Scanning Acoustic Microscope: An Advanced Technique for the Mechanical Characterization of Irradiated Nuclear Fuel

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Abstract. The characterization of mechanical properties constitutes a key challenge for the study of nuclear fuel behaviour under conditions relevant for safe in-pile operation. This applies to both conventional and advanced fuel concepts. At JRC-ITU new tools are being developed to extend the knowledge on the thermo-mechanical behaviour of the fuel during irradiation. In particular, a scanning acoustic microscope was recently tested in hot cell for the measurements of elastic constants of irradiated fuel (in the frame of collaboration with EdF). The technique has been adapted to the conditions characterizing the hot cell environment and the configuration of typical irradiated fuel samples. The results obtained so far are very promising and indicate that this could become a very useful tool not only for the measurement of elastic properties, but also as complementary method to assess bulk properties and initial conditions of fuel samples destined to various types of analysis.

1. WORKING PRINCIPLE

Elastic properties of UO_2 have been studied for a long time. Their determination is relevant particularly to reactor-transients for which creep relaxation is unlikely to happen. This type of situation produces pellet–cladding mechanical interaction (PCMI) and possibly cladding failure. This has been a subject of concern for all reactor types using UO_2 or mixed-oxide fuel. Elastic properties, combined with thermal expansion, determine the level of stress in the fuel and consequently in the cladding when interaction occurs. The pellet stiffness evolves with burn-up because of the pellet cracking (macro and micro cracks) and also with the bulk intrinsic elastic moduli. Under this situation Young's modulus, E, is the most important parameter to assess for code calculations.

It was found that the elastic modulus in oxide fuel decreases both with increasing temperature and porosity volume fraction. The fractional porosity in UO_2 has been extensively studied at room temperature and in the range 90–100 % of theoretical density. Although data on mixed oxides are scarce, it was found that the elastic modulus versus temperature followed the same pattern as for UO_2, but increased with the plutonium content. The most reliable estimate seems to be 3% increase for a 20% PuO_2 fuel.

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The influence of the O/M ratio on Young's modulus was studied, suggesting that Young's modulus is higher for stoichiometric fuel, but decreases with hyper- or hypo-stoichiometric shifts. The influence of stoichiometry is difficult to quantify, because other parameters (e.g., the fabrication route, grain size, etc.) interfere.

1.1. Acoustic waves for elastic modulus of nuclear fuel evaluation

Because of the fractured pattern of irradiated pellets and the variation of mechanical properties along the radius, classical methods (e.g., mechanical traction, global echography) are not applicable and only novel non-destructive and non-invasive methods, applied on a very small area of the sample, such as high-frequency acoustic microscopy or indentation, can give useful results.

The theory of elasticity in an isotropic material such as sintered UO_2 shows that only two ultrasonic velocities are needed to assess the elastic constants. Usually the longitudinal (V_L) and the transverse (V_T) velocities are measured. To perform local measurements, frequencies between 50–200 MHz have to be used. In this range of frequencies, the transverse attenuation in irradiated fuel is very high and the transverse velocity cannot be measured. Furthermore, for transverse waves the coupling between the ultrasonic sensor and the sample has to be made with a viscous liquid. Working on irradiated samples in hot cells with such liquids seems difficult. An alternative possibility is to use another ultrasonic wave, which is the Rayleigh surface wave. Its velocity will be called V_R. With the measurement of V_L and V_R, the transverse velocity V_T is deduced using the Eq. (1),

$$V_T^{\,8} - (V_L^{\,2} + V_R^{\,2})V_T^{\,6} + \frac{3}{2}V_L^{\,2}V_R^{\,2}V_T^{\,4} - \frac{1}{2}V_L^{\,2}V_R^{\,4}V_T^{\,2} + \frac{1}{16}V_L^{\,2}V_R^{\,6} = 0 \tag{1}$$

Available in the range

$$0 \le V_T \le \frac{V_L}{\sqrt{2}} \tag{2}$$

The elastic moduli (E and G) and the Poisson's ratio ν are then calculated as follows:

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

$$G = \rho V_T^{\,2} \tag{4}$$

$$\nu = \frac{1 - 2\left(\frac{V_T}{V_L}\right)^2}{2\left(1 - \left(\frac{V_T}{V_L}\right)^2\right)} \tag{5}$$

Where ρ is the mass density of the material.

For measurements on nuclear fuel oxide, the Rayleigh surface wave velocity is obtained from the acoustic signature (see Section 1.2). Even if theoretically the longitudinal wave velocity can also be assessed with the acoustic signature, in most cases the wave attenuation is too large to permit the detection of the longitudinal velocity in the signature. Therefore, this last velocity has to be measured using a classical echographic technique on a fuel slice about 1 mm thick: with the knowledge of thickness and thanks to the measurement of the propagation time of the ultrasonic wave in the sample, the velocity is simply deduced.

1.2. Rayleigh wave velocity assessment

An ultrasonic focused transducer made of a piezoelectric crystal excited with a sinusoidal voltage, settled on a silica rod in which a spherical lens has been designed is gradually de-focused towards the sample surface along the *z* axis. Thanks to piezoelectric effect, mechanical (ultrasonic) waves are generated and interferences then created between the specular wave (normal ray) and the Rayleigh wave (which propagates on the surface). The ultrasonic signal received by the piezoelectric crystal versus z and reconverted in voltage 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 following relation:

$$V_R = \frac{V_{fluid}}{\sqrt{1-\left(1-\frac{V_{fluid}}{2.f.\Delta z_i}\right)^2}} \tag{6}$$

Where V_{fluid} is the ultrasonic velocity in the coupling fluid and f is the operating frequency. We recall that the coupling fluid (water, ethanol or methanol for $V(z)$ experiments) ensures the propagation between the sensor and the sample. Using high-frequency ultrasonic waves (around 100 MHz), the zone investigated is less than 100 μm, assuming special signal processing is used.

An acoustic image can also be obtained by recording the amplitude of the signal during an x-y scan and transforming it to false colours. It reveals variations of mechanical properties and sub-surface micro-cracks. Such pictures are used to identify adequate zones for $V(z)$ measurements.

1.3. Adaptation of the method to irradiated fuel

When samples are already embedded, echographic measurements are impossible as only one free surface is available. Then we have tried to find a way to assess E and G with V_R alone. Indeed for V_R evaluation, only one free surface is needed. Regarding Eqs (1–5) and assuming that the Poisson's ratio is not very different from 0.3, the elastic moduli E and G can be directly related to V_R as follows:

$$\nu \approx 0.3 \Rightarrow \begin{cases} E \approx 3.000 \rho V_R{}^2 \\ G \approx 1.162 \rho V_R{}^2 \end{cases} \tag{7}$$

In isotropic materials, such as sintered UO_2, the elastic constants can be determined by measuring only two ultrasonic velocities. Usually, the longitudinal velocity V_L and the transverse velocity V_T are measured. At the frequencies used for local measurements (between 50–200 MHz), the strong transverse signal attenuation makes the transverse velocity measurement impossible. Instead, the Rayleigh surface wave can be considered. The transverse velocity can then be trivially calculated from the longitudinal velocity and the Rayleigh velocity V_R. The elastic moduli E and G and Poisson's ratio ν are then calculated from the relations

$$E = \rho V_T^2 \frac{3V_L^2 - 4V_T^2}{V_L^2 - V_T^2} \tag{8}$$

$$G = \rho V_T^2 \tag{9}$$

$$v = \frac{1 - 2\left(\frac{V_T}{V_L}\right)^2}{2\left(1 - \left(\frac{V_T}{V_L}\right)^2\right)}, \tag{10}$$

where ρ is the density of the sample.

The operating principle of the acoustic microscope is shown in Fig. 1.1.

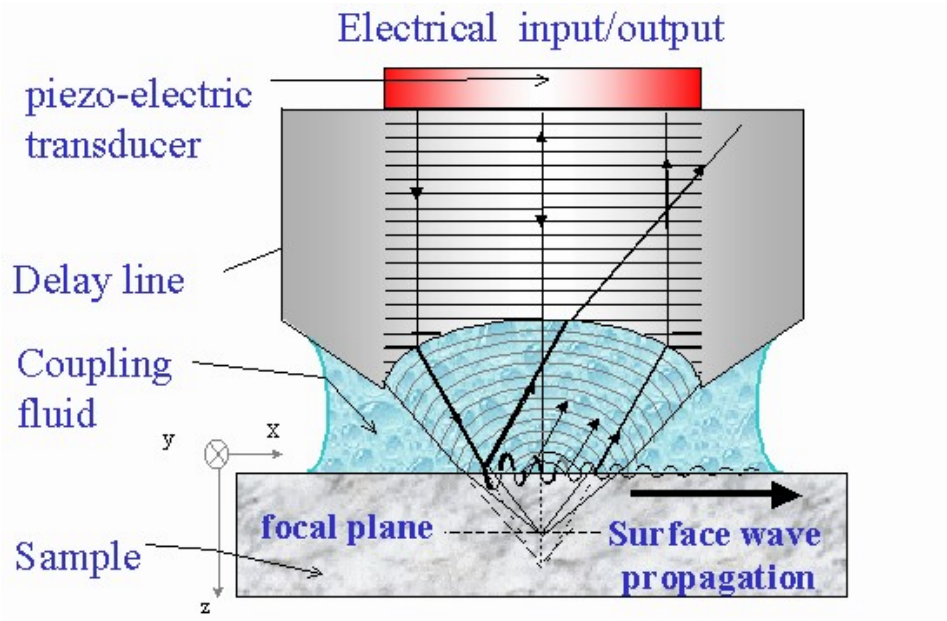

FIG. 1.1 Principle of the acoustic microscope.

The acoustic signal from a transducer is focused by a spherical lens and coupled to the sample by a coupling fluid. A so-called "acoustic signature" is acquired by gradually de-focusing the focused wave of the ultrasonic transducer towards the sample surface. Interference is created between the specular wave (normal ray in the coupling fluid) and the Rayleigh wave which propagates along the surface. The acoustic signature is the pseudo-periodic signal received by the piezoelectric crystal versus the z coordinate. The Rayleigh velocity is then obtained by the Eq. (6), where Δz is the pseudo-periodicity of the signal, V_{fl} is the ultrasonic velocity in the coupling fluid, and f is the operating frequency.

2. SCANNING ACOUSTIC MICROSCOPE IN HOT CELL

2.1. Device installed at ITU

In 2000, an acoustic microscope was built and introduced into ITU's hot cell facilities as part of a collaboration with IES (Institut d'Electronique du Sud) at University of Montpellier and EDF. Fig. 2.1 shows a photograph of the device in a hot cell. Inside the cell are the sensor heads, translation stage, micrometer motors and acoustic sensors. Outside the cell are the complex electronics which consist of remote triple axis sample movement control electronics, RF amplifiers, acquisition boards, computer for data processing are located outside the hot cell (see Fig. 2.2).

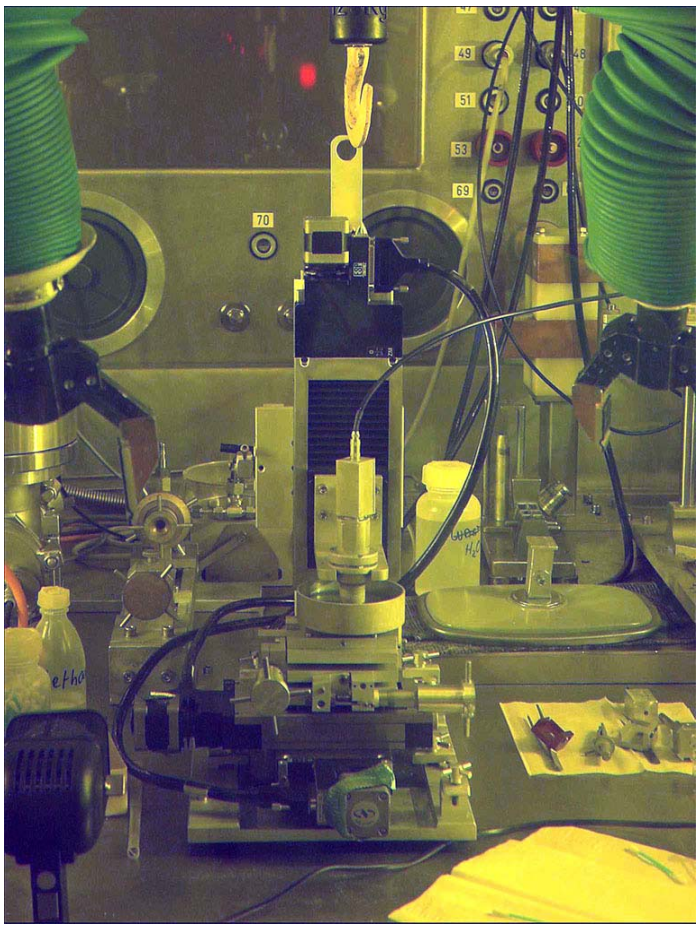

FIG. 2.1. Acoustic microscope installed in hot cell.

The acoustic sensor is connected with a coaxial cable and fixed in the lower part of the z-axis translation stage. Sensors are easily interchangeable and very rigid in order to withstand high doses of gamma radiation for an extended period of time, as normally the case in hot cells. The x-y-z translation stages are each connected with cables and are remotely controlled (see Fig. 2.2). Micrometric adjustment screws are used to level out the sample platform to ensure an equal distance between sensor and sample in the plane of the scanned area during imaging. The focus distance is based on the direct signal from the sensor which gives a feedback to the z translation stage. The size of the sample platform is restricted by the hot cell layout limitations. In this case a 90 mm large coupling liquid holder is used which gives an effective sample imaging area of approximately 50 × 50 mm which is enough to select an appropriate spot for measuring acoustic velocities. Due to the fact that each sensor gives a different resolution one can select a sensor suitable for the application, as illustrated in Fig. 2.3. In this case 140 MHz was found to give satisfactory results for UO_2 irradiated fuel samples.

FIG. 2.2. Overview of the device showing acoustic sensors, coupling liquid holder, sample platform, translation stages.

For each operating frequency a separate sensor head needs to be used (see Fig. 2.3).

Frequency	Resolution
1 GHz	1 µm
100 MHz	10 µm
10 MHz	100 µm
1 Mhz	1 mm

FIG. 2.3. On the left, acoustics sensors (from left to right 10 MHz, 80 MHz and 100 MHz). On the right, frequency/resolution correlation.

Samples with a thickness of about 1 mm are placed in an aluminium sample holder which can hold up to 3 different samples at a time, as shown in Fig. 2.4. After embedding with resin the sample holder is polished to obtain a smooth surface area ready for measurement. Fig. 2.5 shows a SEM image of three embedded samples illustrating that this sample preparation can be used for both SEM and acoustic microscopy.

The methanol coupling liquid is poured in the basket so the sample holder is completely submerged. After horizontal alignment with two adjusting screws the acoustic sensor is lowered in such a way that no air bubble is trapped which could lead to false readings. The sensor is then further lowered until a few µm distance from the sample and defocusing is started in order to get an acoustic image. The signal from the sensor is converted to an optical signal to be displayed on a computer screen. Fig. 2.6a/b shows acoustic and optical images of a UO_2 sample. Micro-cracks can be clearly seen in the subsurface acoustic picture making it helpful to select a suitable area for quantitative measurement.

FIG. 2.4. Sample holder containing 3 samples.

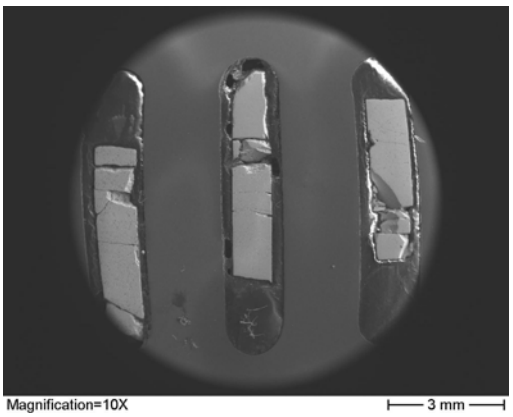

FIG. 2.5. SEM image of 3 samples ready for measurement.

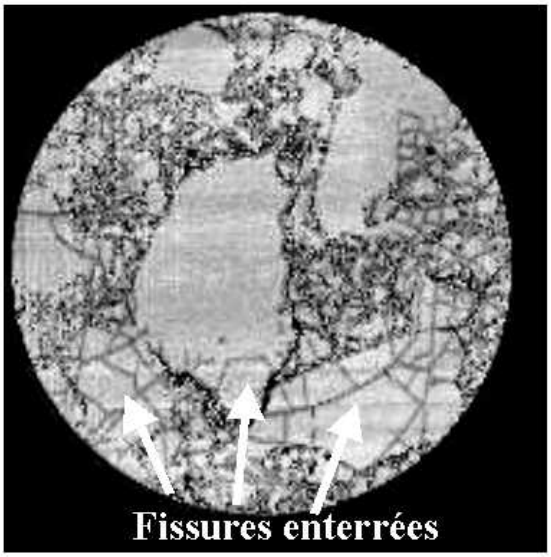

FIG. 2.6a. Acoustical image of a UO_2 high burn-up sample showing sub-surface micro cracks.

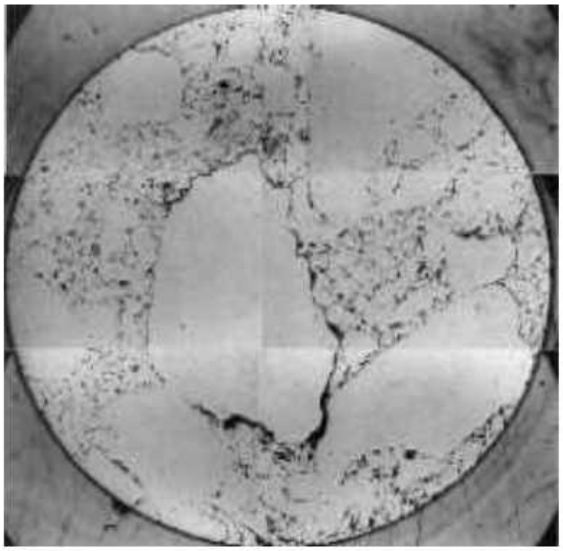

FIG. 2.6b. Optical image of the same sample as in Fig. 2.6a.

3. RESULTS AND CONCLUSIONS

The acoustic microscope was successfully tested at ITU on samples from the High Burnup Rim Project (HBRP). The experimental results were found to be in agreement with those obtained with a microindenter, also deployed at ITU. Since 2000, the apparatus has been preserved in one of the hot cells. In 2010, with the support of EDF it was decided to install a new acquisition hardware at ITU with a view to integrating acoustic microscopy as a standard characterization technique for spent fuel. In addition to the device in the hot cell, a "cold" acoustic microscope is provided to allow easy calibration of the acquisition electronics. By analyzing the available hot cell samples, it was learned how even in the case of well polished specimens some particles could come under the acoustic lens and render the acoustic signature unusable. A specific procedure was developed to eliminate this nuisance. The consistency obtained when measuring at high frequency, obtaining good quality signals indicates that the scanning acoustic microscope is a valid alternative for the evaluation of local elastic properties of irradiated nuclear material. The measurements are local enough to be carried out on pellet fragments. The choice of the acoustic frequency depends on the heterogeneity scale of the

material investigated. It is a non-invasive method for material characterisation, which has shown very good accuracy and reproducibility.

The set of results obtained so far using this technique includes simulated fuel samples manufactured at ITU, two sets of irradiated UO_2 and UO_2+ 5% Gd samples from the High Burn-up Rim Project with irradiation temperatures ranging from 500 to 1200 °C and burn-up ranging from 35–100 $GW{\cdot}d{\cdot}t^{-1}M$, and a N118 BR3 irradiated fuel sample with an average pellet burn up of 68 $GW{\cdot}d{\cdot}t^{-1}M$. For all measurements Young's modulus has been deduced from the V_R value. Despite some scatter in the results, Young's modulus after porosity correction was:

- almost constant between 0–30 $GW{\cdot}d{\cdot}t^{-1}M$
- decreasing by about 25% between 30–80 $GW{\cdot}d{\cdot}t^{-1}M$
- stabilized above 80 $GW{\cdot}d{\cdot}t^{-1}M$ and consistent with SIMFUEL analogues
- similar observations were made for $(U,Gd)O_2$ fuels

the whole body of data showed a general decrease of the elastic modulus with burn-up (pure UO_2 taken as reference), particularly in the burn-up range 0-50 $GW{\cdot}d{\cdot}t^{-1}M$ where in irradiated fuels still no high burn-up transformation has occurred, with an apparent stabilisation above 100 $GW{\cdot}d{\cdot}t^{-1}M$ (to be confirmed by future measurements). New sets of fuel samples will be identified for the next measurement campaigns; ongoing and new projects will ensure the need for measurements in the coming years.

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