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Study by acoustic microscopy of irradiated and non-irradiated uranium dioxide

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Abstract

The investigation of nuclear fuel properties at high burnup often yields difficulties in accounting for the fuel pellet fragmentation, the sharp properties gradient accross the pellet radius and the troubles met in the surface preparation of such a multiphase material. Nevertheless there is a strong need for modelling to assess to the local mechanical properties of the materials. This is the reason for EDF to participate in the developement of new investigation techniques, such as microindentation with the ITU, and microacoustic devices with the Montpellier University. Concerning the second technique, a feasibility phase has been achieved in the years 1996–1997 on irradiated fuels, showing many applications for this non-destructive techniques. The target of this first study was to demonstrate the feasibility of the Rayleigh wave measurement by acoustic microscopy on non-irradiated and irradiated uranium dioxides. This work has confirmed the normal functioning of the whole experimental device and particularly of the acoustic probes in a heavy irradiated environment. The works are still going on, combining microechography and microacoustics. It will be the matter for the next publication to come in 1999, more dedicated to the correlation between the Rayleigh wave velocity and the local porosity volume.

1. Introduction

In the water pressurised nuclear reactors, the fuel rod cladding is the first barrier against radioactive isotopes release. Its integrity must be demonstrated all along the fuel rod irradiation but also after irradiation during handling or dry storage. During the four or five years of irradiation, the zircaloy tubing is submitted to mechanic stresses induced by several mechanisms: the Zircaloy cladding creep down, the fuel pellet solid and gaseous swelling, the materials thermal expansion, mainly during power changes. The fuel pellet fragmentation and its global deformation remain the main components in the cladding loading. As a consequence, the knowledge of the evolution of the nuclear fuel mechanical properties during irradiation looks indispensable to evaluate properly the potential risk for clad failure. In the last 30 yr, the only data available for modelling have been the compressive tests conducted by different laboratories on fresh fuel pellet samples. However, the solicitation of the fuel pellet during power transient is more complex and the fuel chemistry evolution at high burnup let expect a drastic evolution of the fuel mechanical properties, mainly in the 'rim' region at high burnup. The first attempt done by Spino et al. in the ITU laboratories in measuring microhardness on irradiated pellets at room temperature [1,2] confirmed this assumption. In order to measure locally the fuel material visco-plastic properties at high burnup, the development of a microindentation technique has been launched in Karlsruhe, within a collaboration ITU/EDF [3]. The technique include the construction of a microindentator able to operate from

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room temperature to 1000°C and the development of a numerical indentation with the 3D EDF code named 'ASTER' in order to fit viscoplastic modelling parameters on the test results. These tests to be performed are: loading tests, creep tests and relaxation tests. Obviously, the interpretation of such experimental results can be made easier if another technique gives accurate values for the elastic local properties.

Up to now, two kinds of characterisation methods have been used to measure the elastic parameters: the static methods, as the mechanical compression test, and the dynamic ones as the ultrasonic velocity measurement or the determination of the resonance frequency [4]. However, these techniques give an overall evaluation of the pellet properties and cannot carry out local measurements, particularly on fractured materials. So, we propose within this presentation to apply another method to the nuclear irradiated fuel: the scanning acoustic microscopy (SAM) [5]. This brings noticeable profits, allowing a local and accurate non-destructive measurement. It is based on the determination of the propagation velocity of a surface acoustic wave, called the Rayleigh wave. This wave velocity depends on the elastic properties of the material. The aim of this work is to show the feasibility of the method to characterise non-irradiated and irradiated pellets of uranium dioxide. To do so, we have used an experimental device specially designed to operate in high activity areas. This apparatus has been previously introduced into a 'hot cell' of the GDL in EDF Chinon's laboratories, to investigate irradiated stainless steel [6].

2. Principle of acoustic microscopy measurement

An acoustic microscope is composed of a focused probe with the structure and operating described in Fig. 1.

The ultrasound generator is basically a piezoelectric transducer which converts the electrical excitation signal delivered by a frequency tone burst emitter into elastic ultrasound waves. These sound waves are focused onto the surface of the material to be tested through an acoustic lens. The acoustic link between the lens and the sample is made by immersing everything in water.

The acoustic microscope can be used in two ways: either using image formation or acoustic signature. In image formation mode, data are acquired at a constant height. The contrasts obtained are representative of the overall acoustic reflectivity of the material. In acoustic signature mode, we consider the interference between two types of wave (as shown in Fig. 1): wave A, which trajectory is not disrupted by the lens; and wave B, (the so-called Rayleigh wave) which propagates at the material/liquid interface. This surface wave is generated by an acoustic ray with a particular incident angle θ_r and is

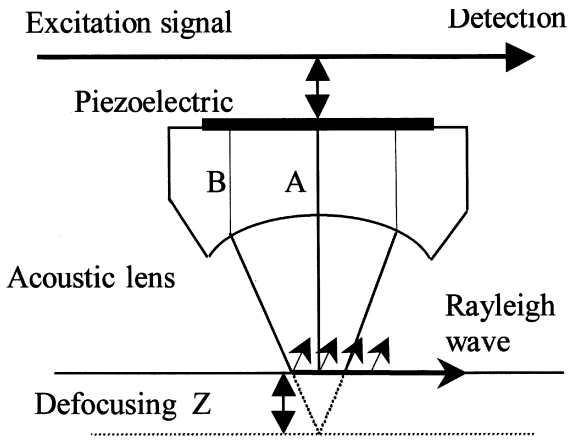

Fig. 1. Principle of the $V(z)$ mechanism.

remitted in the coupling fluid, collected then by the lens. The Snell law enables one to express the angle θ_r as a function of the Rayleigh velocity v_r and the wave velocity in the coupling fluid v_{liq}:

$$\sin(\theta_r) = \frac{v_{liq}}{v_r}. \tag{1}$$

As the acoustic lens is moved towards the surface of the material towards (increasing the defocusing), the measurement of the periodicity ΔZ of the interference between these two wave modes makes it possible to evaluate the propagation velocity of the surface waves, and thus induce the local mechanical characteristic of the material (the modulus of elasticity). It can be expressed in the following way [7]:

$$v_r = \frac{v_{liq}}{\sqrt{1-(1-(v_{liq})/(2F\Delta z_{r,l})^2}}, \tag{2}$$

where F is the frequency and ΔZ the period of the interference. The ΔZ period is determined precisely by the peak position of the acoustic signature Fast Fourier Transform (FFT) (see Fig. 2).

3. Study of a non-irradiated pellet

3.1. Echographic measurements

Firstly, we tried to evaluate the Rayleigh wave velocity of the non-irradiated fuel material using an echographic method. This technique consists in measuring the time of flight Δt between two ultrasonic echoes reflected by the parallel faces of the samples. The velocity 'v' of the wave can be calculated in the following way

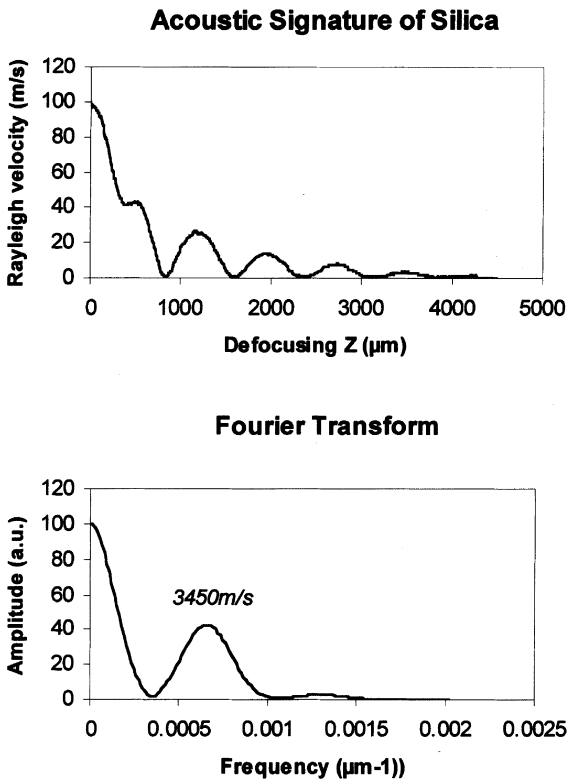

Fig. 2. Acoustic signature of a silica sample, acquired with water as coupling fluid at 15 MHz, and its FFT.

$$v = \frac{2d}{\Delta t},$$

where d is the thickness of the sample. Measurements of the longitudinal and shear velocities v_l and v_t were performed by a conventional ultrasonic echographe which allows to make only overall measurements. A 3 mm thick pellet was studied with a longitudinal and a shear plane transducers working at, respectively 4 MHz and 1 MHz.

From these experimental results and using the relation (3) [8], we can deduce the Rayleigh wave velocity (Table 1):

$$v_r = v_t \frac{0.715 - 0.995((v_t)/(v_l))^2}{0.75 - ((v_t)/(v_l))^2}. \quad (3)$$

Table 1
Ultrasonic velocity measured by time of flight method on non irradiated pellets

Longitudinal velocity (m/s)	Shear velocity (m/s)	Rayleigh velocity (m/s)
4545 ± 40	2720 ± 25	2477 ± 25

Table 2
Elastic constants of the non irradiated pellet calculated from the ultrasonic measurements and comparison with values from literature

Source	E (GPa)	G (GPa)	σ
Ref. [2]	200	75	0.321
Present work	189	77	0.22

The pellet elastic constants (Young's modulus, shear modulus and Poisson's ratio) can be determined from these measurements, using relations (4)–(6), assuming a sample mass density of 10 500 kg/m^3 (Table 2).

$$E = \rho\, v_t^2\, \frac{3v_l^2 - 4v_t^2}{v_l^2 - v_t^2}, \quad (4)$$

$$G = \rho\, v_t^2, \quad (5)$$

$$\sigma = \frac{2v_t^2 - v_l^2}{2\,(v_t^2 - v_l^2)}. \quad (6)$$

Table 1 shows that the longitudinal wave measured differs from the value found in literature (5100 m/s, [9]). This could be explained by the difference between the sample examined and the conventional fuel used in commercial reactor. Nevertheless, the value of the Young's modulus deduced from our measurements agrees within a range of ±5% with the values suggested by other authors [4].

3.2. Acoustic microscopy measurements

Measurements using acoustic microscopy have been carried out on this sample. Before performing acquisitions, we had to choose the operating frequency and the coupling fluid. For such porous material, the Rayleigh wave attenuation depends on the wavelength and thus, according to relation (7), on the operating frequency.

$$v_r = \lambda F, \quad (7)$$

where v_r is the Rayleigh velocity. If the wavelength is smaller or of the same order of magnitude than the material structures, the acoustic energy is scattered and the attenuation is strong. So, higher is the frequency, higher is the attenuation. Operating frequency is also in relation with the defocusing. Three arches at least have to appear in the acoustic signature to enable an accurate surface wave velocity evaluation. The arches period increases when the frequency decreases and as a consequence, lower is the frequency, deeper is the defocusing. The sample zone examined by the acoustic signature depends also on the defocusing: the deeper the defocusing, the greater the area diameter taken into account by the signature and consequently more global is the measurement. Nevertheless, because of the acoustic

energy concentration in the centre of the examined area, the zone involved for the measurement is much smaller than the surface which could be geometrically determined (Fig. 3). Finally, increasing the operating frequency, leads obviously to more and more localised measurements.

The acoustic properties of the coupling fluid are very important as well. Firstly, to generate the Rayleigh wave, the probe half aperture angle (about 50°) must be greater than the surface wave excitation angle θ_c. Relation (1) shows that this angle depends on the coupling fluid velocity. As the Rayleigh velocity of the dioxide uranium is low, the propagation velocity in the coupling fluid has to be slow as well. Moreover, the attenuation of the coupling fluid respect to the ultrasonic waves must be low to preserve the signal noise ratio. The methanol which agrees with these criteria has been chosen as coupling liquid.

Using this liquid, acoustic signature were recorded at 15, 50 and 130 MHz on the non-irradiated samples. The defocusing was set to obtain as many arches as possible in the acoustic signatures. The acquisitions were numerically processed to emphasise the oscillations: a pass band filter was used to eliminate the high frequencies due to the noise and the low frequencies produced by the response of the lens. In Fig. 4 experimental acquisitions at different frequencies are presented after the signal processing described above.

Table 3 presents the Rayleigh velocities measured on the non-irradiated pellet. The agreement between the average Rayleigh velocity measured by acoustic microscopy (2439 m/s) and the value deduced from echographic technique is quite good (Table 1). Thus, the ratio between the wavelength and the average pores diameter is high enough to work at 130 MHz.

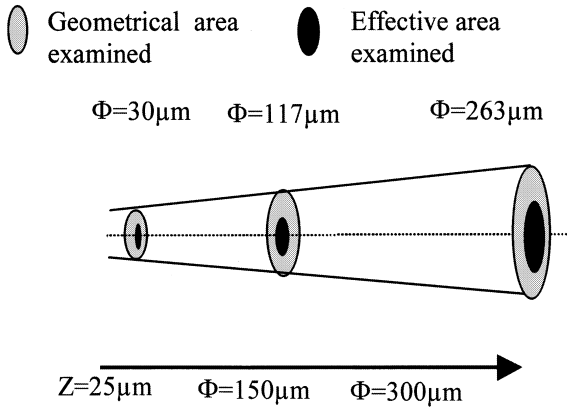

Fig. 3. Examined area of the sample by the acoustic probe at 130 MHz as a function of the defocusing and approximative diameter ϕ of the effective area taken into account by the measurement.

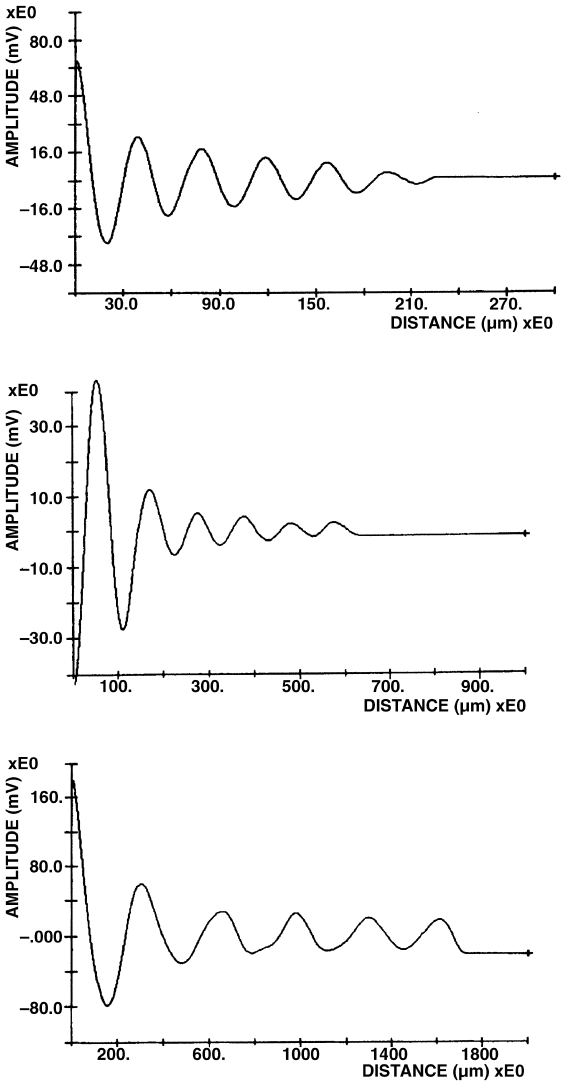

Fig. 4. Filtered acoustic signatures acquired at 130, 50 and 15 MHz on a non-irradiated pellet.

Table 3
Rayleigh velocity measured by acoustic microscopy on the non-irradiated pellet

Frequency (MHz)	15	50	130
Defocusing (μm)	2000	1000	300
Wavelength (μm)	160	50	20
Rayleigh velocity (m/s)	2417	2434	2467

We have also acquired acoustic images on this pellet using a high frequency probe operating at 930 MHz. This probe allows high resolution to be reached, around 1 μm. Thanks to that, one can easily distinguish the pores appearing in black on the bright background (Fig. 5). A manual counting on a part of the image has

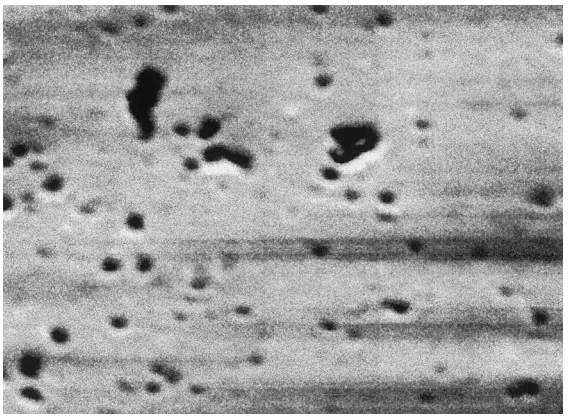

Fig. 5. Image of the non irradiated pellet near the surface at 930 MHz (64×48 μm²).

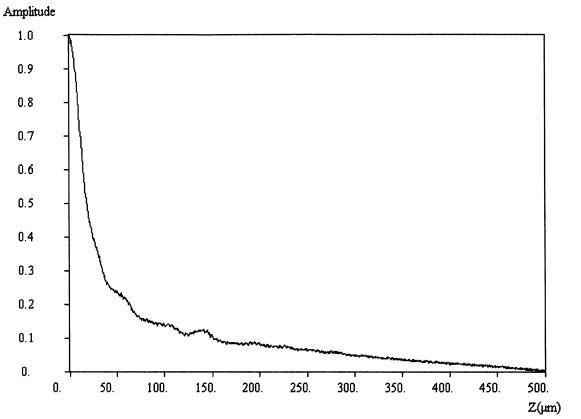

Fig. 6. Acoustic signature at 130 MHz on a 2 cycles irradiated pellet within the schematic effective measured area.

been performed to evaluate the porosity. This methodology gives a volume fraction porosity of about 4%. This value is low enough to express the evolution of the ultrasonic velocities and particularly the Rayleigh velocity by a linear correlation [10]

$$v_r = v_{r0}(1 - p), \tag{8}$$

where v_{r0} is the Rayleigh velocity of the non-porous body. Using this relation, the v_{r0} value for the uranium dioxide can be evaluated: 2538 m/s.

4. Study of the 2 and 5 cycles irradiated pellets

Measurements on 2 and 5 cycles fuel pellets have been carried out. In spite of the difficulty of the experiments, we could verify the normal functioning of the microscope in a strong irradiated area. Acoustic signatures have been first recorded at 130 MHz. Contrary to the previous acquisitions, the oscillations amplitude is very slight and the surface velocity could not be measured with enough accuracy (Fig. 6).

This can be explained analysing the image recorded at 130 MHz (Fig. 7). On this image, the average pores dimension (about 10 μm) is 10 times larger than the ones previously observed on the non-irradiated sample. Consequently, a lower operating frequency (15 MHz) should have been more appropriate. Moreover, this image confirms optical observations which show a strong micropores and cracks concentration.

So, a new acquisition protocol has been used: an image of the whole sample at 130 MHz (Fig. 8) allows one to select the areas for the acquisition of the acoustic signatures at 15 MHz. Thus, the acquisition avoids the fractured zones which could perturb the measurement of the material properties.

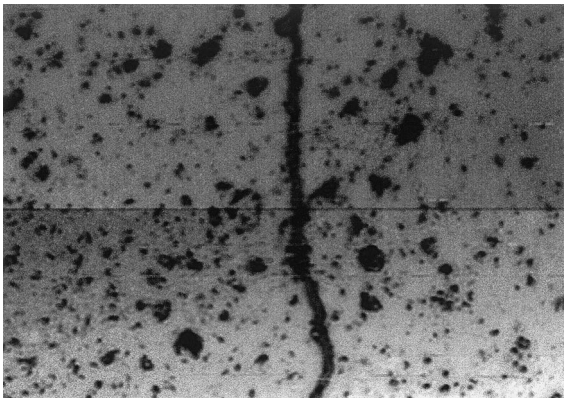

Fig. 7. Acoustic image at 130 MHz of the 5 cycles fuel pellet (1000×700 μm²).

Using this protocol, series of acoustic signatures have been recorded on the 2 and 5 cycles fuel pellets (with discharge burnup about 26 GWd/tM and 60 GWd/tM, respectively) according to the pellet radius. From these measurements, using relation (7) and the v_{r0} value determined previously, the local porosity volume has been evaluated. This is an approximation which supposes that the irradiated and non-irradiated v_{r0} values are similar. This assumes that the fission products have a slight influence on the elastic constants of the nuclear fuel. This assumption will be obviously checked in further works.

The porosity volume fraction have then been deduced from relation (7). Young's modulus can be expressed in the following way:

$$E = 2(1 + \sigma)\rho\, v_r^2 \left(\frac{1 + \sigma}{0.87 + 1.12\,\sigma}\right)^2, \tag{9}$$

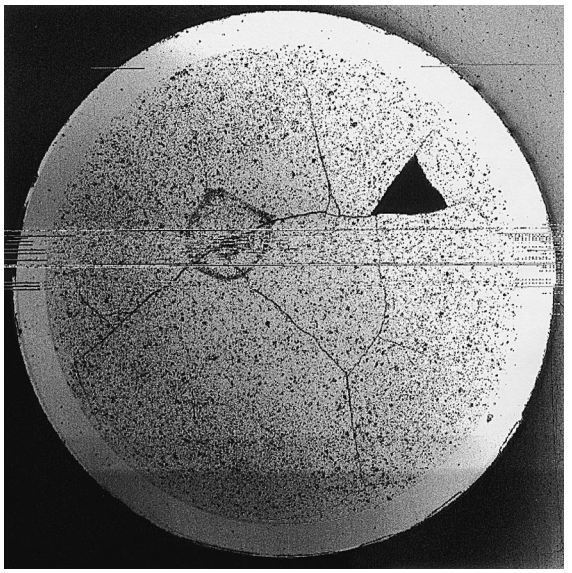

Fig. 8. Acoustic image at 130 MHz of the 5 cycle fuel pellet ($10\,000 \times 10\,000$ μm^2).

where ρ is the mass density and σ Poisson's ratio. The mass density can be expressed as follows:

$$\rho = \rho_0\ (1-p)(1-0.00065 \times BU), \tag{10}$$

where $\rho_0 = 10960$ kg/m^3 is the theoretical mass density and BU is the pellet burnup expressed in GWj/tU. The average rod burnup were, respectively 26 and 60 GWj/tM for the 2 and 5 cycles pellet. The Poisson's ratio of uranium dioxide depends on the volume fraction porosity [4].

$$\sigma = 0.321\ (1-1.03\,p). \tag{11}$$

Then, the Young's modulus can be calculated from relations (8)–(10) according to a diameter of the 2 and 5 cycles pellet (Fig. 9). The average porosity volume fraction determined in this way (about 11%) is much higher than the value measured by optical method (around 5% in most of the surface of a 60 GWd/tM irradiated pellet [11]). This could be explained by the rough determination of the v_{r0} value or/and by the choice of an empirical inadequate relation between the Rayleigh velocity and the porosity volume fraction for irradiated and non-irradiated fuel. Nevertheless, the increase of the porosity recorded at the edge of the pellet (up to 15%) (in the RIM area) agrees with optical results for the two pellet examined. With regard to the elastic parameters of the fuel, none values of Young's modulus on irradiated uranium dioxide are available in the literature and so, no comparison with our results cannot be made.

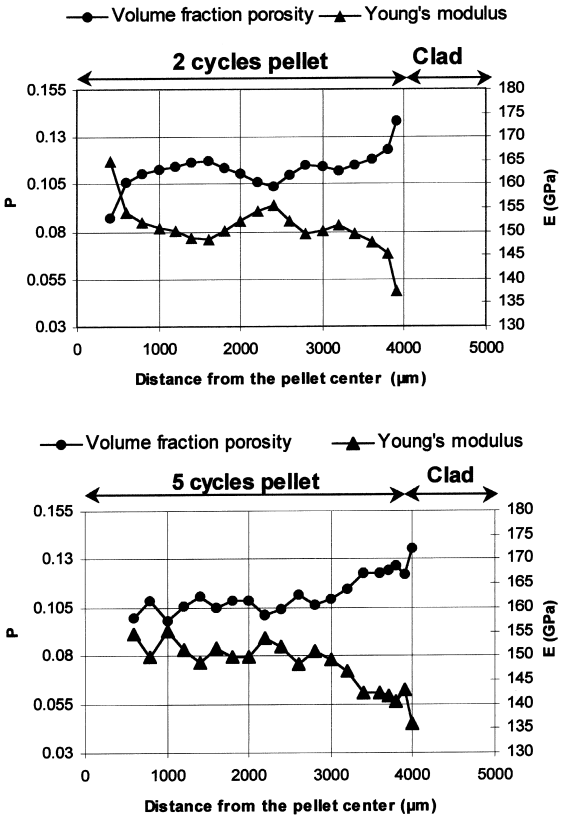

Fig. 9. Volume fraction porosity and Young's modulus according the distance from the centre of the 2 and 5 cycle pellet.

5. Conclusion

The target of this first study was to demonstrate the feasibility of the Rayleigh wave measurement by acoustic microscopy on non-irradiated fuel and on uranium dioxide samples irradiated up to 55 GWd/tM. This work has confirmed the normal functioning of the whole experimental device and particularly of the acoustic probes in a heavy irradiated environment. We could thus develop further more an original methodology to evaluate the local Young's modulus on an irradiated uranium dioxide.

The comparison with the value deduced from echography measurements confirm the validity of the Rayleigh velocity measurements performed on non-irradiated fuel material. On irradiated material, the first porosity volume fraction determined by acoustic microscopy did not agree with optical results from the literature.

However it is possible to improve the accuracy of the methodology, with a previous determination of a parametric relation between the Rayleigh velocity and the material properties. This job has already been performed

on archived fresh uranium dioxide samples in order to investigate the sensitivity of the Rayleigh wave to the material properties. Samples with variable porosity volume fraction, grain size or with additives concentration simulating the fission products are used. These works are still going on and have already shown promising results. It shall be the matter for the next publication to come in 2000. The following planning involves then measurements on irradiated uranium dioxide pellets. The parametric works previously performed on the non-irradiated samples and subsequently the calibration of the methodology will allow an accurate interpretation on the irradiated samples.

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