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Proposal for ultrasonic technique for evaluation elastic constants in UO_2 pellets

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ABSTRACT

Uranium dioxide pellets (UO_2) used as fuel in nuclear power reactors are subjected to high thermal gradients and may undergo swelling during reactor operation. The swelling of UO_2 pellets, resulting from the fission products, causes cracking of the material and it is thus critical to study their behaviour. This study evaluated the application of the ultrasonic method in obtaining the elastic constants of UO_2 pellets. Due to the difficulties in handling nuclear material, we used tablets of alumina (Al_2O_3) for a comparative study. For this, two sets of 10 Al_2O_3 pellets with densities of 92% and 96% were fabricated. Ultrasonic transmission technique was used for obtaining measurements of the travel time of longitudinal and transverse ultrasonic waves. Equations relating material density and velocity of the ultrasonic wave to the elasticity, shear and Poisson's modules allowed the determination of these elastic constants, whose values showed excellent agreement with those available in the literature for Al_2O_3.

Keywords: Ultrasound; Ultrasonic Wave; Elastic Constants; Alumina.

1. INTRODUCTION

In pressurized water reactors (PWR) the fuel element is stacked inside sealed zirconium alloy rods. The rods are made of the following items: fuel pellets, insulator wafer, insert column fixing spring, coating tube and end caps. To maintain the fuel pellets column formed inside the fuel rod and to create voids to accommodate fission gases during irradiation, an insulating pad Al_2O_3 (alumina) is placed inside the rod to reduce the heat flow from the fuel pellet to the spring region (plenum), thus preventing reactions between them, due to the central region of the fuel pads being subjected to high temperatures, (PERROTA [1]).

The UO_2 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_2 powder in the form of pellets and subsequent sintering at about 1600 °C) (PERROTA [1]). The thermal conductivity of UO_2 is a little low and the high power generated in the reactor leads to the existence of high thermal gradients on the fuel pellet. As a result, high power levels may lead to melting of the central part of the pellets. However, this is avoided in thermal reactors since it can lead to performance problems. The major limitations in performance are UO_2 swelling of the tablet caused by fission products (solid and gas) and the 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 the fuel 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 desirable to accommodate these gases, reducing the possibility of damage to the pellets. The effect of porosity in the ceramic material properties has long been studied (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 (PERROTA [1]).

In this work we will study the mechanical aspects relevant for the project of the UO_2 fuel element, determine focussing on the determination of 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

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studied due to its efficiency and easy implementation (PANAKKAL *et al*. [2]; LAUX *et al*. [3]; YOUNG [4]; SMITH *et al*. [5]; ALARCÓN *et al*. [6]; SASMITA *et al*. [7]). Among the mechanical properties, the effect of the porosity has been studied mainly in relation to the modulus of elasticity. For the elastic constants of ceramic materials, destructive techniques, involving several steps and care, are commonly used, what require time and financial resources. In this scenario, ultrasonic techniques are considered an interesting alternative for the characterization of nuclear fuel pellets.

As the resource for obtaining these tablets is limited due to a few safeguards and nuclear safety standards, it was decided to conduct a comparative study on alumina pellets (Al_2O_3). Thus, the tablet of alumina, which is also a ceramic material that 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 (LAMARSH [8]).

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.

The phase velocity of ultrasonic longitudinal and transverse (shear) waves propagating through a solid isotropic or nearly isotropic polycrystalline material with a random distribution of grain orientation, relate to the Poisson ratio (μ), the Young's modulus (E) and shear modulus (G), by means of relations found in reference ANSI/ASTM [9]. The elastic constants μ, E and G, are thus determined as indicated in equations (1) to (3), where V_L is the longitudinal velocity V_T, the transverse velocity and ρ it is the density of the material.

$$\mu = 0,5\frac{V_L^2 - 2V_T^2}{V_L^2 - V_T^2} \tag{1}$$

$$E = \rho\frac{3V_T^2V_L^2 - 4V_T^4}{V_L^2 - V_T^2} \tag{2}$$

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

2. MATERIALS AND METHODS

2.1. 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 the pellets 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 5 MHz brand "Panametrics";
- transducer of transverse waves of 5,0 MHz brand "Panametrics";
- couplant of "couplant" brand, applied between the pellets;
- oscilloscope model "DPO 3032" of "Tektronix" brand;
- micrometre 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 220 g and precision 0,00001 g.

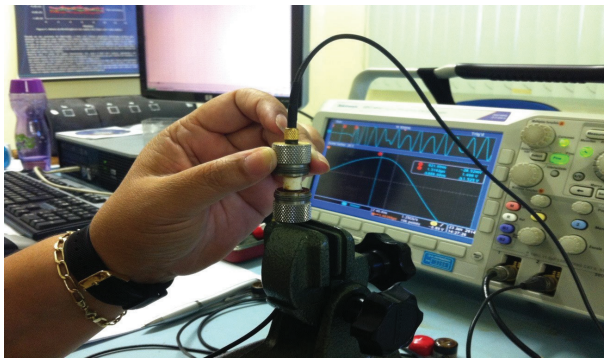

Figure 1: Scheme longitudinal transducers being used in the transmission mode.

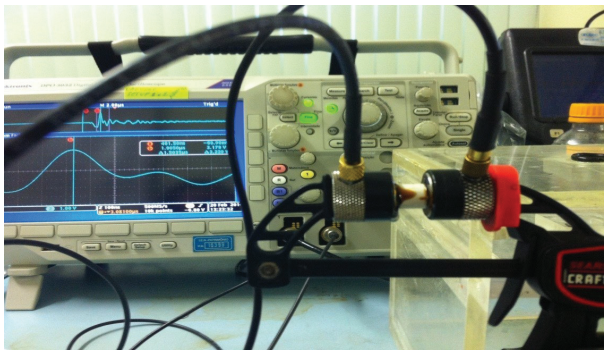

Figure 2: Scheme transverse transducers being used in transmission mode.

2.2. 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 analysed 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 between 23°C and 25°C.

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 analysed pellet.

Using the methodology described above 10 were measured time of flight of longitudinal and transverse ultrasonic waves in each 20 alumina pellets analysed: 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 [10].

3. 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, as showed in Tables 1 and 2.

With the values presented is possible to calculate the elastic constants of the used pellets, shown in Table 3.

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

PELLETS	DENSITY HYDROSTATIC (g/cm³)	DT (g/cm³)	DT (%)	HEIGHT (mm)	LONGITUDINAL TIME (ns)	TRANSVERSE TIME (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 ± 0,06		
Average longitudinal time				882,8 ± 2,49		
Average transverse time				1.541,3 ± 2,51		
Average hydrostatic density				3,83 ± 0,02		

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

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

PELLETS	DENSITY HYDROSTATIC (g/cm³)	DT (g/cm³)	DT (%)	HEIGHT (mm)	LONGITUDINAL TIME (ns)	TRANSVERSE TIME (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	1.708,9
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 ± 0,06		
Average longitudinal time				941,06 ± 2,23		
Average transverse time				1.719,8 ± 2,65		
Average hydrostatic density				3,71 ± 0,02		

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

Table 3: Values found for the elastic constants determined experimentally.

PELLET (%)	υ	E (GPa)	G (GPa)
96	0,2556	326 ± 21	130
92	0,2859	259 ± 17	101

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 by YOSHIMURA, *et al.*[11], to alumina 100% of theorical density, obtained by the conventional process from mechanical tests (Table 4).

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

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

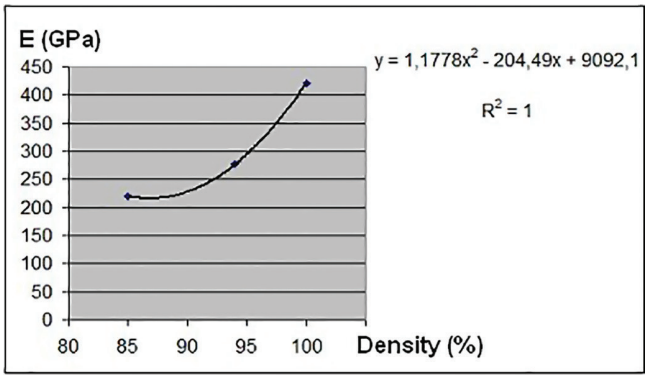

Figure 3: Interpolation to the values E shown in Table 4.

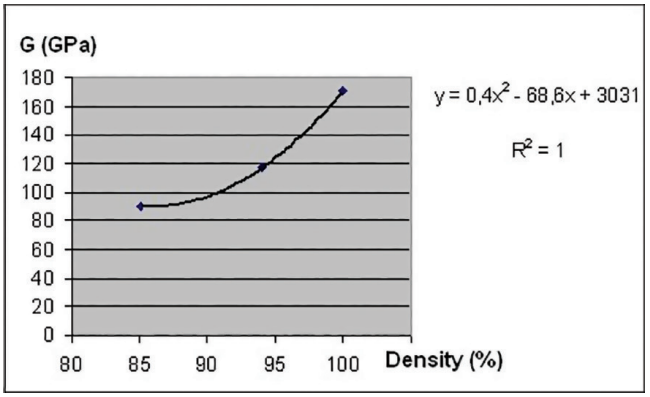

Figure 4: Interpolation for G values shown in Table 4.

The Figure 3 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.

The calculation of the modulus of elasticity (E) through the equation provided by the graphical adjustment shown in Figure 3, to insert porosity 96% theoretical density, provides a value for E = 308 GPa approximately, whereas the value determined by the ultrasonic technique was 326 ± 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 3, to insert porosity 92% theoretical density, provides a value for E = 248 GPa approximately, whereas the value determined by the ultrasonic technique was 259 ± 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 that the polynomial fit fits very well to this data.

The calculation of the shear modulus (G) by the equation provided by the graph shown in Figure 4 setting for the pellets 96% of the theoretical density, provides a value for G of about 132 GPa, while the value determined by the ultrasonic technique was 130 ± 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 4 setting for the pellets 92% of the theoretical density, provides a value for G of about 105 GPa, while the value determined by the ultrasonic technique, was 101 ± 5 GPa (deviation of 3.8% compared to the interpolated value).

4. CONCLUSIONS

The results of this study showed that the use of ultrasonic technique for the determination of elastic constants of alumina ceramic pellets gave results compatible with those found in the literature for the porosity range evaluated. The UO_2 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 the elastic constants of UO_2 pellets, offering advantages to be a nondestructive, offering greater security since it would not generate waste, which must be monitored. Further technical details and discussions about this work can be found in reference (LOPES, [12]).

5. ACKNOWLEDGMENTS

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