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ULTRASONIC SPECTRAL ANALYSIS FOR NUCLEAR FUEL CHARACTERIZATION

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ABSTRACT

Ceramic materials have been widely used for various purposes in many different industries due to certain characteristics, such as high melting point and high resistance to corrosion. Concerning the areas of applications, automobile, aeronautics, naval and even nuclear, the characteristics of these materials should be strictly controlled. In the nuclear area, ceramics are of great importance once they are the nuclear fuel pellets and must have, among other features, a well controlled porosity due to mechanical strength and thermal conductivity required by the application. Generally, the techniques used to characterize nuclear fuel are destructive and require costly equipment and facilities. This paper aims to present a nondestructive technique for ceramic characterization using ultrasound. This technique differs from other ultrasonic techniques because it uses ultrasonic pulse in frequency domain instead of time domain, associating the characteristics of the analyzed material with its frequency spectrum. In the present work, 40 Alumina (Al_2O_3) ceramic pellets with porosities ranging from 5% to 37%, in absolute terms measured by Archimedes technique, were tested. It can be observed that the frequency spectrum of each pellet varies according to its respective porosity and microstructure, allowing a fast and non-destructive association of the same characteristics with the same spectra pellets.

1. INTRODUCTION

Uranium dioxide (UO_2) ceramic pellets are widely used as nuclear fuel in PWR (Pressurized Water Reactor) reactors, as is the case of Angra 1 and 2. When UO_2 fuel pellets are thermally analyzed, one can notice that a number of factors influence their thermal conductivity, for instance, temperature, porosity, oxygen / uranium atomic ratio, plutonium dioxide (PuO_2) content, pellet cracking degree and burning rate (burn up).

On the whole, the thermal conductivity of solids decreases as the porosity in its structure increases. A low porosity, therefore, is aimed to maximize the conductivity, enhancing heat flow in the pellets.

However, the gaseous fission products accumulated in the pellets from nuclear fissions result in internal pressures, which can cause cracks and deformations in the pellets. Thus, a certain degree of porosity is desired to accommodate fission gases, limiting a possible deformation of the fuel by swelling or cracking. Porosity can be considered one of the most important factors as it is not only related to structural integrity of the fuel, but also directly linked to thermal conductivity.

Porosity degree, therefore, must be carefully controlled for optimal operation and efficiency of the reactors, especially in fast reactors (breeders). In these reactors, the high specific power increases the production rate of fission gas per unit volume.

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 [1],[2],[3]. These techniques use ultrasonic pulse analyses in time domain, associating the variation of pulse speed with the characteristics of the analyzed material. Then, specimen thickness (e) and time interval (Δt) between the emission and reception of ultrasonic pulse must be measured. Based on these data, speed must be calculated through equation 1 [4].

$$V = \frac{2e}{\Delta t} \qquad \text{Eq. 1}$$

In this equation, time is conventionally determined by the interval between two consecutive echoes measured by an oscilloscope, as shown in Figure 1.

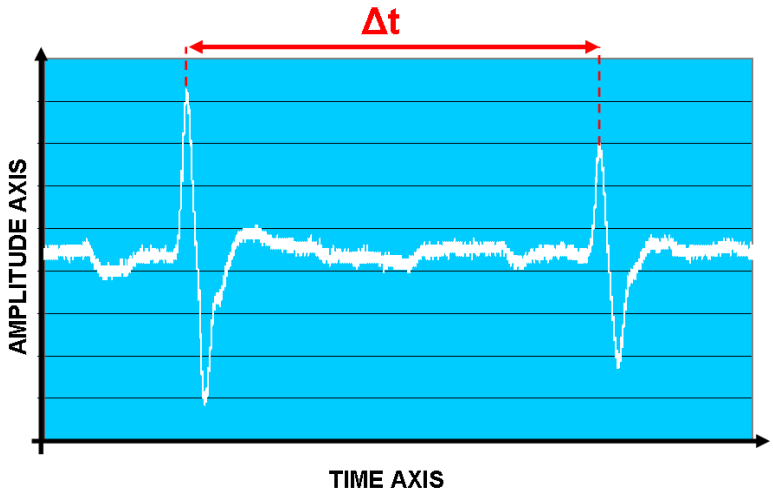


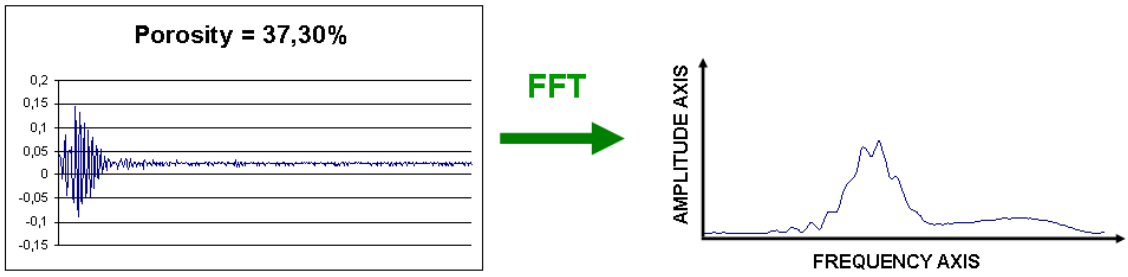
Figure 1 – Time interval between two consecutive echoes from the analyzing ultrasonic pulses in time domain.

Based on a set of several samples with different porosities a correlation between propagation speed and material porosity can be derived. This correlation allows evaluations of porosity by means of simple measurements of the ultrasonic pulse speed in other materials.

Although simple and efficient this technique is limited by high porosity, as it demonstrates the experimental results in the Figure 2.a. In these high porosity materials, ultrasonic pulse-echo detection is unfeasible due to the strong attenuation by the voids (pores). Another drawback of this technique lies in the relation between the ultrasonic wave speed and the specimen thickness, which promotes the echo overlapping [5] depicted in the Figure 3-a

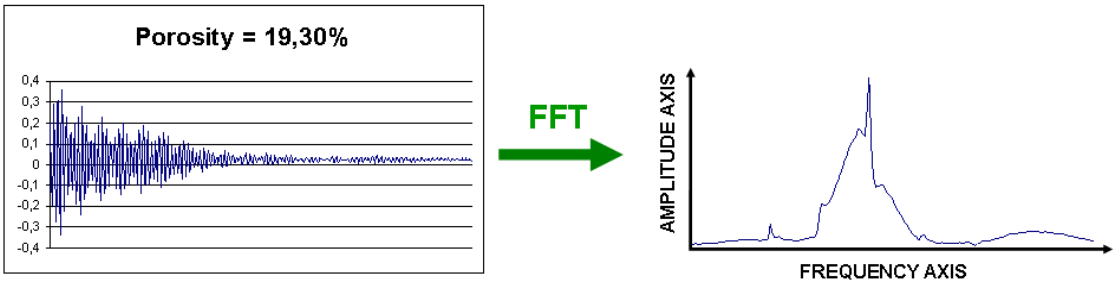
When conventional assessment can not be performed, researches have shown the advantages of using frequency domain in relation to time domain [6], [7], [8]. In these cases, FFT (Fast Fourier Transform) provides a map of the frequencies that make up this signal, enabling a spectral analysis of the ultrasonic pulse.

Regarding frequency domain, it is possible to observe how the frequency spectrum of the ultrasonic pulse interacts with the internal structure of the analyzed material, altering its initial shape according to these interactions. After passing through the material, the signal is deformed in a unique way depending on the characteristics of the sample under analysis (Figure 2-b and Figure 3-b)



a) **b)**

Figure 2 - (a) ultrasonic pulse in time domain strongly attenuated due to the high porosity of the sample and in (b) frequency spectrum of this pulse.



a) **b)**

Figure 3 - (a) Return echoes from overlapped ultrasonic pulse due to the high propagation speed and small sample thickness (4mm); (b) frequency spectrum of the pulse.

The analysis of frequency domain provides information that can be used to characterize the analyzed material from different aspects (porosity, pore size distribution etc). Unlike time domain analysis, it functions as its "fingerprint ultrasound".

The present work, thus, shows the influence of porosity and microstructure of ceramic samples in the frequency spectrum of an ultrasonic pulse.

2. MATERIALS AND METHODS

This section will list all materials utilized and methods adopted.

2.1. Sample description

The aim of this paper is to present an ultrasonic technique that allows the characterization of UO_2 pellets; however, due to the special care required when handling these pellets, 40 Alumina (Al_2O_3) ceramic pellets were used to simulate the nuclear fuel produced in uniaxial press using 25x25mm pellets, generating specimens at about 5mm thick (Figure 4).

Figure 4 – Alumina Pellets

Two compacting pressures, 50 and 100Kgf/cm2 were applied. Samples were sintered in resistance furnace at temperatures of 1400, 1480, 1540 and 1580 ° C for 1 hour in order to obtain pellets with porosities that vary according to differences in compaction pressure and sintering temperature. Table 1 shows the classifications used for manufactured pellets, the manufacturing conditions of each pellet and the porosity values obtained by the Archimedes method.

Table 1 - Values of porosity of each pellet produced according to pressure and sintering temperature.

Pellet	Pres.(kgf/cm^2)	Time.(ºC)	Porosity (%)
AA1	50	1150	40,60
AA2	50	1150	39,40
AA3	50	1150	39,00
AA4	50	1150	39,40
AA5	50	1150	39,10
AB1	50	1400	30,20
AB2	50	1400	28,50
AB3	50	1400	29,70
AB4	50	1400	30,50
AB5	50	1400	29,00
AC1	50	1480	21,40
AC2	50	1480	21,20
AC3	50	1480	21,20
AC4	50	1480	20,60
AC5	50	1480	20,60
AD1	50	1540	12,30
AD2	50	1540	11,90
AD3	50	1540	11,70
AD4	50	1540	11,10
AD5	50	1540	11,10
AE1	50	1580	6,90
AE2	50	1580	6,54
AE3	50	1580	6,50
AE4	50	1580	5,93
AE5	50	1580	5,67

Pellet	Pres.(kgf/cm^2)	Time.(ºC)	Porosity (%)
BA1	100	1150	37,30
BA2	100	1150	37,20
BA3	100	1150	37,20
BA4	100	1150	37,10
BA5	100	1150	36,80
BB1	100	1400	29,00
BB2	100	1400	28,70
BB3	100	1400	28,70
BB4	100	1400	28,70
BB5	100	1400	28,70
BC1	100	1480	19,90
BC2	100	1480	19,70
BC3	100	1480	19,60
BC4	100	1480	19,40
BC5	100	1480	19,30
BD1	100	1540	10,90
BD2	100	1540	10,80
BD3	100	1540	10,60
BD4	100	1540	10,60
BD5	100	1540	10,50
BE1	100	1580	5,81
BE2	100	1580	5,54
BE3	100	1580	5,43
BE4	100	1580	5,20
BE5	100	1580	5,09

It is possible to observe that the pellets produced under the same conditions presented variations of porosity - probably resulting from small oscillations in pressing and sintering conditions.

2.2. Experimental procedure

The process of ultrasonic inspection of the pellets started after the open porosity of the sintered ceramics was determined by ASTM 15.02(8) (Archimedes method).

The experimental setup (Figure 5) used a longitudinal wave transducer with 5 MHz of nominal frequency, applied in pulse-echo mode. Explorer II Matec Instruments was used for generation and signal acquisition. This equipment also performs the conversion from pulse time domain to frequency domain by FFT. Afterwards, a computer connected to Explorer II analyzes and stores the spectra.

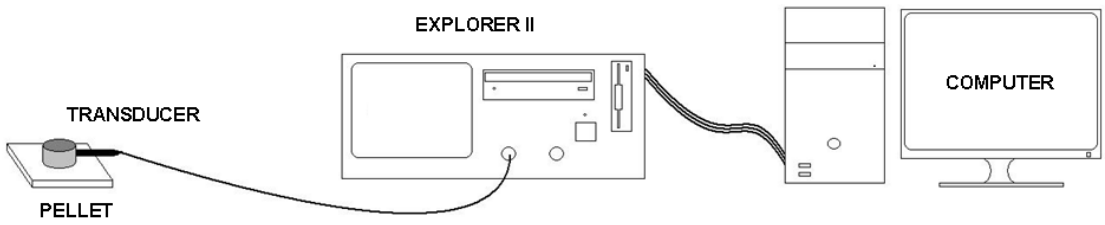


Figure 5 – Experimental set up

In pulse-echo mode, the transducer emits an ultrasonic pulse that passes along sample thickness being reflected by the opposite surface and returning to the transducer. Based on the FFT, it is possible to observe a map of the frequencies that make up this signal.

When observed in the frequency domain, after passing along crossing the entire sample, the ultrasonic pulse shows changes in its shape when compared to the initial spectrum. These changes are the result of the interaction of ultrasonic pulse with the structure of the analyzed material. Then, the frequency spectrum has its form changed due to the interactions with the structure of the material (Figure 6), serving as its identifier. This enables pellet characterization through the changes it promotes in the frequency spectrum of the ultrasonic pulse.

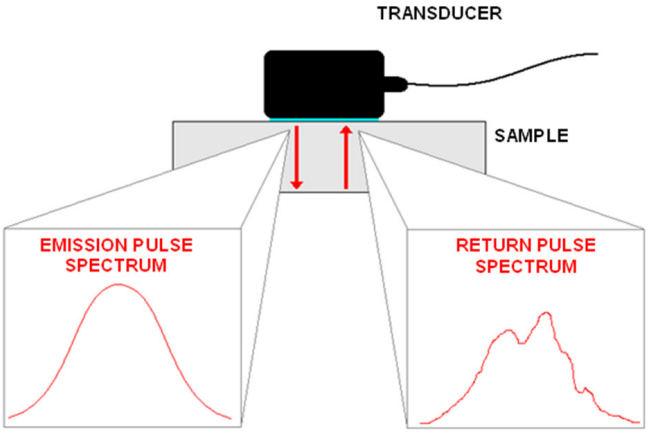


Figure 6 – Operation scheme of the transducer in the pulse-echo mode, showing the change in the frequency spectrum shape according to interactions with the sample

The frequency spectrum of the return ultrasonic pulse was then obtained from all the 40 pellets.

3. RESULTS AND DISCUSSIONS

It is important to mention that pellets produced under the same compaction pressure, sintering temperature and same porosity range generated similar spectra, as shown in Figure 7.

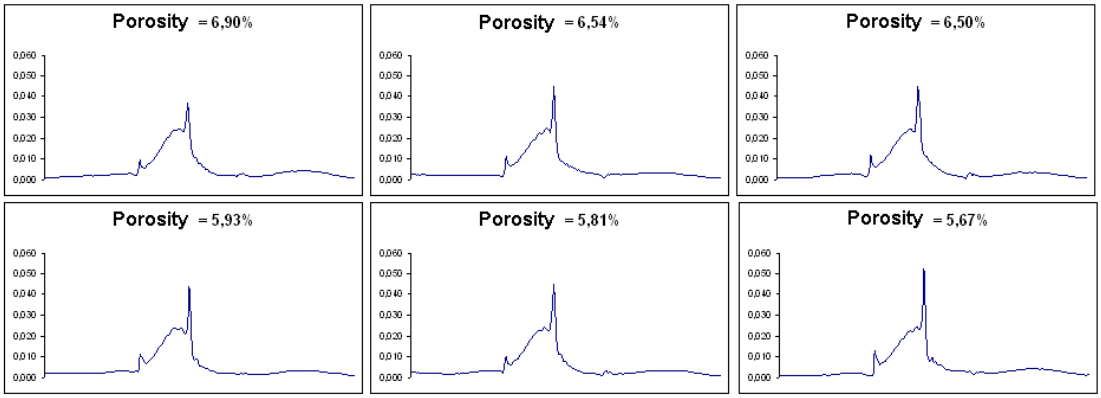


Figure 7 – Pellet spectra with the same porosity range

Similarly, pellets with different porosities showed distinct frequency spectra as shown in Figure 8.

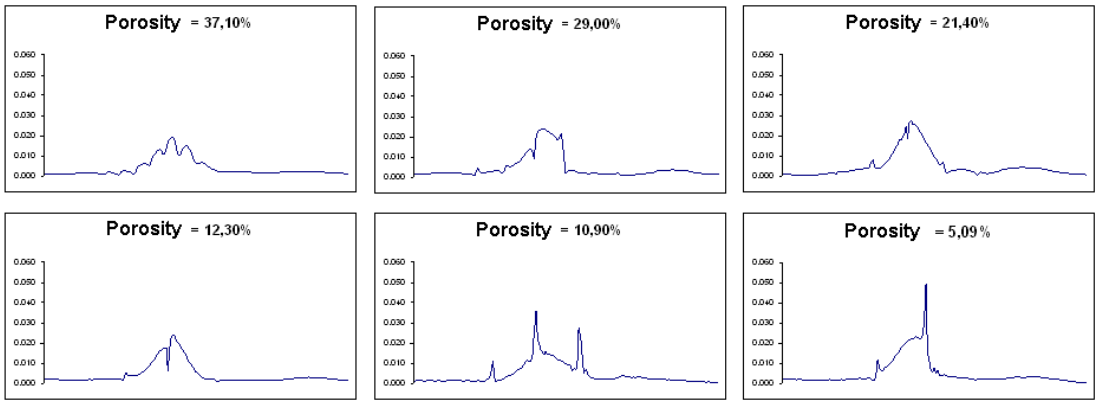


Figure 8 – Different porosity pellet spectra.

Despite having the same porosity, AC2 and AC3 pellets manufactured under the same conditions of temperature and pressure showed different spectra. This represents different microstructural properties according to ultrasonic technique.

The samples, therefore, were analyzed through scanning electron microscopy aiming at a more detailed characterization as well as porosity measurements of the pellets.

Although pellets AC2 and AC3 present the same porosity, the scanning electron microscopy revealed that these pellets, measured by Archimedes technique, have completely different microstructures (Figure 9). These differences could not be detected by the Archimedes technique once this method is not sensitive to microstructural variations. However, the ultrasonic technique enabled the identification of microstructural variations.

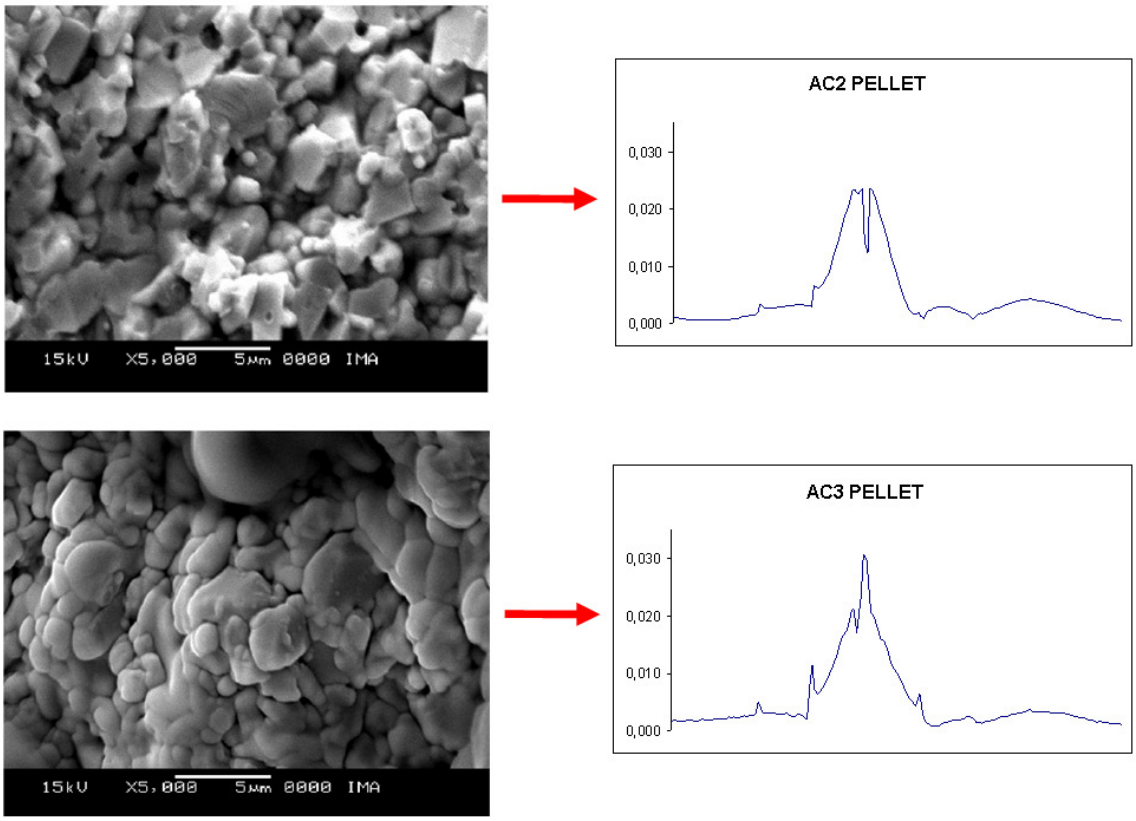


Figure 9 - AC2 and AC3 pellets, both with 21.2% porosity, showed different spectra, which is justified by their different microstructures.

Regarding these samples, it was observed that AC2 pellets were sintered. However, AC3 pellets presented an intermediate degree of sintering, as shown in Figure 9. These microstructural differences may be associated with oscillations in the manufacturing conditions of the pellets.

Therefore, other pellets were analyzed by SEM. The results showed that all the pellets that presented frequency spectrum with the same shape had similar microstructure, as seen in Figure 10.

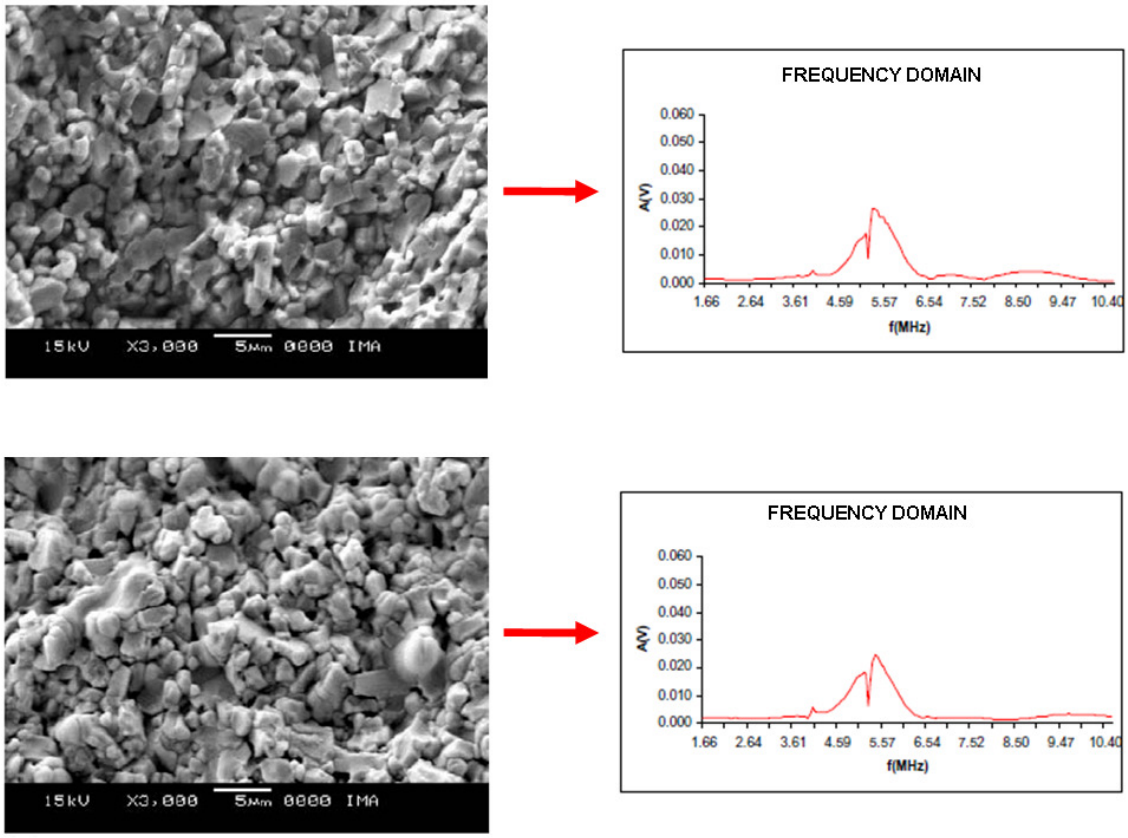


Figure 10 – Interaction of ultrasonic pulse with similar microstructure provides similar frequency spectra.

Each sample served as a filter, reducing the frequency of the greatest interactions that occurred due to microstructural features such as porosity, pore size, grain size, presence of other phases, among others. Then, each pellet can be characterized and identified by the changes it causes to the initial shape of the ultrasonic spectrum.

4. CONLUSIONS

Based on the analysis of the frequency spectrum of the pellets together with their respective microstructures observed by SEM, it was possible to notice that similar structures generate similar spectra. This type of analysis enabled the identification of porosity rate a group of 40 Alumina ceramic pellets.

The ultrasonic pulse was sensitive to changes in the microstructure of the pellets, allowing the identification of microstructural differences even in pellets with the same porosity value.

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REFERENCES

1. 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.
2. 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.
3. 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.
4. J. G. Santos, E. E. M. Oliveira, M.S.Q. Bittencourt, C. A. Lamy "*Estudo Preliminar de Caracterização de Alumina Através de Ondas Ultra-sônicas"*. 42° Congresso de Cerâmica, Poços de Caldas, MG, **v.2**, pp 792-795. 1998.
5. A. L. Santos, "*Estudo de uma Nova Técnica de Medida de Tempo de Percurso da Onda Ultrassonica Usando o Espectro de Freqüência"*. Dissertação de Mestrado. Instituto de Engenharia Nuclear, Rio de Janeiro, 2010.
6. D. B. Baroni, "*Desenvolvimento de técnica ultrassônica para medida de porosidade em pastilhas de UO2"*. Dissertação de Mestrado. Instituto de Engenharia Nuclear, Rio de Janeiro, 2008.
7. A. M. L. M. Costa, "*Estudo da Influência das Micro-estruturas e da Porosidade das Pastilhas de Alumina no Pulso Ultra-sônico no Domínio da Frequência"*. Dissertação de Mestrado. Instituto de Engenharia Nuclear, Rio de Janeiro, 2009.
8. C. S., Lucas "*Medida de Tamanho de Poro em Membrana Polimérica de Microfiltração Utilizando Técnica Ultrassônica e Redes Neurais Artificiais"*. Dissertação de Mestrado. Instituto de Engenharia Nuclear, Rio de Janeiro, 2009.