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ULTRASONIC METHODS APPROPRIATE FOR FURTHER PROGRESS IN NUCLEAR MATERIALS EXAMINATION

Ultrasonic Measurement of High Burn-up Fuel Elastic Properties

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ABSTRACT

The ultrasonic method developed for the evaluation of high burn-up fuel elastic properties is presented hereafter. The objective of the method is to provide data for fuel thermo-mechanical calculation codes in order to improve industrial nuclear fuel and materials or to design new reactor components. The need for data is especially crucial for high burn-up fuel modelling for which the fuel mechanical properties are essential and for which a wide range of experiments in MTR reactors and high burn-up commercial reactor fuel examinations have been included in programmes worldwide. To contribute to the acquisition of this knowledge the LAIN activity is developing in two directions: The development of an ultrasonic focused technique adapted to active materials study. This technique was used few years ago in the EdF laboratory in Chinon to assess the ageing of materials under irradiation. It is now used in a hot cell at ITU Karlsruhe to determine the elastic moduli of high burnup fuels from 0 to 110 GWd/tU. Some of this work is presented here.
The second on going programme is related to the qualification of acoustic sensors in nuclear environments which is of a great interest for all the methods which work in a hostile nuclear environment. Results concerning PZT piezoelectric sensors, under gamma irradiation, are presented in the previous paper given in this conference.

1. HIGH BURNUP FUEL ELASTIC PROPERTIES EVALUATION

1. Experimental method [1] [2]

An ultrasonic focused transducer is defocused towards the surface sample along the z axis. Interference is then created between the specular wave (normal ray in the coupling fluid) and the Rayleigh wave (which propagates on the surface). The signal received by the piezoelectric crystal versus z is then pseudo-periodic and is called the acoustic signature V(z) From the measurement of the pseudo-periodicity Δz, V_R is deduced using the relation 1. In this relation, V_{fluid} is the ultrasonic velocity in the coupling fluid and f is the operating frequency.

$$V_R = \frac{V_{fluid}}{\sqrt{1-\left(1-\frac{V_{fluid}}{2.f.\Delta z}\right)^2}} \qquad (1)$$

Then, the elastic properties are deduced with the relation (2), assuming that the Poisson's ratio is not far from 0.3 (this assumption has been checked on non irradiated and on irradiated pellets):

$$\begin{cases} E \approx 3\rho V_R{}^2 \\ G \approx 1.162\rho V_R{}^2 \end{cases} \qquad (2)$$

ρ is the density.

At the moment, we are using ultrasonic frequencies around 150 MHz, leading to an investigated area of less than 100 μm^2.

The experimental device developed and improved over a period of 6 years has been introduced in the ITU hot cells in 2001. First acquisitions on irradiated samples were obtained in December 2001

2. Non irradiated UO_2 study

Although the macroscopic elastic properties of the fuel are important for calculation codes, the elastic properties of the matrix (this means between the pores) are also of high interest from a scientific point of view to understand HBS or “Rim” structures formation. The ultrasonic waves are at a first estimate a function of porosity, metallic FP, FP in solid solutions, gazeous FP, defects, O/M ratio. This make the problem quite complex. From an experimental point of view, performing measurements on each parameter separately is impossible. That is the reason why we have simplified the problem. Using various publications we can assume that: the effect of simple porosities and the effect of surpressurized bubbles are quite equal, the irradiation defects have an effect of attenuation on the signal but does not change the velocity and it has been shown that grain size does not affect velocity. The effect of metallic FP and FP in solid solution can be evaluated with Simulated Fuels (SIMFUEL). Furthermore, measurements on variable O/M ratio samples have shown that this parameter was not on influence on water reactor fuel. In conclusion only the effect of burn-up and porosity has to be quantified.

2.1. Porosity

The results have been obtained on depleted Uranium with 0,3 % of ^{235}U and a bulk density of 10960 $Kg.m^{-3}$. For porosity ranging from 0 to 20 % we have found [2]:

$$V_R = 2593.(1 - 0{,}91p - 0{,}68p^2) \tag{3}$$

Both Berryman's model [3][4] and periodic homogenisation methods [5] lead to pore factor shapes equal to 0.25. On irradiated samples part of the initial porosity is disappearing by densification. Additional intra-granular porosity can then be created by coalescence of vacancies allowing the accommodation of gaseous fission products. These gases allow the stabilisation of the porosity. These intra-granular pores are “bubbles” and have a quasi spherical shape in order to minimize the energy of the system. The rim type pores are multi-faceted pores and can be considered as quasi spherical pores. Consequently in order to evaluate the intrinsic elastic modulus, we have developed a pore correction calculation assuming a combination of oblate and spherical pores [5]:

$$\frac{E}{E_0} = (1 - 2.000 * p_{spherical}) * (1 - 2.692 * p_{lenticular}) \tag{4}$$

$$\frac{G}{G_0} = (1 - 1.920 * p_{spherical}) * (1 - 2.458 * p_{lenticular}) \tag{5}$$

The total porosity volume (p) is given by :

$$p = p_{spherical} + p_{lenticular} \tag{6}$$

2.2. Effect of additives: study of Simulated Fuel

The samples on which the experiments have been performed were manufactured by ITU and by AECL, Chalk River [6]. Figure 2 gives the variation of the elastic constants versus the simulated burn-up. The global trend is a decrease of about 25 % from 0 to 100 GWd/tM followed by a stabilisation when solubility threshold is reached. Although only a few samples have been studied at low simulated burn-up, the decrease appears to start around 30 GWd/tM.

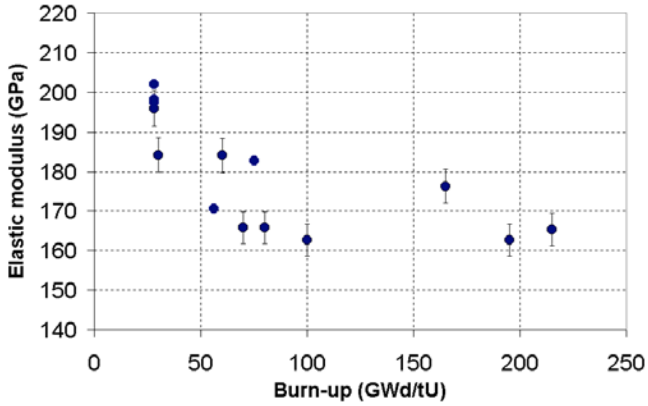

Figure 2 – Elastic modulus measured versus simulated burn-up

3. High burn-up fuel analysis : HBRP samples

Samples have been manufactured for the "High Burn-up Rim Project" which was started in 1993. Hundred eighty two 5 mm diameter wafers (UO_2, $(U,Ce)O_2$, $(U,Gd)O_2$ and $(U,Mg)O_2$) have been especially manufactured [7] [8][9]. Measurements on some of these samples is given in figure 3 together with the results obtained on SimFuel. The shear modulus is not reported here, but the global trend is similar to the Young's modulus. On this graph, a large number of data are reported, and the first impression is a large scattering. This is due to other influencing parameters such as the average porosity volume. However a global decrease of the Young Modulus can be noticed.

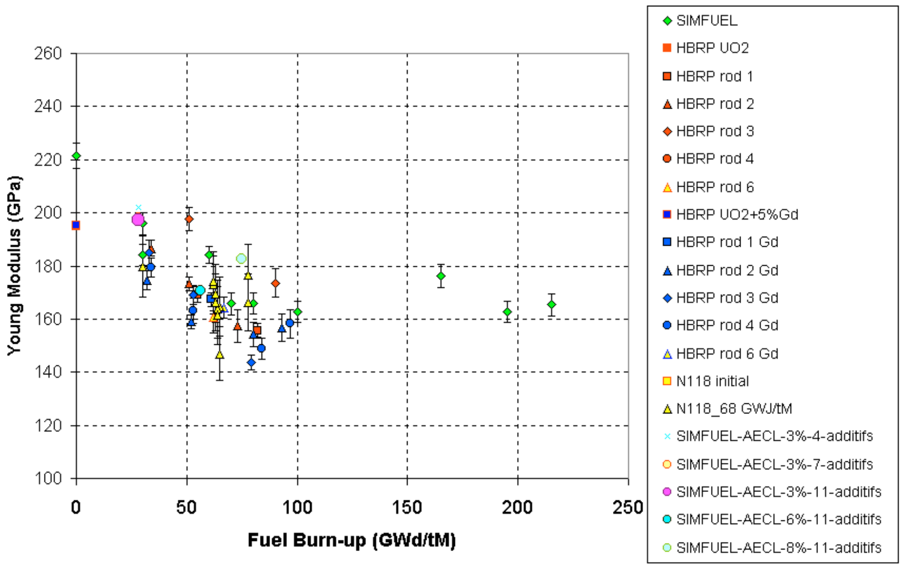

Figure 3 – Young modulus versus burnup for irradiated samples

At this point two important conclusions can be drawn concerning the global mechanical properties of the pellets :

- A decrease of 25 % of the elastic modulus between 0 and 100 GWd/tM
- A similar behaviour between Simfuel and irradiated material

In order to deduce the elastic intrinsic properties of the irradiated UO_2 matrix, apart from the porosity effect, all the data were extrapolated to 0% porosity using the relations (4), (5) and (6). This gives the following expression of the Young modulus :

$$E_{p=0} = \frac{3\rho V_R^{\,2}}{(1-2.00 * p_{spherical})*(1-2.69 * p_{lenticular})} \qquad (7)$$

With the following conditions :

$$\begin{cases} p_{lenticular} = p \\ p_{spherical} = 0 \end{cases} \quad \text{if } p \le p_{initial} \text{ (densification)} \qquad (8)$$

$$\begin{cases} p_{lenticular} = p_{initial} \\ p_{spherical} = p - p_{initial} \end{cases} \quad \text{if } p > p_{initial} \qquad (9)$$

The results have been divided in two sets of results: figure 4 shows the results for Simfuel and irradiated UO_2 and figure 5 gives the results for irradiated $(U,Gd)O_2$.

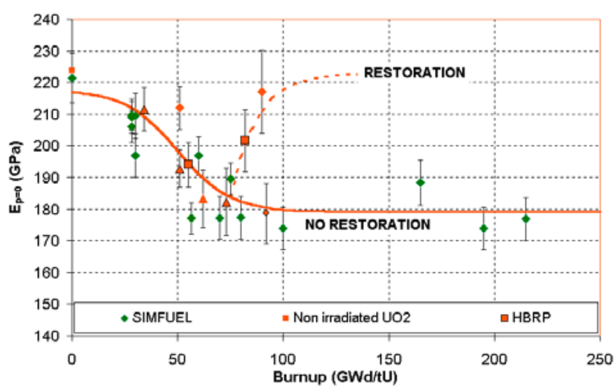

Figure 4

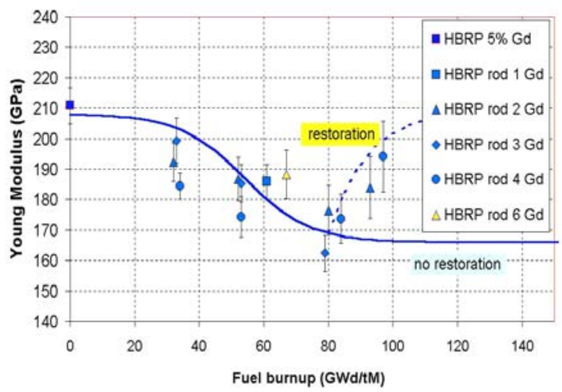

Figure 5

Figure 4 – Young modulus of UO_2 and Simfuel (reduced to 0 % of porosity)
Figure 5 – Young modulus of $(U,Gd)O_2$ (reduced to 0 % of porosity)

For the UO_2 samples like for the UO_2+$Gd_2 0_3$ samples, the trend shown with SIMFUEL is confirmed: an apparent stability of the elastic properties up to 30 GWd/tM (to be confirmed in the future by additional data on low burn-up samples), followed by a rapid decrease from 30 to 70 GWd/tM which, in turn is followed by a stabilisation above 100 GWd/tM. A possible explanation is that, around this burn-up the solubility saturation of the metallic and oxide fission products in the matrix is reached. This increase of precipitates in the matrix with the burn-up beyond 70 GWd/tM is then globally balancing the additional deterioration of the elastic properties of the fuel beyond this burn-up.

Above 70 GWd/tM, a restoration of the elastic properties is observed for all the samples where material restructuration has been reported. This restoration starts at a threshold burn-up around 75 GWd/tM for the UO_2 samples and 80 GWd/tM for the UO_2+5%Gd samples.

4 . Discussion

The method presented here is based on small perturbations of the material and gives an overall decrease of the elastic modulus with burn-up. However, other work conducted in ITU by Pujol et al [10] using a synchrotron method under very high constraints (1Gpa to 22 GPa) reports a different trend (increase from 0 to 60 GWd/tU and then stabilisation), consistent with Knoop indentation works. We have no definitive justification of such a discrepancy at this stage but we can propose some elements:

- When performed the acoustic method induces a small perturbation (traction/compression) in the material tested. In this case, the result obtained is likely comparable to an uniaxial traction test. Despite the fact that traction tests are not available on irradiated fuel it should be noticed that the available tests on other homogeneous materials confirm that acoustic and traction tests are comparable.
- when performing the Knoop or the indentation tests, a high compressive stress field is applied. It seems that the apparent elastic modulus deduced from this measurement could also be sensitive to the already present stress induced by the presence of Fission Products. This suggests that there is a fair difference of behaviour between compression and traction in elastic modulus measurement of irradiated fuel.

The synchrotron analysis is conducted with high compressive stress (pressure). The imposed constraint is ranging from 1 to 23 Gpa which is very high..In these conditions the response could be away from the linear elastic behaviour of the material. When compressing atoms, due to the potential barrier, the energy is not linear. What could be the behaviour between 0 and 1 Gpa ?

No clear conclusion can be drawn yet and discussions are underway between the two teams in order to find a way to explain discrepancies between the two methods.

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