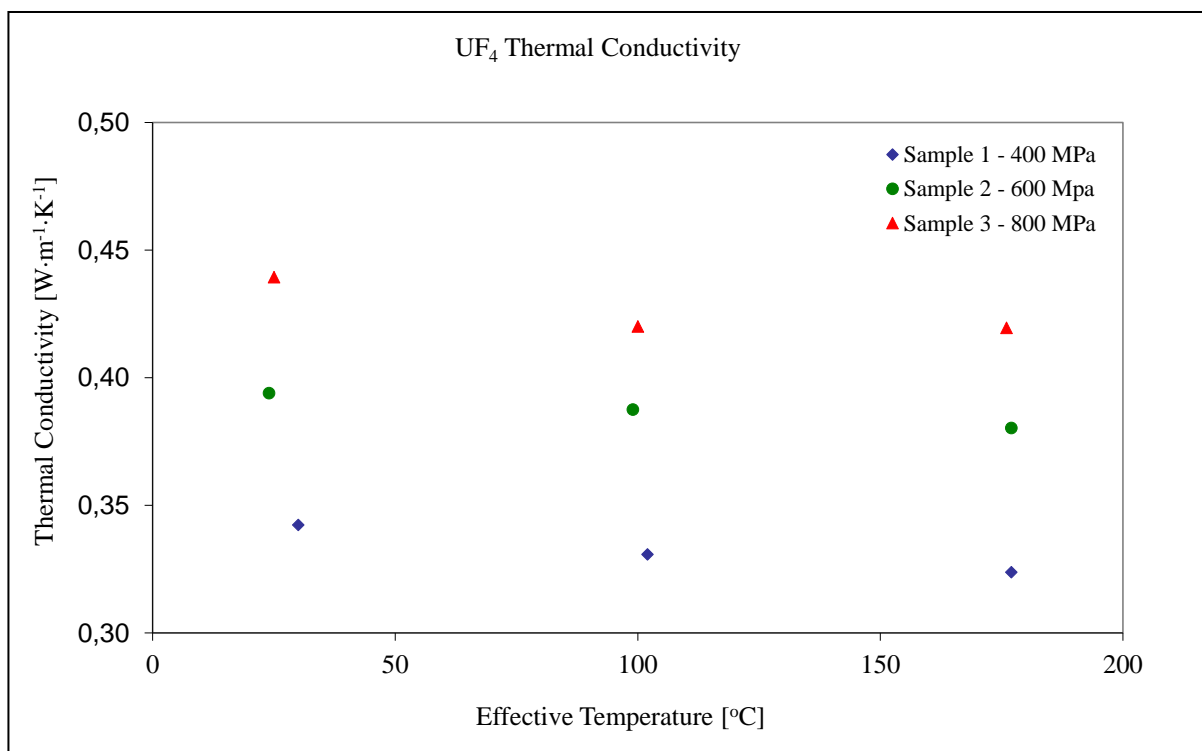


Figure 10. Discrete decrease of the UF₄ thermal conductivity with the increase of the effective temperature.

An extrapolation using the graph of Figure 8 results in an estimative of $0.24 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for the UF₄ powder thermal conductivity at room temperature in the non-pressed state, i.e., corresponding to a compaction pressure equal to zero. This value will be used in the modeling of the UF₄ metalothermic reduction process of IPEN to obtain metallic uranium, whose results will be subject of another publication. For temperature of ca. 177 °C was found by extrapolation, a value of $0.22 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$.

For the powder thermal diffusivity, an extrapolation using the graph of Figure 6 results in a value of $0.28 \times 10^{-6} \text{ m}^2\cdot\text{s}^{-1}$ at room temperature and $0.26 \times 10^{-6} \text{ m}^2\cdot\text{s}^{-1}$ at temperatures of ca. 177 °C, and for the powder bulk density, $3700 \text{ kg}\cdot\text{m}^{-3}$, using the graph of Figure 4.

4. CONCLUSIONS

Pellets of uranium tetrafluoride were produced by uniaxial pressing of the powder produced at IPEN, employing the same technology developed at CDTN to produce nuclear fuel pellets. It was possible to obtain an extrapolated value of ca. $0.24 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for the thermal conductivity of the UF₄ powder sample at room temperature and in the loose state, from the pellets thermophysical properties measurements results obtained applying the laser flash method over these pellets. These value will be employed as input data of a mathematical model that numerically simulates the reduction process of uranium tetrafluoride to metallic uranium, in order to optimize the efficiency of the IPEN's metalothermic process, whose results will be subject of a future publication.

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REFERENCES

- [1] A. M. Saliba-Silva, E. F. Urano de Carvalho, H. G. Riella and M. Durazzo, *Radioisotopes-Applications in Physical Sciences, Chapter 2: Research Reactor Fuel Fabrication to Produce Radioisotopes*, InTech, Rijeka, Croatia, (2011).
- [2] A. M. Saliba-Silva, M. Durazzo, E. F. Urano de Carvalho, H. G. Riella, "Fabrication of U_3Si_2 powder for fuels used in IEA-R1 nuclear research reactor," *Materials Science Forum*, **591-93**, pp.194-199, (2008).
- [3] A. M. Saliba-Silva, I. C. Martins, E. T. Oliveira, J. V. Pereira, M. Durazzo, "Estudos Térmicos e Físicos para Viabilizar a redução Metalotérmica de Urânio Metálico a partir de UF_4 ," *Revista Brasileira de Pesquisa e Desenvolvimento*, **8**, pp.85-90, (2006).
- [4] BELTRAN, A.D., RIVAS DIAZ, M., SANCHEZ, A. F., *Fabricacion de uranio metal. [ed.] Publicacion Bimestral de la Junta de Energia Nuclear*, Energia Nuclear, Madrid, Spain, (1972).
- [5] Ferreira, R. A. N., "Desenvolvimento no CDTN de Processo de Fabricação de Pastilhas Combustíveis," *VII CGEN- Congresso Geral de Energia Nuclear*, Belo Horizonte, MG, Brazil, 31 de Agosto a 3 de Outubro, CD-ROM (1999).
- [6] Parker, W. J., Jenkins, R. J., Butler, C. P., Abbott, G. L., "Flash method of determining thermal diffusivity, heat capacity and thermal conductivity," *J. Appl. Phys.*, **32**, no.9, pp.1679-1684, (1961),
- [7] Ferreira, R. A. N., Miranda, O., Dutra Neto, A.; Grossi, P. A., Martins, G. A., Reis, S. C., Alencar, D. A., Soares Filho, J. G., Lopes C. C., Pinho, M. G., "Implantação no CDTN de Laboratório de Medição de Propriedades Termofísicas de Combustíveis Nucleares e Materiais através do Método Flash Laser," *International Nuclear Atlantic Conference - INAC 2002*, Rio de Janeiro, RJ, Brazil, 11 to 16 August, CD-ROM (2002).
- [8] Grossi, P. A., *Modelagem físico-matemática de sistemas de determinação de propriedades termofísicas com aplicação ao método flash laser*, 107p., Dissertação (Mestrado em Engenharia Mecânica) - Escola de Engenharia, Universidade Federal de Minas Gerais, Belo Horizonte, Minas Gerais, Brazil, (2003).