

IES skills for ultrasonic innovative instrumentation in hot labs

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Abstract

Specialized in non destructive characterisation by ultrasounds, we developed specific devices for operation in nuclear environment for EDF and CEA. For this purpose, the piezoelectric elements constituting the active part of our systems have been characterized for Gamma ray and neutron irradiation (BR1 SCH-CEN reactor. For their operations in hot labs, the specific electric problems induced by long distance transmission lines and nuclearized connectors have been solved. Thus, we have succeeded in introducing recently an Acoustic Microscope as an easy handling measurement system of the mechanical properties of fuel pellets in the hot lab of TUI (Germany). This implantation is the result of a collaboration with EDF to assess the Young's modulus variation for nuclear materials submitted to high burn up rate (NFIR). Secondly, for investigation of fission gas release in very harsh environment, we have designed a new sensor to measure the internal pressure and the composition of Helium-Xenon in a specifically instrumented fuel rod in pile or above the plenum of fuel rods for hot labs. This non destructive device is a safe and real-time measurement system. It is planned to operate in hot labs (CEA Cadarache) and first validation experiments have shown good agreement versus puncture validation measurement. Its design and performances have been successfully validated in November 2012 at the Osiris experimental reactor (CEA Saclay) providing the first kinetics of the fission gaz mixture evolution for an instrumented fuel rod during a real irradiation cycle. In conclusion, our skills for ultrasonic propagation, mechanical coupling, impedance matching and Electronics enable us to assess numerous applications for research activity in hot labs.

Introduction

Ultrasonic waves are widely used to provide non destructive evaluation means for Industry. At the University of Montpellier, two specific applications are developed: the local characterisation of surface elastic properties by high frequency Acoustic microscopy and the thermo physic characterisation of gas in sealed cavities to measure for instance the fission products release inside fuel rods plenum.

Studies of surface acoustic wave propagation may give access to the elastic constants on very small areas using the acoustic signature method. Indeed, by using high frequency focused sensors around 500 MHz, the investigated volume may be limited to a diameter of 100 μm and a thickness of few microns. As these two dimensions are inversely dependent on frequency, this last parameter is chosen to fit with the sample size. For example, this local characterisation tool has been successfully applied for research on thin ion implanted layers for nuclear glasses used to trap high activity wastes for very long term storage. As acoustic sensors have a good resistance to radiation, we have designed a specific acoustic microscope for its introduction and operation inside hot cells. With this specific research device, the effects of long activity inside civil reactors on the fuel pellets mechanical properties have been investigated for numerous fuel rod samples with different high burn-up levels (NFIR program). In specific conditions, high frequency ultrasonic waves can be also used to enable gas characterization within small cavities. Despite the very high echo attenuation induced by acoustic impedance mismatch between solid and gas, we have succeeded in the development of acoustic sensors adapted to measure the inner pressure and the He/Xe mixture ratio of fission products contained in the plenum volume of fuel rods in a non destructive way. Used

in hot cells, these sensors have been qualified in hot cells from comparison with chromatography measurement after the tube puncture.

I. Acoustic microscopy for mechanical characterisation of irradiated materials

Acoustic Microscopy is a powerful non destructive tool to reveal the material microstructure from the detection of the reflection conditions of a focused ultrasonic beam. With high frequency waves, the spatial resolution of acoustic images can go down to just few microns. Simultaneously, variations of elastic properties can be locally investigated in solid materials from surface wave velocimetry or acoustic signature technique.

I.1 Application to nuclear glasses

Acoustic signature has been successfully used to study the alteration mechanisms of glasses selected for confinement of high-level long-lived nuclear waste. Indeed, to realize accelerated tests, these effects are investigated on Au ion implanted materials. In this way, defaults equivalent to real irradiation are generated in the glass matrix but the thickness of the altered layer is nevertheless limited to few microns. So, very high frequency ultrasonic sensors have to be used to generate surface waves with a small enough penetration depth compared to the altered layer thickness. Thus, by using Rayleigh waves present in bulk materials, characterisations of the mechanical properties on a limited depth of few microns under the sample surface have been obtained with a focused sensor operating at 600 MHz. With this acoustic microscope, the effect of this ion implantation on the surface mechanical properties has been detected in a non destructive way [1]. This technique has revealed the decreasing of the Young's modulus of the glass surface layer at the beginning of the ion implantation and next the saturation of this effect.

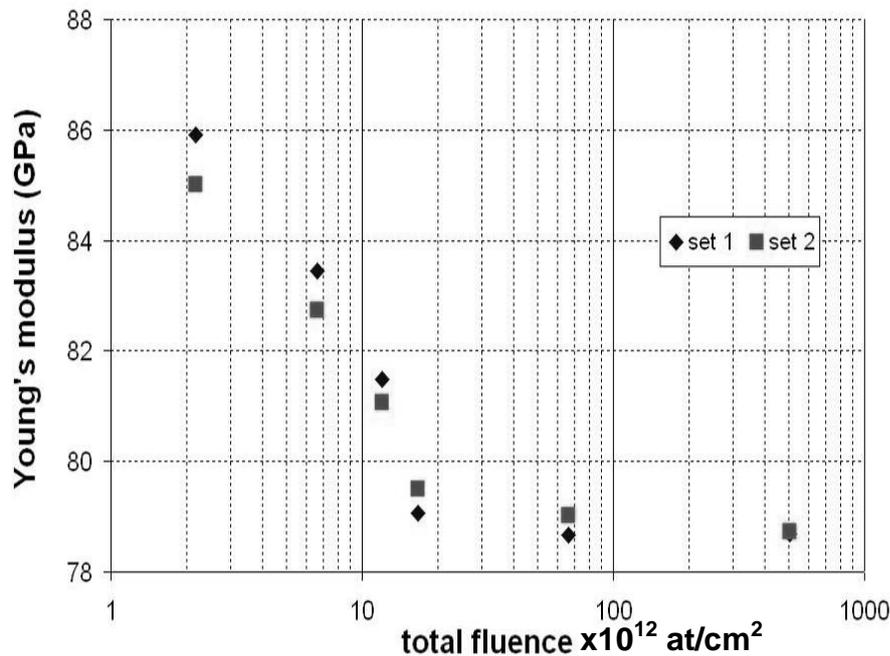


Figure 1 : Characterisation of the alteration of the surface of a nuclear glass sample exposed to ion implantation to simulate effect of radionuclids for long term storage

I.2 Application to irradiated fuel pellets in hot cells

As the sensors used for acoustic microscopy can be done with piezoelectric materials such as LiNb crystals or PZT, they are not strongly affected by the radiations levels reached in hot cells. For instance, we have shown experimentally that they can pass Gamma rays exposure larger than 1 MGy (IRMA irradiator) and neutron irradiation up to 10^{17} n/cm² (BR1 reactor) without any breakdown [2]. Even under this neutron irradiation, a limited shift of their electrical resonance response has been detected but this effect has been followed by a natural recovery process. For this reason, these ultrasonic sensors seem to have a large enough lifetimes for long term operation inside hot cells.



Figure 2 : nuclear setup for acoustic microscopy in a hot cell

To characterize the mechanical properties by acoustic signature or to reveal the microstructure by acoustic imaging inside hot cells, acoustic microscopes have to specifically designed to introduce and to fix, with robot arms, the three axis motorized system used to scan the surface samples and to adjust the focus position. Next, for operation inside hot cells, specific handling systems adapted for remote handling are used to change the sensors, to manipulate the samples or for tilt surface compensation. Two acoustic microscopes have been recently installed at ITU for mechanical characterisation outside and inside hot cells.

Previously, for a collaboration with EDF, acoustic imaging has been a very valuable method to investigate the properties of irradiated samples made of spent fuel pellets issued from different civil reactors. The aim of this study was to investigate the effect of high burn-up levels on such highly irradiating materials [3]. From acoustic images obtained in hot cells, the different porosity levels of the fuel microstructure have been revealed (cf. figure 3). This investigation technique has been particularly helpful to locate cracks especially when they were closed ones or partially filled after cutting or polishing operations. Indeed, underground cracks are inappropriate for Knoop indentation characterization.

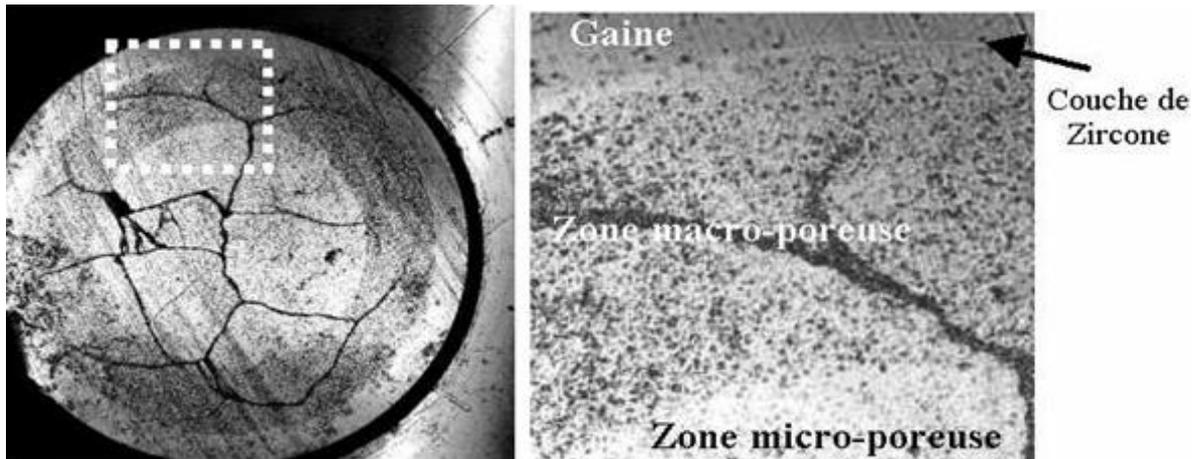


Figure 3 : acoustic images of the cross section of spent fuel rod with a burn-up level of 68 GWj/tU

This crack localization was useful also to locate homogeneous and large enough area to perform accurate acoustic signatures to quantify the fuel mechanical properties from measurements of the Rayleigh wave propagation velocity and attenuation.

These investigations have been done with spent fuels and with SIMFUEL samples i.e. non irradiated fuels with additives to simulate the role of different fission products.

From these acoustic signature measurements on fuel, the gradual decrease of its Young's modulus has been quantified on numerous samples (cf. figure 4)

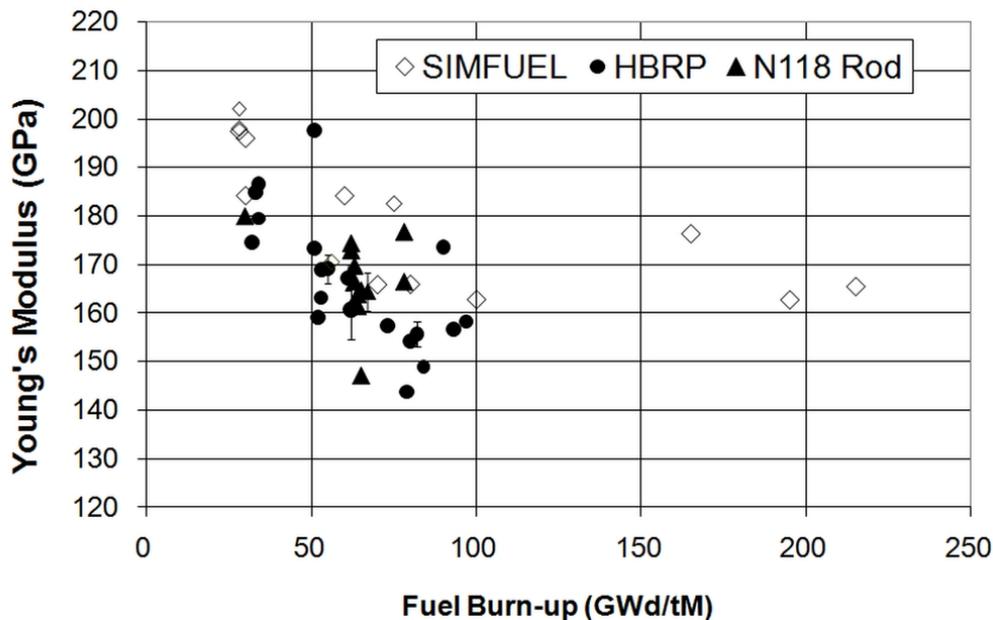
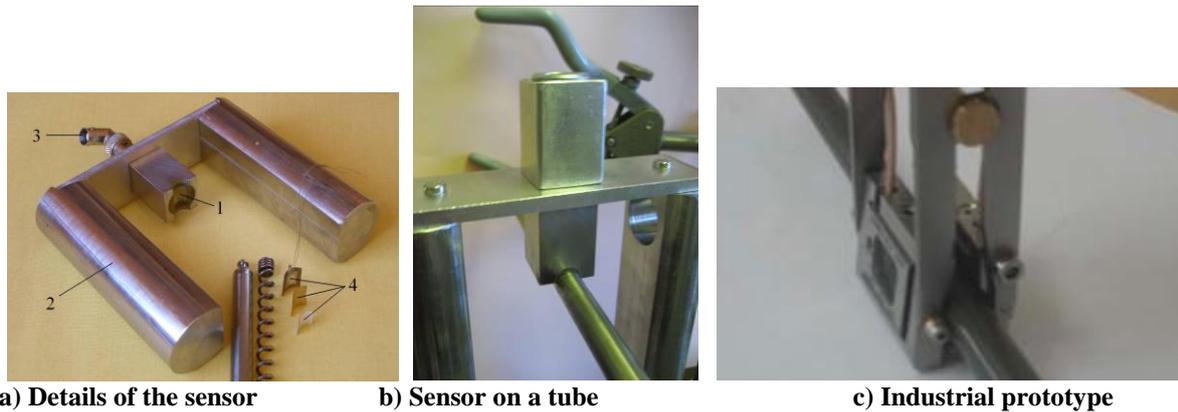


Figure 4 : detection of the Young's modulus decrease for fuel burn-up increase for spent fuel rods (HBRP and N118) and equivalent non irradiated samples (Simfuel)

II. Fission gas analysis on fuel rods by acoustic resonators for hot cells

To control the inner pressure and the gas mixture composition inside fuel rods in a totally non destructive way, we have developed a new method based on the propagation of ultrasonic waves inside the rod plenum [4, 5] by placing a specific sensor directly upon the tube. The design of this device has been modified for remote handling by robot arms for operation inside hot cells (cf. figure 5). This measurement technique has been also optimized to deal with the presence of the metallic spring present inside the fuel rod plenum.



a) Details of the sensor b) Sensor on a tube c) Industrial prototype
Figure 5 : acoustic sensor for inner pressure and gas composition evaluation on cylindrical cavities such as fuel rod plenums

From optimized coupling conditions, we managed with this device to inject enough acoustic wave energy through the tube wall despite the very high reflective conditions between solids and gas. With the right configuration, the gas filled metallic cavity acts as a kind of acoustic resonator. Consequently, numerous round trip echoes are available to get the value of the velocity of ultrasonic waves in the gas inside the cavity with a very good accuracy by using sophisticated signal processing. This velocity is characteristic for any gas and depends on its purity. Wave damping is mainly inversely proportional to the gas pressure.

In the case of real fuel rods, the plenum cavity is initially pressurised with Helium. Due to irradiation, fission gas are released, mainly Xenon. So, using thermo physics equations, the proportion of Helium versus Xenon in the rod plenum can be deduced from the acoustic velocity measurement.

This sensor has been tested on several fuel rods in the hot cells of the Leica-star facility (CEA Cadarache). Acoustic measurements were made and next, these samples were carefully punctured to measure their inner pressure and to analyse the fission gas composition by chromatography. Acoustic measurements were found in good agreement with chromatography results with a maximum discrepancy of only 4% and a resolution better than $\pm 0.3\%$. For pressure, in the range between 40 and 60 bars, acoustic measurements are not always in perfect agreement with the destructive measure. Some disturbances could be attributed for example to dispersion of the tube dimensions or to their surface states due to corrosion, etc. At the moment, the pressure incertitude by acoustic measurement is closed to ± 5 bars compared to ± 2 bars by chromatography. Further works are planned to improve this. Nevertheless, this non destructive method seems to be sufficiently accurate for non destructive control applications for fuel rods in hot cells or even in storage pool with further developments.

This sensor technology can resist to hot cell environment and also to much higher radiation rate and temperature. In collaboration with CEA, a specific cavity and its acoustic sensor has been added to an instrumented fuel rod to measure the fission gas mixture composition in experimental reactor during irradiation campaigns. Recently, this experimental device has given the first insight of the kinetics of the gas fission release in pile at the Osiris reactor (CEA Saclay) for a temperature up to 200°C and a total epithermal neutron dose around 10^{17} n/cm^2 .

Conclusion

Acoustic sensors can efficiently resist to radiation levels reached in hot cells. They are also sufficiently robust and easy to handle for operation with robot arms. We have demonstrated that they can operate in imaging and acoustic signature modes to reveal the microstructure modifications and to quantify the variations of the elastic constants of fuel

pellets from cross sections of fuel rods exposed to high burn-up levels. This complete acoustic set-up is now available for investigations of solid materials at the ITU research centre. They can detect very thin modifications of elastic properties such as ion implantation effect in nuclear glasses or control in depth interface such as braze interface for instance. Further developments are planned to image the pellet-cladding interface all along fuel rods in hot cells.

With a specific system, fission gas analysis can be also performed in a totally non destructive way in hot cells by placing our acoustic sensor upon the plenum cavity of fuel rods. This direct and almost instantaneous measure gives access to the composition ratio of He/Xe with incertitude of only $\pm 0.3\%$ in good agreement with qualification tests obtained by chromatography after the tube puncture. The inner pressure can be simultaneously estimated. This could be useful for safety controls of irradiated fuel rods. The same technology has been implanted in a specific instrumented fuel rod and is actually tested in pile at the experimental Osiris reactor (CEA Saclay).

References

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