# Nuclear Fuel Elastic Properties :
## Application of local non destructive acoustic methods on $UO_2$, UMo, HTR fuels

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## ABSTRACT

In 2001 we have presented a paper in which our approach and first results concerning the effect of porosity or simulated burnup on $UO_2$ mechanical properties were given. In this new communication, these data have been extended. On irradiated fuel, we show that from 0 to 100 GWj/tU a decrease (25 %) of the elastic moduli, mainly due to porosity and fission products occurs. Furthermore, the intrinsic elastic properties of the fuel (this means corrected from porosity) decrease between 30 GWj/tU and 75 GWj/tU. From 75 to 100 GWj/tU, an increase is then observed and the fuel recovers elastic moduli similar to these measured on non irradiated $UO_2$. This increase, which is not observed on SimFuel seems to be an intrinsic characteristic of high burn-up restructured fuels. More recently, measurements on new nuclear fuels such as UMo and HTR particles have been performed and show that acoustic microscopy could also give interesting information.

## INTRODUCTION

Since 1996, an increase of the average load discharge burn-up from 35 GWd/tM to 60 GWd/tM is in progress. In order to ensure a constant level of security, new simulation codes such as Cyrano 3 have been written to forecast the rods behaviour at high burn-up. Due to a lack of mechanical parameters at high burn-up, two projects have been launched. The first one, the micro-indentation, developed by Dr J.Spino in ITU will give indications on the evolution of the fuel thermal visco-plastic properties with irradiation from 0 to 1200 °C. The second technique, the acoustic microscopy gives the evolution of the elastic constants with the burn-up.

A second aspect of our work concerns innovative nuclear fuels for experimental reactors or for civil generation IV power plants. A collaboration has begun with the CEA of Cadarache. As all these fuels are inhomogeneous one or very small, only local methods such as acoustic microscopy, indentation are suitable for mechanical characterisation. In order to install a microscope in a non irradiated samples lab, tests have been performed to evaluate the capacities of acoustic approach on these composite fuels (UMo particles and HTR kernels).

## 1. ACOUSTIC SYSTEMS DEDICATED TO NUCLEAR FUEL INVESTIGATION

### 1.1. Recalls of elasticity

In order to measure the elastic constants from ultrasonic waves velocities, people usually measure the longitudinal ($V_L$) and the transverse ($V_T$) velocities [1]. To get local measurements, one has to use frequencies between 50 and 200 $MH_Z$. In this range, the transverse attenuation on irradiated fuel is so high that the transverse velocity is not obtained. Consequently, we have chosen to use another ultrasonic wave : the Rayleigh surface wave. Recently [2] [3] we have demonstrated that for $UO_2$ study , assuming that the Poisson's ratio is not far from 0.3 (this assumption has been checked on non irradiated and on irradiated pellets) , the elastic moduli E and G can be directly linked to $V_R$ as follows :

$$\begin{cases} E \approx 3\rho V_R^2 \\ G \approx 1.162\rho V_R^2 \end{cases} \quad (1)$$

### 1.2. Rayleigh wave velocity assessment and acoustic pictures acquisition

An ultrasonic focused transducer is gradually defocused towards the surface sample along the z axis. Interference are 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 $\Delta z$, $V_R$ is deduced. If during an X-Y scanning, the amplitude of the signal is acquired and transformed in colours, acoustical pictures are obtained. They reveal variations of mechanical properties and sub-surface micro-cracks. Such pictures are used to perform the V(z) on interesting zones. The experimental device developed and improved since 6 years has been introduced in the ITU hot cells in 2001. First acquisitions on irradiated samples have been operated in December 2001 [4] .

## 2. MAIN RESULTS

With the ultrasonic device, we can directly assess in a local way to the elastic moduli, on nuclear fuel cross section samples prepared for ceramography . Furthermore, as the ultrasonic waves are very sensitive to the volume fraction of porosity, acoustic approach could constitute an alternative tool for porosity assessment in a local way. If the porosity is measured we can also deduce the elastic properties of the matrix (between the pores). Such information could be very interesting for "Rim" or "HBS" structures understanding. On irradiated fuel, the problem is quite complex because we have to determine the effect of the different parameters modified by the irradiation : fission products in solution, precipitates, gaseous fission products, irradiation defects, stoechiometry evolution , grain size. Using literature we can assume that :

the effect of simple porosity and the effect of surpressurized bubbles are quite equal, the irradiation defects have an effect on attenuation and not on the velocities, the effect of grains size has no effect on velocities , the effect of metallic FP and FP as solid solutions can be evaluated with Simulated Fuels . Concerning the O/M ratio we have demonstrated that this parameter could be neglected for $UO_2$ fuel [3]. Consequently, we can assume that the ultrasonic waves velocities are only influenced by the porosity and a global effect of burnup. These two parameters are now studied in the following paragraphs.

## 2.1. Porosity

The results on depleted Uranium with 0,3 % of $^{235}U$ and a bulk density of 10960 $Kg.m^{-3}$ presented in reference [5] have been extended to 20 % of porosity. We have found :

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

Then, using periodic homogenisation technique, we have found general relations accounting for the pore shape [6]. For example for the Young modulus, we have :

$$\frac{E}{E_0} = 1 - (0.36 * \ln^2(w) + 2.000) * p \qquad (3)$$

$E_0$ is the intrinsic Young modulus, p the relative porosity volume, $w$ the ratio between the two axis length of a lenticular shape : $w = \frac{c}{a}$ ; for a spherical shape, $w = \frac{R}{R} = 1$ .

On irradiated samples part of the initial porosity is disappearing by densification. Then additional intra-granular porosity can be created by coalescence of vacancies and accommodate the gaseous fission products. These gases allow the stabilisation of this 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 well as quasi spherical pores. Consequently in order to evaluate the intrinsic elastic modulus, we have to make a pore correction assuming a combination of oblate and spherical pores :

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

with the total porosity volume (p) given by : $p = p_{spherical} + p_{lenticular}$ (5)

## 2.2. Effect of additives : study of Simulated Fuel

The samples on which the experiments have been performed have been manufactured in ITU and in Chalk River (AECL) [7]. On figure 1 we have reported the variation of the elastic constants versus the simulated burn-up. To establish accurate conclusions, one should have to extrapolate these results to 0 % porosity. But as all these SimFuel had quite the same volume porosity fraction, a partial conclusion can be established right now : the global trend is a decrease of about 25 % from 0 to 100 GWd/tM and then a stabilisation when solubility threshold is reached. Even if only a few samples have been studied at low contents, the decrease appears to start around 30 GWd/tM.

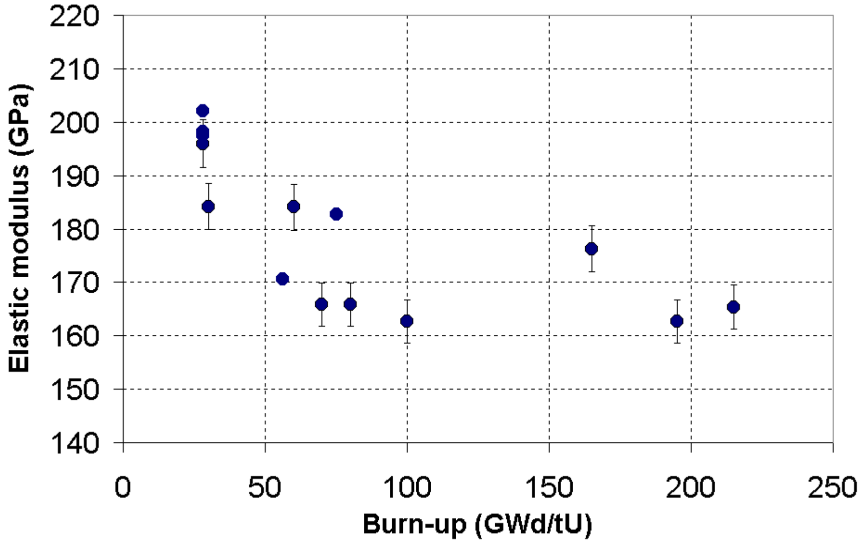


FIGURE 1 – ELASTIC MODULUS MEASURED VERSUS SIMULATED BURN-UP

### 2.3. High burn-up fuel analysis : HBRP samples

2.3.1. HBRP fuel wafers

The first set of samples have been manufactured for the international project : High Burn-up Rim Project which has started in 1993. The main goal of this project was to obtain a better understanding on the conditions of the high burn-up microstructure transformation so called "RIM" structure or "cauliflower" structure, and the evolution of the physical properties of such high burn-up structure (HBS). Consequently, 182 wafers ($UO_2$, $(U,Ce)O_2$, $(U,Gd)O_2$ and $(U,Mg)O_2$), 5 mm diameter, have been especially manufactured [8] [9][10].

2.3.2. BR3 N118 fuel rod samples

The second samples have been extracted from the CEA N118 fuel rod irradiated in the BR3 core from 1976 to 1983. The cladding is a recristallized zircaloy 4 and the fuel $UO_2$ standard pellets. The average fuel rod burn-up is about 56 GWd/tM and the axial peak burn-up is about 68 GWd/tM. Detailled characteristics of this rod can be obtained in reference [11].

2.3.3. Observations and discussion on the data acquired

The plot obtained is presented on figure 2 conjointly with the results obtained on SimFuel. The shear modulus is not reported here, but the global trend is similar to the Young's modulus one. On this graph, a large number of points are reported, and the first impression is a large scattering of the data base. This is due to the presence of other influent parameters behind this plot such as the average porosity volume. Looking closer, a global trend can be however noticed.

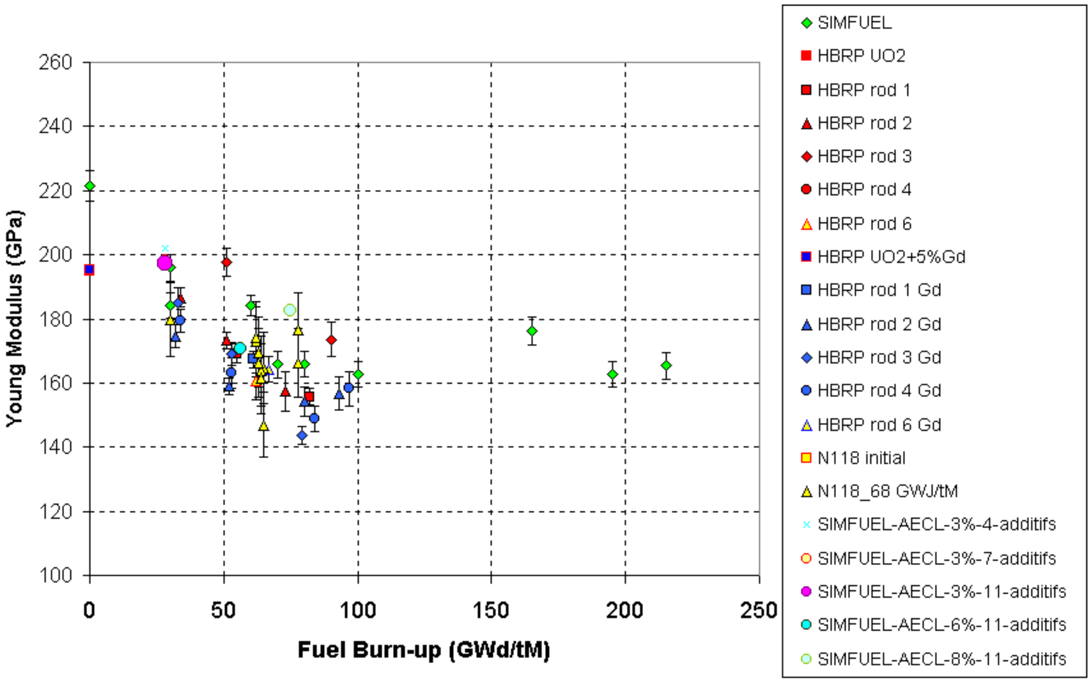


FIGURE 2 – YOUNG MODULUS VERSUS BURNUP FOR IRRADIATED SAMPLES

Two important conclusions can be deduced concerning the global mechanical properties of the pellets :

- A decrease of 25 % of the elastic moduli up to 100 GWd/tM
- A similar behaviour between Simfuel and irradiated material

As reported before in this paper, this elastic properties degradation has several origins : modification of the material porosity volume, accumulation of fission products in substitution or interstitial position, accumulation of ceramic and metallic precipitates, modification of the oxygen sub-lattice, global accumulation of irradiation defects.

In order to deduce the elastic intrinsic properties of the irradiated $UO_2$ matrix (Fig 3 and 4), apart the porosity effect, we have reduced all the data to 0% porosity using the relations (2), (3) and (4). They give for the intrinsic Young modulus :

$$E_{p=0}=\frac{3\rho V_R^2}{(1-2.00*p_{spherical})*(1-2.69*p_{lenticular})} \qquad (6)$$

With the following conditions :

$$\begin{cases} p_{lenticular} = p \\ p_{spherical} = 0 \end{cases} \quad \text{if } p \leq p_{initial} \text{ (densification)} \qquad (7)$$

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

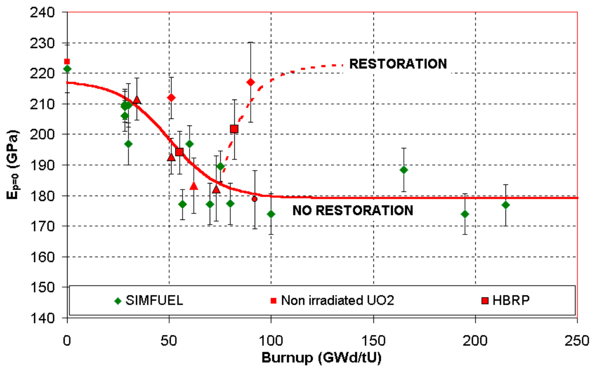


FIGURE 3

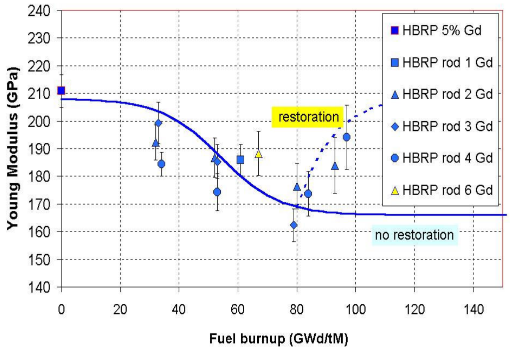


FIGURE 4

FIGURE 3 – YOUNG MODULUS OF $UO_2$ AND SIMFUEL (0 % OF POROSITY)
FIGURE 4 – YOUNG MODULUS OF $(U,GD)O_2$ (0 % OF POROSITY)

For the $UO_2$ samples and the $UO_2$+Gd sample as well, the trends confirm what has been observed for the SIMFUEL samples : an apparent stability of the elastic properties up to 30 GWd/tM (to be however confirmed by furthermore data in the future on low burn-up samples), and then, a rapid decrease from 30 to 70 GWd/tM followed by a stabilisation above 100 GWd/tM. This can be explained by a solubility saturation of the fission products in the matrix and then an increase of the metallic and ceramic precipitates which have a lower effect on the global elastic properties.
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.

## 2.4. 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, an other work conducted in TUI by Pujol et al [12] using a synchrotron method under very high constraints (1Gpa to 22 GPa) reports a different trend, consistent with Knoop indentation works. This work shows an increase of the elastic modulus with burn-up instead of a decrease. This means that instead of a softening, these alternative techniques show a global hardening. Nevertheless, a stabilisation is obtained in both case when the average fission product solubility limit is achieved, and a consistency is observed within the Simfuel set. Therefore, how to explain such a discord ? We have no definitive answer at this stage but propose some elements in the following :

- Acoustic method induces a small perturbation (traction/compression) and measures probably a “theoretical” material Young modulus. In this case, the result obtained should be consistent with a uniaxial traction test if such a test was available on irradiated samples.

- Accounting for the high compressive stress field induced when performing the Knoop test or the indentation it appears that the apparent elastic modulis is sensitive to the internal stress of the material induced by the presence of Fission Products, mainly the Xenon. We could conclude for a difference of behaviour between compression and traction.

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

What is the best technique to assess to the true elastic coefficient really needed for the codes analysis ? The answer is not so obvious. Works have to go on.

## 3. FIRST RESULTS ON INNOVATIVE NUCLEAR FUELS : $UM_O$ AND HTR

### 3.1. HTR particles characterisation [13]

HTR fuel is basically based on spherical coated particles dispersed in a graphite element named compact in the US concept and pebble in the German one. Classical dimensions of US cylindrical compacts are about 1 to 2 cm diameter and 5 cm long. The overall diameter of the German spherical element is 6 cm with a 0.5 cm thick fuel free shell.
The HTR coated particle consists of a $UO_2$ kernel which is surrounded by a porous pyrolytic carbon buffer layer, an inner dense pyrolytic carbon layer (IPyC), a silicon carbide layer (SiC) and an outerdense pyrolityc carbon layer (OPyC). The diameter of the $UO_2$ kernel is about 500 µm whereas the diameter of the coated particle is about 900 µm. Each layer in the TRISO particle design has a specific function in fuel performance and fission product retention. The buffer layer provides a void volume for gaseous FP and accommodates kernel swelling. The dense IPyC reduces tensile stress on SiC and acts as a diffusion barrier to metallicFP. The SiC layer ensures leak tightness to metallicFP during normal and accidental situations. The dense OPyC layer reduces tensile stress on SiC as IPyC and provides bonding surface for matrix material.

Modelisation of the whole coating in service conditions is necessary to define how to prevent crack initiation and leak. Consequently, FEM calculations are performed and require a perfect knowledge of the elastic parameters of each part of the particles.

Acoustic microscopy has been applied in the kernels using a 140 MHz operating frequency. For the moment no result can be obtained in the layers because they are to small. In this case other methods (indentation for instance) can be employed. In order to evaluate the effect of the fabrication process on elastic moduli, measurements have been performed on three batches presenting different microstructures (porosity, grain size...). Using the results obtained on classical $UO_2$, the Rayleigh wave velocity has been measured and used to calculated the Young modulus of the $UO_2$ kernels. The results (see tab. 1) show that there is no influence of the fabrication process on the elastic properties of TRISO kernels. Furthermore, these results are in keeping with literature data available for $UO_2$ pellets produced by classical powder metallurgy route.

| | $V_R$ | E |
|---|---|---|
| Batch 1 | 2545±20 | 205 ±5 |
| Batch 2 | 2477±25 | 192 ±6 |
| Batch 3 | 2500±40 | 198 ±5 |

TAB 1 – YOUNG MODULUS MEASURED ON $UO_2$ KERNELS [13]

### 3.2. UMo composites study [14]

For the new experimental reactor RJH (reactor Jules Horowitz) to be built in France, innovative UMo fuels are designed in CEA and manufactured in CERCA - Romans in France. These UMo fuels are made of an aluminium matrix in which UMo particles are dispersed (Figure 5). High frequency acoustic microscopy (600 MHz) has been applied in Al plates and in the matrix. In the two cases, the same value of Young modulus has been obtained : 72 GPa.

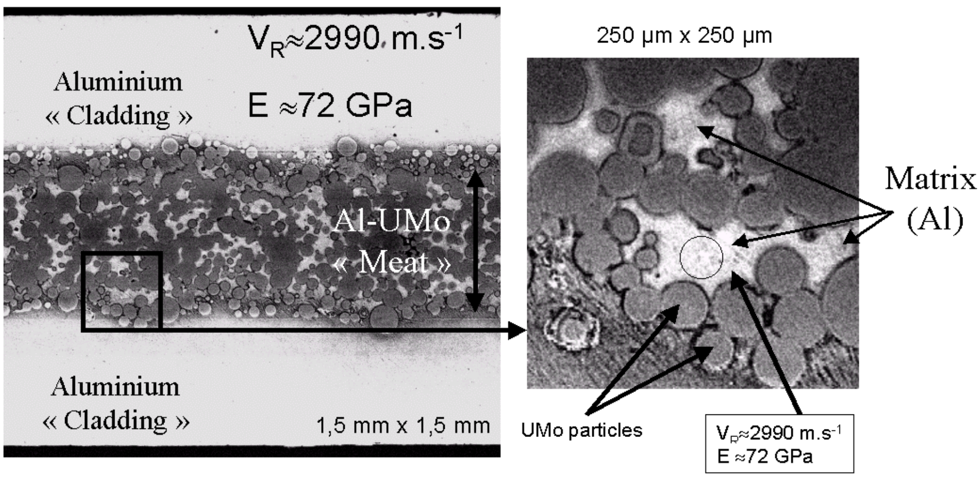


FIGURE 5 – HIGH FREQUENCY 600 MHZ ACOUSTIC PICTURE OF $UM_O$ COMPOSITE

Then, using macroscopic echographic technique the Young modulus of the whole composite has been estimated leading to $Ex \approx Ey \approx Ez \approx 80 \pm 20$ GPa and showing

a good isotropy of the sample [14]. The large error on this first result is mainly due to high uncertainties on each part thickness. In the "meat", a rough estimation of E between 100 and 130 GPa can be given and is quite surprising regarding literature data : a value of 57 GPa is recommended.
In order to explain this result, experiments have been performed in UMo particles thanks to Atomic Force Microscopy in resonant contact. This method is based on the measurement of the resonant frequency of the AFM cantilever settled on the sample. Using calibration laws between the Young modulus and the resonant frequency, one can deduce the elastic modulus of an unknown material. In the Al matrix a value of 70 GPa has been obtained and is in good agreement with ultrasonic result. In UMo particles a value of 180 GPa has been obtained. Such a value is also quite surprising regarding literature data. Indeed a value of 80 GPa is recommended for massive UMo. This hardening has also been observed using indentation technique measurements, performed in the CEA (Restricted diffusion). Consequently, high values obtained for $E_{meat}$ is due to an hardening of UMo particles. These discrepancies between literature and these measurements are certainly due to fabrication process and to the presence of uranium phase $\alpha$.

## CONCLUSION

The elastic data acquired and presented in this publication are the very first collected on irradiated fuel or more widely on highly active materials. Some first conclusions on the elastic properties evolution with the Burn-up can be given:

- a linear decrease of the global elastic properties with an apparent stabilisation above 100 GWd/tM to be confirmed,
- above a threshold burn-up (70 for $UO_2$ and 80 for $UO_2$+5%Gd), a restoration of the intrinsic elastic properties is observed as soon as the vacancies and gas mobility is acquired. It can be thermal activated mobility or rim structure formation.
- the consistency between the irradiated $UO_2$ samples and the SIMFUEL samples let expect that the decrease of the elastic moduli with burn-up is mainly due to the fission products present in the matrix. The restoration should be mainly due to the Xenon redistribution.

Comprehensive works must go on. More data is needed in the future in order to enlarge the basis of these first conclusions. The method presented here is based on small perturbations of the material and gives an overall decrease of the elastic properties with burn-up. Other works, conducted by Dr Jose SPINO's team and using a synchrotron method under very high constraints (> 1GPa) report a different trend. Discussions are engaged between the two teams in order to clarify the discrepancy between the two methods.

Concerning innovative fuels, capacities of acoustic microscopy have been clearly demonstrated. More results will be obtained since an acoustic microscope is being installed in Cadarache in the next months.

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