PRESSURE AND COMPOSITION OF GAS MIXTURES IN FUEL RODS FOR PRESSURISED WATER REACTORS BY AN ULTRASONIC SENSOR.

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Abstract

Non destructive evaluation is widely used in the nuclear power plants in order to insure security but also to follow the evolution of numerous parameters. We propose a non destructive method based on acoustics and more precisely on the design of a new sensor in order to determine the composition of the Helium-Xenon mixture in the free volumes of a fuel rod by measuring the time of flight. Thus we present an original application of acoustics in the non destructive testing nuclear industry.

Introduction

Even though in most countries nuclear energy is not the major way of producing electricity, in France on the contrary over 80% of its energy is produced that way [1]. This is why researches in collaboration with EDF have been carried out in order to study and optimise the UO_2 fuel pellets combustion in fuel rod.
The majority of the studies and the developments in France in the nuclear energy field during the past twenty years have focused on pressurised water reactors (PWR). Fuel pellets made of Uranium Dioxide (UO_2) are stacked up into a Zircaloy pipe called the fuel rod. The clad is closed under a Helium atmosphere at a pressure equal or superior to 25.105 Pa before the reaction starts, which partly counterbalances the effect of the external coolant pressure (water at 155.105 Pa). During the nuclear reaction in pressurised water reactors, solid and gaseous fission products are created. However the gases and solids produced do change state from gas to solid and from solid to gas along the irradiation process [2]. The gases, mainly Xenon and Krypton, are released in the free space of the fuel rod. Moreover, the UO_2 pellets are under various stresses during the reaction, they change physical-chemical properties and therefore put a strain on the Zircaloy cladding. A layer of zirconium oxide of several microns also appears on the inside and outside of the pipe as the reaction goes on. All of these phenomena put the pipe under stress, corrode it and may lead to the cladding rupture. For obvious security reasons but also for economical interest, the quantity of released gases in the cladding has to be evaluated.

So far in the study of PWRs, various destructive and non-destructive means have been used to measure the pressure inside the claddings. Gamma spectrometry for instance allows one to estimate the average burn-up of the fuel element, which can be related to the quantity of released gases in the cladding. However rod-puncturing [3] leads to the knowledge of the pipe inner pressure, the quantity of released gases and the free volume of the pipe in a definite way.

We propose a method based on acoustics to determine, via the speed of ultrasounds, the composition of a gas mixture from the outside of the pipe and thus propose an answer to this non-destructive evaluation application. We will describe the experimental set-up used to achieve those measurements. We will recall the principle of the specific sensor we designed in order to transfer energy through the pipe wall, which is the first step before performing time of flight measurements.

Experimental set-up and method

Some authors have already described a technique using echography to measure ultrasonic velocity in gases in a vessel [4]. But this method requires the introduction of sensors inside the pipe, which is not possible in the case of nuclear fuel rods. An experimental methodology needs then to be defined. The main problem, before measuring precisely the time of flight of an acoustic wave in a medium, is to make the wave propagate in the medium.

The gas is contained in a 1 cm Zircaloy pipe. The thickness of the rod is 570 µm. Small acoustic impedance of gas compared to solid one induces a strong insertion loss which could prevent any propagation of ultrasonic waves inside pipes. Indeed, in the case of a Zircaloy/Helium-Xenon mixture interface, the coefficient of refection (defined as below) is pretty equal to 1.

$$R = \frac{Z_{Zirc} - Z_{gaz}}{Z_{Zirc} + Z_{gaz}} \qquad (1)$$

where $Z_i = \rho_i . v_i$, Z_i is the acoustic impedance of medium i, ρi the density and v_i acoustic waves velocity.

α^T is the coefficient of transmission of the acoustic wave trough a layer of thickness *e.*

$$\alpha^T = \frac{4Z_1Z_3}{(Z_1+Z_3)^2\cos^2\frac{2\pi e}{\lambda_2}+(Z_2+\frac{Z_1Z_3}{Z_2})^2\sin^2\frac{2\pi e}{\lambda_2}} \quad (2)$$

λ_2 is the wavelength of the acoustic waves propagating in the layer.

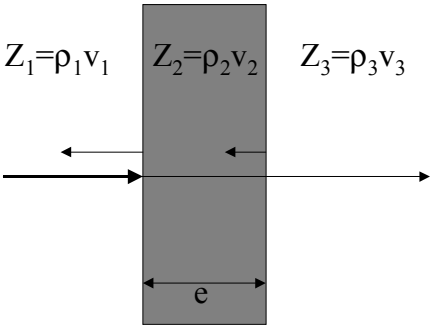

Figure 1 : impedance matching for three layers.

When $e = \lambda/2$, the layer is transparent. When $e = \lambda/4$, then $\alpha^T=1$ for $Z_2 = (Z_1Z_3)^{1/2}$. The adequate choice of material allow to get higher impedance matching.

For this reason, we proposed to adapt the piezoelectric sensor frequency to the 570µm wall thickness of these rods and to the water thickness layer (figure 2) to reduce the different acoustic impedance mismatches to ensure sufficient wave propagation inside the gas.

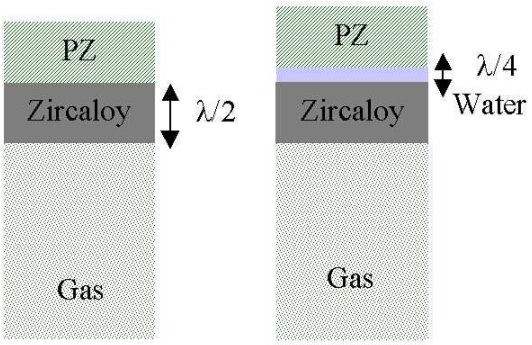

Figure 2 : adaptation of the piezoelectric sensor frequency to the 570µm wall thickness of the rods and to the water thickness layer

We chose a frequency of 4MHz in order to let the ultrasound go through the pipe wall. Indeed the pipe wall being 570µm thick, it is half wave for this frequency. The Zircaloy layer is then transparent. Then impedance matching concerns piezoelectric material and gas. A quarter wavelength water layer allow to increase the transmission : Z_{water} is near than $(Z_{gas.}\, Z_{Pz})^{1/2}$. This interferometric structure guarantees an optimal transmission of the ultrasonic energy inside pipe free volume.

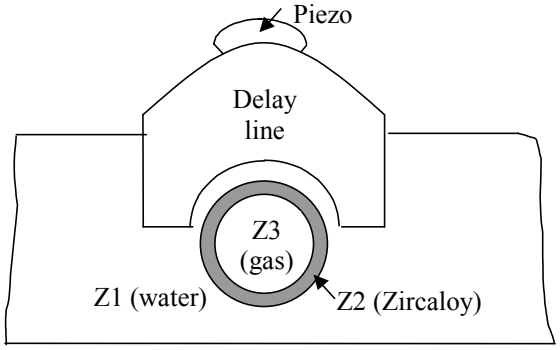

Figure 3 : Schema of the sensor on the Zircaloy pipe

With our device, we reduce the reflection coefficient at the Zircaloy/gas interface so that performing time of flight measurements is possible without the use of signal processing developments. We realised a half-cylindrical sensor for this purpose, which is described on figure 3. The pipe is immersed in water. A specific mechanical holder has been designed to fix this transducer to the external surface rod. Then we use the relative position of the echoes in order to perform time of flight measurements.

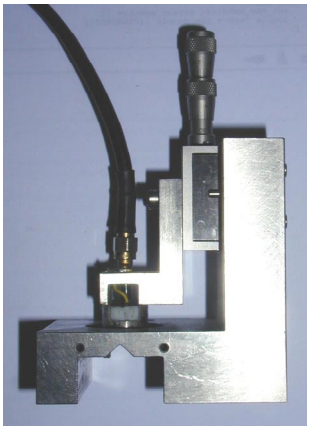

Figure 4 : A specific mechanical holder to fix the transducer to the rod.

Any complementary information that could be carried by the amplitude of the reflected echoes such as the reflection coefficient or the attenuation that are both dependent on the pressure, are not reliable enough. In an ideal case, the reflection coefficient is measurable and close to the models for normal incidence of the ultrasonic beams. But the system is resonant and there are many defects in the thickness of the layers, which lead to a significant misprecision. Indeed, the lack of knowledge in the dimensions of tubing walls (570µm +/-5%) as well as their mechanical properties evolution when under irradiation affects the accuracy of measurements taken of the echo amplitude.

Experimental results and discussion

Pressure determination

We are going to give the experimental results obtained for one gas (Nitrogen, N2) and for the Helium-Xenon gas mixture. First we show on figure 5 an echogram representative of the kind of signal we are dealing with.

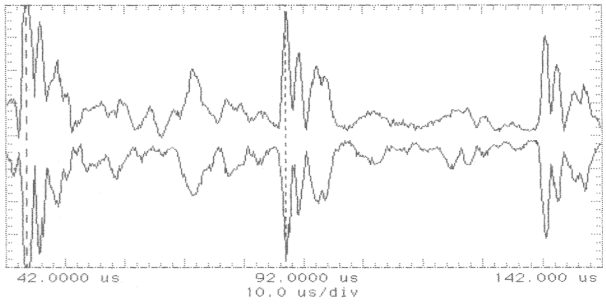

Figure 5 : Echogram to show the return journeys inside the pipe.

This figure represents the observation of 3 echoes in the tube filled with nitrogen at 130.105 Pa. The times of flight are 45.6µs, 88.8µs and 132µs.
The method lies in the measurement of the times of flight between several return journeys (up to ten) of the acoustic wave inside the gas. From this time value one can get the velocity of the ultrasonic waves from the relation:

$$v = \frac{2.d}{t} \qquad (3)$$

for one return journey where d is the inner diameter of the pipe (d=8.36mm +/- 28.5µm) and t the time of flight for one return journey across the pipe.

Therefore we can represent (figure 6) the propagation speed in gas of the ultrasonic waves versus the pressure for one gas such as nitrogen.

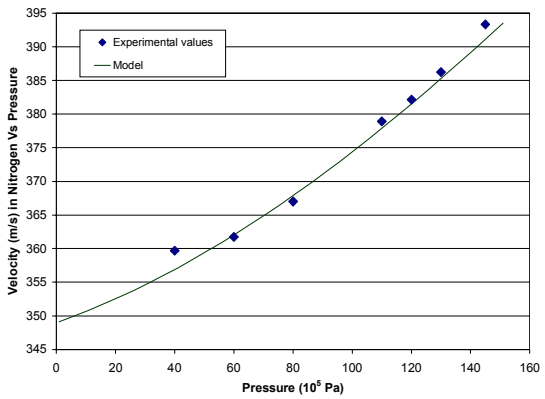

Figure 6 : Theoretical and experimental velocities for Nitrogen, T=300K.

Using a thermodynamic model and virial expansion which is not detailled in this paper, a velocity law versus pressure has been obtained and validated experimentally from 40 to 140 bar with our sensor for a fuel rod filled with nitrogen gas. For this pressure range, velocity deduced from flight time measurement varies from 360 to 390m/s.

Composition determination

The range of pressure and temperature for the Zircaloy tubing which interest us is 60-90.105 Pa and T=298-450K, with a proportion of Xenon up to 5%.
In these conditions the sound velocity variations versus the composition of the mixture are obtained for different pressures .

The composition can be deduced from velocity measurements with the relations (4) and (5) .

$$v = \sqrt{\frac{\gamma.R.T}{M}} \qquad (4)$$

$$M = x_{He}.M_{He} + x_{Xe}.M_{Xe} \qquad (5)$$

where γ is the ratio of the specific heat of the gas at constant pressure c_p to the specific heat at constant volume c_v. R is perfect gas constant. T the temperature. M is the molecular weight of the gas and x_i the volumic fraction of the gas i.

A virial expansion allows to take into account pressure at second order.

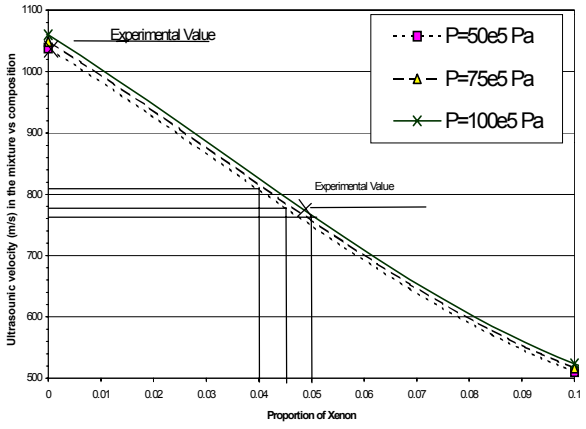

Figure 7 : Ultrasounds velocity in the Helium-Xenon mixture at various pressures versus the proportion of Xenon at 4MHz, T=300K.

We can see on figure 7 that in the case of fuel rods initially filled with pure Helium gas, 10% molar fraction of Xenon decreases velocity from 1050 to 520m/s whereas pressure variation from 50 to 100 bar induces only a 20 m/s increase. These predictions

have been found in good agreement with measurement on Helium-Argon equivalent gas mixtures.

For a pressure between 50 bar and 100 bar and for a Xenon composition between 0 and 10%, determining the ultrasound velocity obtained with our method gives the composition of the gas mixture with a margin of error around 10%.

With an estimated error margin of 4-5% on the velocity, the composition of gas can be ascertained to a 99% degree of accuracy. Thus using our experimental set up, we can determine the composition of a gas mixture for this pressure range with a 90% degree of accuracy (10% error margin).

So, velocity is mostly sensitive to the gas mixture proportion rather than to pressure.

It is interesting to note that if the composition is known through the experimental knowledge acquired during the past 35 years, an accurate enough velocity measurement could lead to the pressure. As for one gas, the speed of ultrasound measurements will directly lead to the determination of the pressure via a specific thermodynamic models which is not detailed here. We did not take into account the Krypton since it represents only 1.1% of the total weigh of the fission products (in comparison, Xenon represents 12.7%). Rapid evaluation showed it would not affect the measured ultrasound velocity.

Conclusion

In the nuclear industry, non-invasive measuring techniques are widely used, especially to control the reaction advancement for which the knowledge of the pressure and the composition of the gases involved is needed. The pressure inside the rods is related to the combustion rate and its optimisation allows longer fuel rod lifetime. This study is part of a feasibility project financially funded by EDF whose people could think of modifying some of their tubes in order to be able to install there our system which could allow to follow on the site the evolution of the Xenon concentration.

We developed a specific half-cylindrical sensor in order to perform the time of flight measurements. We proposed to adapt the piezoelectric sensor frequency to the 570µm wall thickness of these rods and to the water thickness layer to reduce the different acoustic impedance mismatches to ensure sufficient wave propagation inside the gas. This interferometric structure guarantees an optimal transmission of the ultrasonic energy inside pipe free volume. We showed that the measurements were accurate enough to reach, via the ultrasound velocity, the pressure of a single gas or the composition of a gas mixture such as the Helium-Xenon one found in the Zircaloy tubing of PWR reactors of nuclear power stations.

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