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# Proposal for Ultrasonic Technique for Evaluation Elastic Constants in UO<sub>2</sub> pellets

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### ABSTRACT

Pellets of uranium dioxide are used as fuel in nuclear power reactors, in which are exposed to high thermal gradients. This high energy will initiate fusion in the central part of the pellet. The expansion of the uranium dioxide pellets, resulting from fission products, can cause fissures or cracks, therefore, the study of their behavior is important. This work aims to develop and propose an ultrasonic technique to evaluate the elastic constants of  $UO_2$  pellets. However, because of the difficulties in handling nuclear material, we proposed an initial study of alumina specimens. Alumina pellets are also ceramic material and their porosity and dimensions are in the similar range of dioxide uranium pellets. They also are used as thermal insulation in the fuel rods, operating under the same conditions. They were fabricated and used in two different sets of 10 alumina pellets with densities of 92% and 96%. The developed ultrasonic technique evaluates the traveling time of ultrasonic waves, longitudinal and transverse, and correlates the observed time and the elastic constants of the materials. Equations relating the speed of the ultrasonic wave to the elastic modulus, shear modulus and Poisson's ratio have led to these elastic constants, with graphics of correlation that showed excellent agreement with the literature available for Alumina. In view of the results and the ease of implementation of this technique, we believe that it may easily be used for dioxide uranium pellets, justifying further studies for that application.

# 1. INTRODUCTION

The reactors PWR, the fuel element is within the zirconium alloy rods containing in isolation fissile material. It consists of the following: fuel tablets, insulator wafer, mounting the wafer Spring column jacket tube and end caps. To maintain the column together pellets within the fuel rod and create voids to accommodate fission gases during irradiation, it is placed an insulating insert  $Al_2O_3$  (alumina) to reduce the heat flow of tablet fuel to the spring region (plenum), and to prevent reactions between the insert and the spring due to the central region of the pads being subjected to high temperatures [1].

The UO<sub>2</sub> fuel is most commonly presented in the form of sintered cylindrical pellets with a density in the range of 92 to 98% of the theoretical density (Manufacturing Process: compacting the UO<sub>2</sub> powder in the form of pellets and subsequent sintering at about 1600  $^{\circ}$  C). [1]. The thermal conductivity of UO2 is a little low and high power generated in the reactor leads to the existence of high thermal gradients on the chip fuel. As a result, high power levels may lead to fusion of the central part of the wafer. However, this is avoided in thermal reactors, since it can lead to performance problems. The major limitations in performance are UO<sub>2</sub> swelling of the tablet caused by fission products (solid and gas) and release of gaseous fission products into the environment contained by the coating, deteriorating the fuel heat transfer to the coolant.

Gaseous fission products generated in pellets as a result of nuclear reactions increase the internal pressure and can cause cracks, breaks and deformation of pellets. Thus, a certain porosity is desired to accommodate these gases, reducing the possibility of damage to the pellets. The effect of porosity in the ceramic material properties has been studied a long time (since 1950), for the main processing route of these materials is the powder technology, which generally results in unintended residual fraction of pores, due limiting the densification process in sintering or the technological optimization of cost / benefit. [1].

In this work we will study the mechanical aspects with the particularity to determine the Poisson's ratio, elastic modulus and shear modulus. The use of several techniques for nuclear fuel characterization, including destructive, further complicates the pellet handling process. Hence, the development of ultrasonic techniques for inspection and characterization of nuclear fuel has been extensively studied because of its efficiency and easy implementation [2],[3] e [4]. Among the mechanical properties, the effect of the porosity has been studied mainly in modulus of elasticity. For the elastic constants of ceramic materials commonly are used destructive techniques, involving several steps and care, and therefore require time and financial resources. In this scenario, ultrasonic techniques are considered interesting alternative for the characterization of nuclear fuel pellets.

As the resource for obtaining these tablets is limited due to a number of safeguards and nuclear safety standards, it was decided to conduct a comparative study on alumina pellets (Al2O3). Thus, the tablet of alumina, which is also a ceramic material which also forms part of the fuel rod assembly as insulation at the ends, was the predominant choice for the analysis of the elastic constants.

As the sound wave propagation depends on the internal structure of the discontinuities in the material, we can associate the speed of wave propagation to their elastic properties [5].

The aim of this work is to develop an ultrasonic technique that enables the determination of elastic constants (elastic modulus, shear modulus and Poisson's ratio) of ceramic materials that can be used in the future for the characterization of  $UO_2$  fuel pellets.

### 2. FUNDAMENTALS

The phase velocity of ultrasonic longitudinal waves and transverse (shear) propagating through a solid isotropic or nearly isotropic polycrystalline material with a random distribution of grain orientation, relate to the Poisson ratio ( $\mu$ ), the Young's modulus (E) and shear modulus (G), by means of relations found in reference [6]. In order to explain the calculation  $\mu$ , E and G, by determining the longitudinal velocity (V<sub>L</sub>) and transverse velocity (V<sub>T</sub>), these relationships can be rewritten as in equation (1) to (3), where  $\rho$  it is the density of the material.

$$\mu = 0.5 \cdot \frac{V_L^2 - 2 \cdot V_T^2}{V_L^2 - V_T^2}$$
(1)

$$E = \rho \cdot \frac{3 \cdot V_T^2 \cdot V_L^2 - 4 \cdot V_T^4}{V_L^2 - V_T^2}$$
(2)

$$G = \rho . V_T^{2} \tag{3}$$

### 3. MATERIALS AND METHODS

### **3.1 Materials**

To be used as body proof were produced two batches of alumina pellets made of uniaxial press simple action:

- 10 pure alumina pellets with densities of around 92%;
- 10 pure alumina pellets with density around 96%.

The average sizes of inserts are approximately:

- 9,0 mm diameter;
- 8,0 mm height.

The experimental apparatus consisted of:

- an ultrasonic pulse transceiver device model "Epoch Plus" brand "Panametrics";
- transducer of longitudinal waves 5MHz brand "Panametrics";
- transducer of transverse waves of 5,0MHz brand "Panametrics";
- couplant of "couplant" brand, applied between the pellets;
- oscilloscope model "DPO 3032" of "Tektronix" brand;

• micrometer millesimal brand name "Tesa" (with an accuracy of 0.001 mm) to measure the diameter and height of the pellets;

• a scale brand "METTLER TOLEDO" model "AX205 DELTA RANGE" capacity 0 to 220g and precision 0,00001g.

### **3.2 Methods**

Initially we tried to use the transducer in pulse-echo mode, however, it was not possible to identify the echoes and accordingly determining the wave time of flight in the analyzed pellets. Thus, obtaining the ultrasonic signal is accomplished by transmission technique, where the ultrasound wave is generated by a transducer used as a transmitter, runs through the material (alumina pellets), and its acquisition is done by another transducer used as a receiver as shown in Figures 1 and 2. In these experiments, the temperature was maintained between 23°C and 25°C.



Figure 1: Scheme longitudinal transducers being used in the transmission mode



Figure 2: Scheme transverse transducers being used in transmission mode

The determination of the ultrasonic wave time of flight followed the following steps:

- **1.** Place the two transducers (transmitter and receiver) in contact (no insert between them) so as to generate a reference signal on time base;
- **2.** positioning of the oscilloscope cursors on the peak of that sign to be used as initial reference (t = 0) for measurements of time;
- **3.** put a pellet between the transducers, generating a new signal, shifted relative to reference previously generated due to the time spent to complete the sample;
- 4. position the other oscilloscope cursor at the peak of this new signal;
- **5.** perform the reading interval, the time base between a slider and the other, obtaining directly on the oscilloscope, the time of flight of the ultrasonic wave to the analyzed pellet (Figure 3).



Figure 3: Getting the signals of ultrasonic waves

Using the methodology described above 10 were measured time of flight of longitudinal and transverse ultrasonic waves in each 20 alumina pellets analyzed: 10 pellets with a density of 92% and 10 pellets with 96% of theoretical density (DT). The time of flight (longitudinal –  $V_L$ , and transverse -  $V_T$ ) for each of the pellets was determined as the average of these measured 10 times in the respective insert. The densities of the pellets were determined by the Archimedes method, in accord with ABNT NBR ISO 5017:2015 [7].

# 4. RESULTS AND DISCUSSIONS

Pellets 1 and 2 provide the necessary data for calculating the longitudinal and transverse velocities in tablets with 92% and 96% relative density.

Table 1. Data of the alumnia penets with 90 % relative density							
Pellets	Density	DT	DT	Height	Longitudinal	Transverse	
	Hydrostatic	$(g/cm^3)$	(%)	(mm)	time	time	
	$(g/cm^3)$				(ns)	(ns)	
1	3,83	3,96	96,90	8,96	877,4	1.505,5	
2	3,82	3,96	96,69	8,91	875,6	1.504,5	
3	3,82	3,96	96,39	8,93	883,6	1.519,1	
4	3,81	3,96	96,67	8,99	888,1	1.523,5	
5	3,81	3,96	96,67	8,99	888,2	1.555,3	
6	3,84	3,96	97,10	8,96	879,7	1.511,4	
7	3,82	3,96	96,35	8,99	887,2	1.570,9	
8	3,86	3,96	97,64	8,96	876,4	1.570	
9	3,81	3,96	96,48	8,95	892,4	1.583,5	
10	3,82	3,96	96,25	8,95	879,8	1.569	
Average height pellet			$8,97 \pm 0,06$				
Average longitudinal time			882,8 ± 2,49				
Average transverse time			$1.541,3 \pm 2,51$				
Average Hydrostatic Density			$3,83 \pm 0,02$				

 Table 1: Data of the alumina pellets with 96% relative density

# Transverse velocity ( $V_T$ ) = 5822 ± 136, 7 m/s Longitudinal velocity ( $V_L$ ) = 10160, 9 ± 67, 45 m/s

Pellets	Density	DT	DT	Height	Longitudinal	Transverse
	Hydrostatic	$(g/cm^3)$	(%)	(mm)	time	time
	$(g/cm^3)$				(ns)	(ns)
11	3,73	3,96	93,20%	9,11	938,2	1.704,2
12	3,71	3,96	92,71%	9,11	945,0	1.708,6
13	3,72	3,96	92,92%	9,15	948,5	1.7089
14	3,73	3,96	92,96%	9,07	939,2	1.811,4
15	3,70	3,96	92,89%	9,11	944,0	1.702,5
16	3,69	3,96	92,14%	9,09	944,0	1.717,1
17	3,69	3,96	92,45%	9,08	942,8	1.697,4
18	3,71	3,96	92,98%	9,04	932,6	1.665,9
19	3,70	3,96	92,80%	9,13	933,6	1.676,4
20	3,72	3,96	92,80%	9,13	941,6	1.725,1
Average height pellet				$9,1 \pm 0,06$		
Average longitudinal time				941,06 ± 2,23		
Average transverse time				$1.719,8 \pm 2,65$		
Average Hydrostatic Density				$3,71 \pm 0,02$		

 Table 2: Details of the alumina pellets with 92% relative density

# Transverse velocity ( $V_L$ ) = 9532 ± 52,2m/s Longitudinal velocity ( $V_T$ ) = 5219,2 ± 117,93 m/s

With the values presented is possible, using the equations (1), (2) and (3) calculating the elastic constants of the used pellets, shown in Table 3.

Pellet (%)	υ	E (GPa)	G(GPa)
96	0,2556	$326 \pm 21$	130
92	0,2859	$259 \pm 17$	101

## Table 3: values found for the elastic constants determined experimentally

Due to difficulty in performing mechanical testing of ceramic specimens, the determination of reference values for the modulus of elasticity of the specimens used in this study were obtained from the interpolation of the results found in reference [8] to alumina 100 % of theoretical density, and in reference [9] to 85% alumina and 94% of the theoretical density, all obtained by the conventional process from mechanical tests (Table 4).

Relative density (%)	E (GPa)	G (GPa)	μ
85	220	90	0,22
94	277	117	0,21
100	421	171	0,24

Table 4: elastic constants of the alumina values in the literature for tablets with 85%,94% and 100% of theoretical density

The Figure 4 graph shows a polynomial fit second order for the values of E shown in Table 4. In all tested settings, this is what provided the best correlation coefficient, obtaining an R = 1 value, which shows the polynomial fit, fits very well to this data.



Figure 4: interpolation to the values E shown in Table 4

The calculation of the modulus of elasticity (E) through the equation provided by the graphical adjustment shown in Figure 4, to insert porosity 96% theory, provides a value for E = 308 GPa approximately, whereas the value determined by the ultrasonic technique was  $326 \pm 21$  GPa (5.8% offset in relation to the interpolated value).

The calculation of the modulus of elasticity (E) through the equation provided by the graphical adjustment shown in Figure 4, to insert porosity 92% theory, provides a value for E = 248 GPa approximately, whereas the value determined by the ultrasonic technique was  $259 \pm 17$  GPa (4.4% offset in relation to the interpolated value).

The Figure 4 graph shows a second-order polynomial fit to the E values shown in Table 4. In all the tested settings, this is what gave the best correlation coefficient, obtaining a value R = 1, which shows the polynomial fit fits very well to this data.



Figure 5: Interpolation for G values shown in Table 4

The calculation of the shear modulus (G) by the equation provided by the graph shown in Figure 5 setting for the wafer 96% of theoretical density, provides a value for G = about 132 GPa, while the value determined by the ultrasonic technique was  $130 \pm 7$  GPa (1.5% deviation from the interpolated value).

The calculation of the shear modulus (G) by the equation provided by the graph shown in Figure 5 setting for the wafer 92% of theoretical density, provides a value for G = about 105 GPa, while the value determined by the ultrasonic technique, was  $101 \pm 5$  GPa (deviation of 3.8% compared to the interpolated value).

### **5. CONCLUSIONS**

The results of this study showed that the use of ultrasonic technique for the determination of elastic constants Alumina ceramic pellets gave results compatible with those found in the literature for the porosity range evaluated. The UO<sub>2</sub> pellets, used as nuclear fuel, in addition to presenting the same nature (ceramic), has very similar dimensions and porosities of alumina pellets used in this work. Thus, it is expected that in future work, it is possible to verify that this technique can also be used in the determination of elastic constants  $UO_2$  pellets, offering advantages to be nondestructive, offering greater security since would not generate waste, which must be monitored.

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### REFERENCES

- 1. Perrota, J. A; Curso de Difusão Nuclear; USP; Julho 2012.
- 2. J. P. Panakkal, J. K. Ghos, P. R. Roy, "Use of ultrasonic velocity for measurement of density of sintered fuel pellets". J. Phys. D: Applied Phys. 17, p. 1791-1795, 1984.
- 3. D. Laux, et al., "Ultrasonic study of UO2: effects of porosity and grain size on ultrasonic attenuation and velocities". J. Nucl. Mat. 300, p.192-197, 2002.
- 4. M. C. Young, et al., "Non destructive Evaluation of Nuclear Fuel Rods Using Ultrasonic Resonance Phenomena". 15th International Conference on Structural Mechanics in Reactor Technology, Seoul Korea, 1999.
- 5. LAMARSH, J.R. Introduction to Nuclear Engineer, 3° Edição, Addison-Wesley Publishing Co.1982, New York, USA
- 6. ANSI/ASTM. ASTM E 494. Standard Recommended for Measuring Ultrassonic Velocity in Pratice Materials, Philadelphia.1995: 11p.
- 7. Produtos refratários conformados densos Determinação da densidade de massa, porosidade aparente e porosidade real. ABNT NBR ISO 5017:2015
- 8. Yoshimura, H.N.; Molinasi, A.L; Siqueira, G.R; Camargo, A.C; Narita, N.E.; Cesar, P.F.; Goldenstein, H., Efeito da porosidade nas propriedades mecânicas de uma alumina de elevada pureza. Associação Brasileira de Cerâmica. 2005. Vol. 51: p 239-
- 9. Alumina as a Ceramic Material. The American Ceramic Society-Special Publication no 4; Mechanical Proprieties of Alumina. 1964; p.46.