Ultrasonic critical angle reflectometry applied to porous nuclear fuel mechanical characterization

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In the present study, an ultrasonic 5 MHz reflectometer has been built in order to assess the elastic moduli of uranium dioxide in a non destructive way. Ultrasonic reflection coefficient was measured for different samples of UO_2 with various volume fractions of porosity. All of these samples have already been studied with acoustic microscopy and echography a few years ago. We show that the longitudinal, shear and Rayleigh waves velocities can be measured very accurately with this reflectometry method leading to a rapid and accurate assessment of elastic constants. Indeed, the new values of velocities obtained, match very favourably with the previous results. The elastic moduli obtained are in good agreement with the literature data. The relations between V_L, V_S and V_R and the porosity (p) on the one hand, and E and G with p on the other hand were presented and discussed. In particular, a link between these relations and the pores morphology were pointed out.

1 Introduction

In order to simulate the in pile behaviour of nuclear fuel, the knowledge of elastic moduli of non irradiated fuel is of first interest. In fact, Young, shear modulus and Poisson's ratio of uranium dioxide have to be introduced in numerical calculation codes. As these elastic moduli strongly depend on the volume fraction of porosity, the establishment of laws between Young, shear modulus, Poisson's ratio and porosity (volume fraction, morphology of the pores…) is very important and is the object of many studies. However, because of the small size of the UO_2 pellets and of their fragile behaviour [1, 2], classical techniques, compression tests for instance, are not applicable. In order to overcome this problem, other methods must be used. For this reason, for many years, we are developing specific ultrasonic methods (acoustic signature and micro-echography) especially dedicated to elastic moduli of non irradiated or irradiated fuel assessment, in a non destructive way [3-6]. The aim of this paper is to present a specific ultrasonic reflectometry method which is, on some points of view, better than acoustic signature and micro-echography [7-12].

Therefore, we will give theoretical elements concerning the ultrasonic reflection coefficient, likewise that the experimental device built, the data acquisition and processing used to measure the ultrasonic velocities (which are needed to calculate the elastic moduli).

Finally, a comparison between the evolution of elastic moduli with respect to the volume fraction of porosity obtained in the previous works [3-6], and with this new method will be performed and a link between the elastic moduli and the pores morphology will be pointed out.

2 Ultrasonic critical angle reflectometry

2.1 Basics on bulk samples

Ultrasonic Critical angle Reflectometry (UCR) is a non-invasive and non-destructive technique dedicated to the elasticity assessment in different types of materials [13-18]. When an ultrasonic wave encounters an interface between two materials, a proportion of the wave energy is reflected. This proportion (known as the reflection coefficient, *r*) depends on the difference between the acoustic impedances of the two materials, and on the incidence angle of the wave.

So, let us consider an incident longitudinal ultrasonic wave propagating in a liquid medium (density ρ and velocity c). This wave reaches the interface with a solid and generates one reflected and two refracted waves (Figure 1). For special values of angles (critical angles), surface waves can be generated [19]. The first surface wave, is a longitudinal wave and appears for θ_{CL}=Asin(c/V_L). The second is a shear surface wave with θ_{CS}=Asin(c/V_S). And at last, for θ_{CR}=Asin(c/V_R) the Rayleigh surface wave is generated.

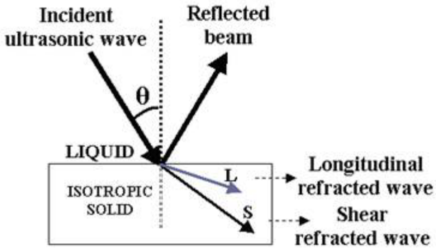

Fig.1 Description of the incident, reflected and refracted waves at an interface liquid-solid, for small values of the incidence angle θ.

These surface waves produce an alteration in the amplitude of the reflection coefficient (peaks/valleys) (Figure 2). From these peaks/valleys, one can calculate the longitudinal, shear and Rayleigh velocities using the following relationship:

$$V_i = \frac{c}{\sin(\theta_{ci})} \tag{1}$$

where the index i = L, S or R refers respectively to longitudinal, shear and Rayleigh waves.

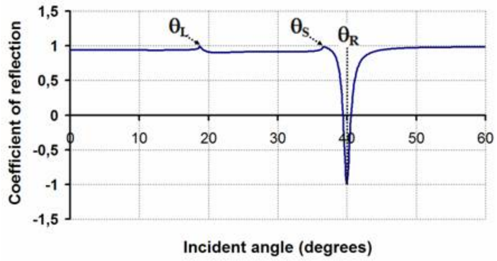

Fig.2 Example of a reflection coefficient r as a function of the incident angle θ_i for a bulk material. θ_L, θ_S and θ_R are the longitudinal, shear and Rayleigh critical angle respectively.

Then, using these velocities, the elastic constants (necessary to the comprehension of the mechanical behaviour of materials): the Young (E) and shear (G) modulus can be calculated by the following relationships:

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

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

where ρ is the density of the material, V_L and V_S are the longitudinal and shear velocities respectively.

2.2 Extension to guided modes: thin plates and Lamb waves

When a sample (plate with infinite dimensions in X-Y plane and thickness d) is thin enough to allow the penetration of ultrasonic waves to the opposite surface, and if its thickness is of the same order of magnitude as the wavelength ($d \leq \lambda$), guided waves are generated in the sample, and propagate in the X-Y plane. These guided waves are called Lamb waves [20-23].

As in optic guides, these modes are dispersive. In this case, an infinity of modes theoretically exist, which vibration nature and velocity are function of the product of the thickness by the frequency.

The reflection coefficient of these materials (Fig.3), because of dispersion, does not allow the direct assessment of velocities (longitudinal, shear and Rayleigh) in a simple way. In fact, all of the dispersive modes generated, have their own velocity and one has to use numerical simulations to adjust the curves relating the velocity to the product of the thickness by the frequency. As this curves are function of the longitudinal, shear and Rayleigh velocities of the equivalent bulk material, an accurate adjustment of the dispersion network leads to V_L, V_S, V_R and consequently to E, G [24].

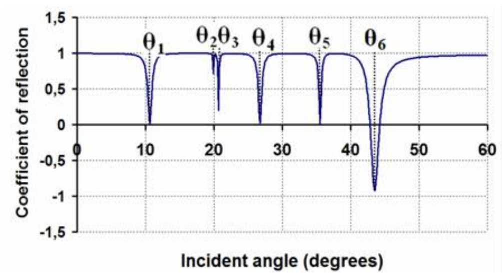

Fig.3 Simulation of a reflection coefficient as function of the incident angle θ_i for a thin plate for a frequency of 5MHz. V_L=4300m.s^{-1}, V_S=2355m.s^{-1} and thickness d=1mm.

An example of such a dispersion network is presented in Fig.4. The velocity of the lowest mode tends to V_R if the product of the thickness by frequency is high enough. We will see further that, for the ceramics pellets, namely UO_2, the velocity of the lowest mode tends to V_R if the product of the thickness by the frequency is superior than 4000 MHz.mm.

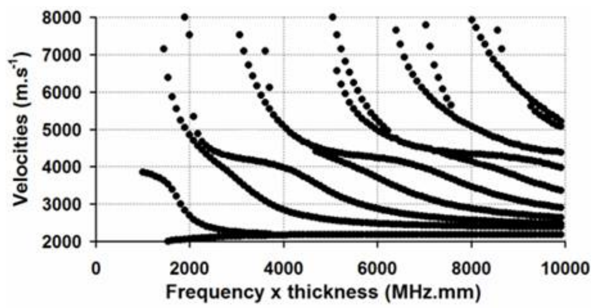

Fig.4 Simulation of the dispersion modes as a function of the product of the thickness by the frequency. V_L=4300m.s^{-1}, V_S=2355m.s^{-1}and thickness d=1mm.

3 Experimental setup

3.1 General Aspects

This experimental setup (Fig.5) is constituted by a trapezoidal system (which changes a movement of translation ensured by a stepper motor in a rotation movement), two plane ultrasonic transducers and a water tank that ensures the propagation of the waves from the sensors to the sample. The transducers are excited by a pulse/receiver (Parametrics Sofranel 5900 PR). The major frequency of the sensor is 5 MHz with a band-width of 6 MHz. So these sensors are efficient from 2 to 8 MHz. The received signal is acquired on a computer via a standard GPIB interface.

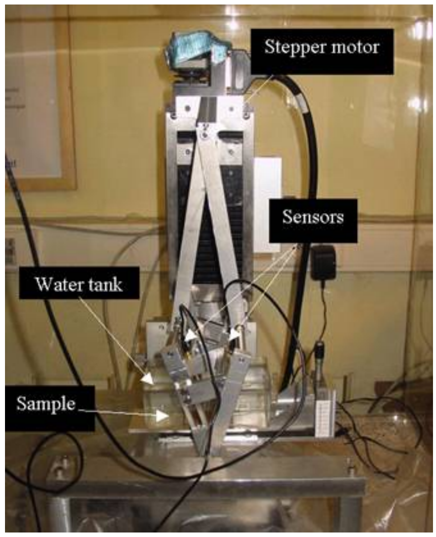

Fig.5 Photo of the experimental device.

3.2 Data acquisition and processing

After the echoes acquisition, the Fast Fourier Transform (FFT) of the received signals is performed on the whole band-width of the transducers leading to the evaluation of the amplitude of the signal for each frequency as a function of the angle. Hence, the reflection coefficient is obtained for all the frequencies of the band-width of the sensor. If the sample is massive ($\lambda \ll d$), the reflection coefficient is identical on all the band-width. Then, V_L, V_S and V_R are deduced as explained in section 2.1. If the sample is a thin plate ($\lambda \sim d$ or $\lambda > d$), the reflection coefficient is a function of the frequency. Thus, the procedure presented in section 2.2 is used. On a practical point of view, after the acquisition of the reflection coefficient, the dispersion curves are deduced by a software elaborated under Labview©. As said previously, the lowest mode velocity is equal to V_R if the frequency by thickness product is high enough (superior than 4000 MHz.mm), that is the case for all the samples studied, as it will be shown in the section 4. From this mode, V_R is directly deduced. Indeed, the critical angle for this mode does not depend on the frequency. Therefore, V_R is deduced from relation (1). Then, using a simulation code (made with Labview©) and the Rayleigh velocity, the longitudinal and shear velocities are deduced.

4 Results

The elastic constants of the samples (measured by microechography and V(z)) have been detailed in 2005 [6]. The volume fractions of porosity of all the samples are given in Table A. The samples thickness is around 1 mm.

Reference samples	Porosity measured (hydrostatic - %)	Reference samples	Porosity measured (hydrostatic - %)
2792	1.9±0.02	111	10.1±0.02
2796	2.75±0.02	113	12.08±0.02
2795	4.1±0.02	112	12.33±0.02
2793	4.95±0.02	115	14.8±0.02
2797	6.02±0.02	114	20.4±0.02

Table A - Characteristics of the samples and the corresponding porosity.

In the graphics shown in figures 6 and 7, the velocities and elastic moduli obtained by reflectometry are compared with the previous ones which can be found in [6][25] or more recently in [26]. In figures 6and 7, the solids lines represents adjustment of data proposed in [25]. The standard error made in the measurements i.e. the accuracy of the method is around ±50m.s^{-1} for V_L and V_S, ±20m.s^{-1} for V_R and ±2GPa for E and G. The results obtained by ultrasonic critical angle reflectometry method match very well with the results obtained by microscopy and echography demonstrating that reflectometry could constitute a powerful tool for nuclear fuel materials mechanical evaluation.

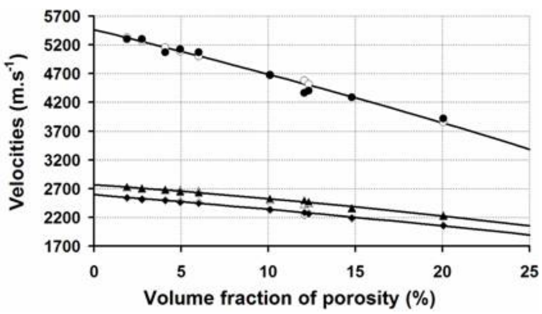

Fig.6 Comparison between longitudinal (●), shear (▲) and Rayleigh (◆) velocities by reflectometry (5 MHz), and the longitudinal (○), shear (△) and Rayleigh (◇) velocities found by the classical methods [25].

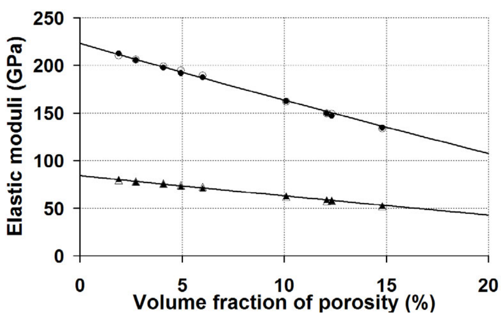

Fig.7 Comparison between Young (●) and shear (▲) modulus by reflectometry (5 MHz), and the Young (○) and Shear (△) modulus by classical methods [25].

Using these curves and a periodical homogenisation technique it also possible to evaluate the pores morphology as presented in [6]. We found that the pores can be considered as oblate one with a morphology factor around 0.4.

5 Conclusions

The main advantage of the reflectometry method is that it is possible to measure the reflection coefficient (which contains all the information on the mechanical properties), directly. Such a measurement is usually impossible using the other methods. Furthermore, difficulties existing in classical methods are overcome with ultrasonic critical angle reflectometry. For instance, if theoretically the V(z) allows to measure V_L and V_R, in experiments, only V_R is determined with precision. Hence, V_L or V_S have to be evaluated with echographic methods. This step of the manipulation is the most difficult one, because it is necessary to know with much precision the thickness. Moreover, the faces of the samples must be parallel.

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