Acoustics'08 Paris
June 29-July 4, 2008
www.acoustics08-paris.org

Effect of intense neutron dose radiation on piezoceramics

Franck Augereau[a], Jean-Yves Ferrandis[b], Jean-François Villard[c], Damien Fourmentel[c], Mark Dierckx[d] and Jan Wagemans[d]

[a]IES / Université Montpellier II, Université Montpellier II, Place Eugène Bataillon, 34095 Montpellier, France
[b]Institut d'Electronique du Sud UMR-CNRS 5214, Université Montpellier 2 CC082, Place Eugène Bataillon, 34095 Montpellier, France
[c]CEA Saclay, 91191 Gif sur Yvette Cedex, France
[d]SCK-CEN, Boeretang 200, B-2400 Mol, Belgium
franck.augereau@ies.univ-montp2.fr

Four grades of commercial PZT materials have been exposed to nuclear radiation during five months in an irradiation channel of the BR1 research reactor at SCK•CEN (Belgium). This experimental study was performed in the framework of the Joint Instrumentation Laboratory with the CEA French Commission of Atomic Energy to validate these materials for future applications in severe conditions such as online measurements in irradiation experiments performed in research reactors. For this purpose, thin piezoelectric discs were irradiated while a remote network analyser continuously monitored the frequency response of their electrical impedance. The total neutron dose has reached a level of $1.5.10^{17}$ neutrons/cm^2. Positive and negative shifts of the peak resonance frequency have been recorded but in any case with a variation lower than 1%. On the other hand, the amplitude of the electrical impedance at resonance frequency has largely decreased with even a reduction by factor two or three for some piezoelectric cells. Transitory effects have also been detected for these two parameters as function of the reactor activity. Additional thermal and gamma radiation effect have been investigated. Similarly, some piezoelectric cells glued on glass delay line have been tested with satisfactory results to these stresses.

1 Introduction

This experimental investigation takes part into an attempt to create a non destructive tool to measure the internal pressure rising due to gas fission release inside fuel nuclear rods. For this purpose, the active part of this sensor has to be qualified for gamma and neutron radiations. Piezoceramics provide an efficient means to generate and detect ultrasonic waves in the frequency range suitable for our application. For this reason, various products such as PZT27 and 46 were exposed to an high activity Cobalt source for few days. Next, these devices were placed inside a research nuclear reactor to investigate their reliability towards neutron activity. The corresponding damages or modifications of the devices under test were detected from the frequency measurement of their electrical impedance. These variations were compared to pure thermal tests to separate this effect to the radiation one.

2 Test for gamma irradiation effects

These tests were performed at the IRSID irradiation facility in France. This experimental campaign was a preliminary investigation of the transitory and long term effects of gamma radiation for piezoelectric cells used for non destructive testing in radioactive environment such as at vicinity of nuclear core. For these tests, different sets of four piezoelectric cells were fixed on specific handlers and moved by robot arms inside the irradiation chamber to be exposed momentary to 10kGy/h, 1kGy/h, 0.1kGy/h or 0.01kGy/h flence rate. Depending on the different sensors sets, the overall cumulated fluence ranged from 50 kGy up to 2 MGy. Four kinds of piezoceramics delivered by Ferroperm ™ company have been tested: PZT 26, 27, 34 and 47. Some of these devices were glued to a backing element and/or glued to a glass delay line. As resonant devices, their electrical properties were characterized by the frequency and amplitude of the maximum of the real part of the electrical impedance respectively noted F_{max} and A_{max}. The two electrodes of each sensor were soldered to a coaxial wire and linked to a RF multiplexer. Sequentially, a network analyser measured the electrical impedance on a given frequency range for each channel. Eighteen channels were available and the scanning repetition rate was around one minute. Each output of the multiplexer was initially calibrated with specific loads. For one channel, the piezoelectric device under test was replaced by a 50Ω load to investigate any possible impedance drift induced by the gamma exposition of the coaxial wires. For this campaign, this variation has not exceeded 1% for the 2-5 MHz investigated range.

None of the piezoelectric assemblies has lost irremediably its property and in fact, most of the variations were extremely small especially for F_{max} with variations never higher than 2%. For instance, this effect corresponds for a two 2 mm thick PZ27 piezoelectric cell in free mode (no backing or delay line) to a gradual increase for F_{max} up to 1.1% for a 200 kGy total dose (cf. figure 1). Moreover, for the last part of this curve (4500 to 5000 min time interval), the Cobalt source has been removed and a recovery process is visible with a 20 min constant time but the resonance frequency does not reach its initial value even 3 days after.

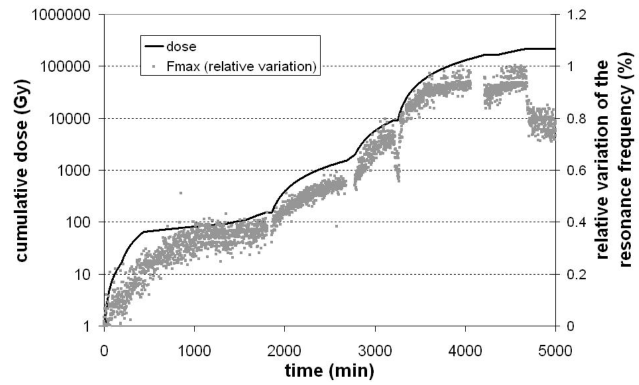

Fig. 1 Effect of gamma radiation for the resonance frequency of a 2mm thick PZ27 cell

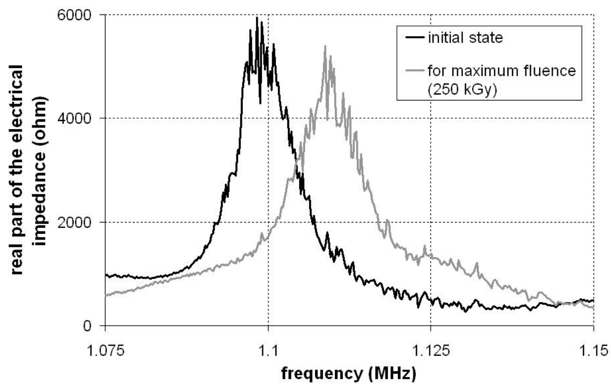

Fig. 2 Effect of gamma radiation on the electrical impedance of a 2mm thick PZT27 cell in free mode

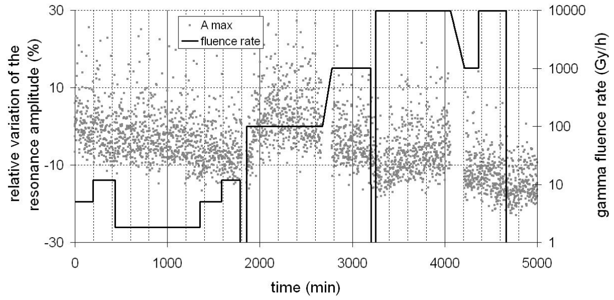

Fig. 3 Effect of gamma radiation for the resonance amplitude of a 2mm thick PZ27 cell

Similarly, the resonance amplitude decreased gradually with some transitory effects linked to manual fluence rate modifications in the irradiation chamber.

These effects of gamma radiation for the PZT27 material seem to be reduced for smaller cell thickness. Indeed, for a 0.6mm thick PZT27 cell, the maximum variation of the resonance frequency did not exceed 0,4 % even for 1,5MGy total dose (cf. figure 3). A recovery is also visible after gamma irradiation ending but with a longer constant time.

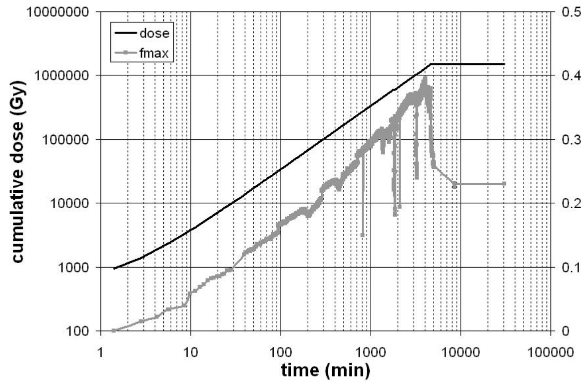

Fig. 4 Effect of gamma radiation for the resonance frequency F_{max} of a 0,6mm thin PZ27 piezoelectric cell in free mode

The amplitude of the real part of the electrical impedance of this 0,6 mm thin PZT 27 cell first increased in the early stage of this test and next resonance amplitude gradually decreased with the exposure time (cf. Fig 5). Surprisingly, this parameter recovered immediately its initial value any time the irradiation source was switched off. Moreover, a kind of memory effect occurred for any new following radiation exposition since this parameter returned to its previous minimum value.

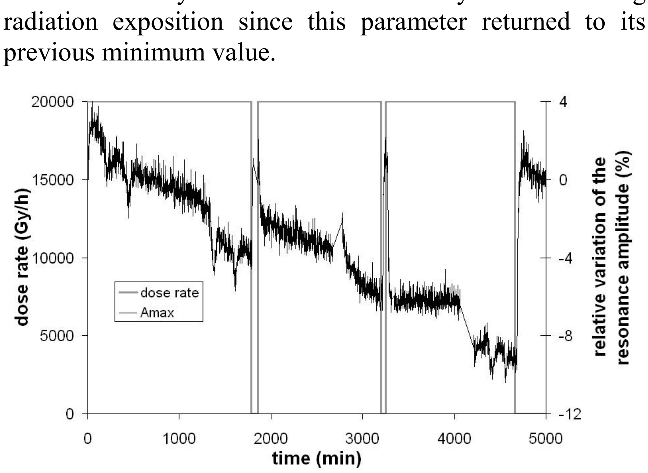

Fig. 5 Effect of gamma radiation for the resonance amplitude A_{max} of a 0,6mm thin PZ27 piezoelectric cell in free mode

Gamma irradiation is known to produce temperature rising of any material depending mainly on its mass density or effective cross section but this effect was theoretically limited to only few Celsius degrees in our case. To quantify this impact on the electrical impedance measurement, all our piezoelectric cells have been tested versus temperature before gamma radiation exposition. The figure 6 shows the direct correlation of the resonance amplitude decreasing with temperature but this variation was five times smaller than the irradiation one at maximal dose. Consequently, the variations of A_{max} on figure 5 were not mainly created by the temperature rising induced gamma irradiation.

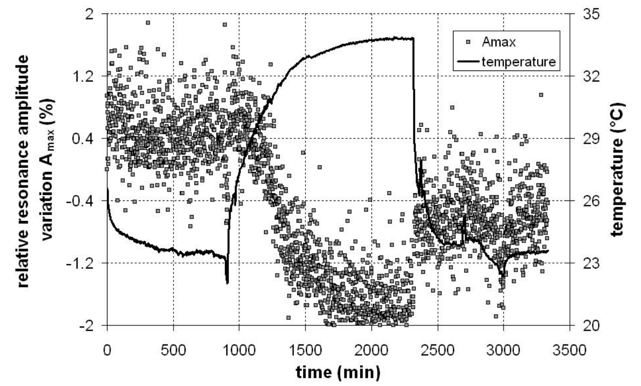

Fig. 6 Temperature dependence of the resonance amplitude A_{max} for the 0,6mm thin PZT27 piezoelectric cell

For F_{max}, the comparison of the figures 4 and 7 reveals that a small pure thermal effect exists but it is largely compensated and negligible compared to the gamma irradiation one.

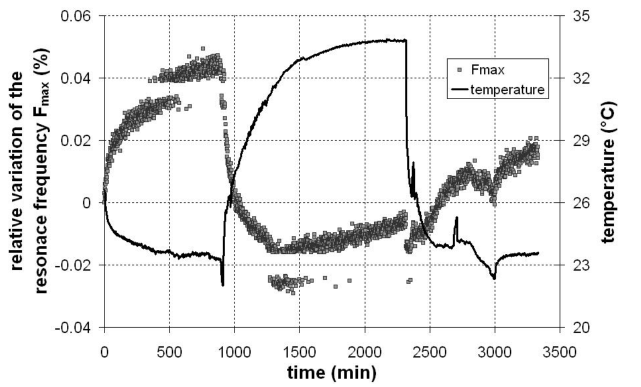

Fig. 7 Temperature dependence of the resonance frequency F_{max} for the 0,6mm thin PZT27 piezoelectric cell

Next, PZT43 piezoceramics dedicated for high temperature applications (Curie temperature above 600°C) have been also tested. Once more, the remaining effect of irradiation on the resonance is small (cf. Fig 7). Transitory effects due to sudden dose rate variation have not been observed for this material.

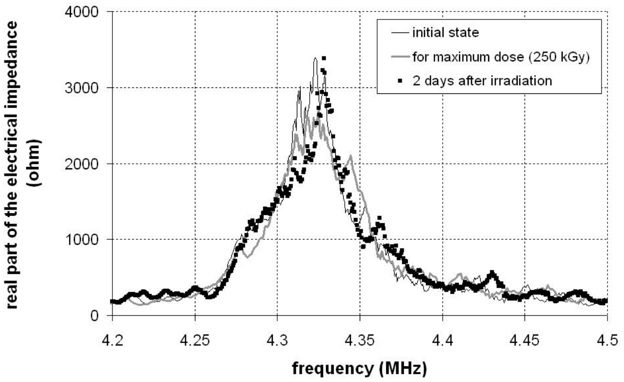

Fig. 8 Effect of gamma radiation on the electrical impedance of a PZT43 cell in free mode

Finally, even piezoelectric devices made with glued delay line or backing resin elements have not been damaged by these gamma irradiation tests.

3 Test for neutron irradiation effects

The BR-1 research reactor at SCK•CEN, Mol (Belgium) has a graphite core matrix loaded with fuel rods consisting of a natural uranium slug in an aluminium cladding. This research reactor is used as neutron source for physics experiments and as a training tool for new plant operators.. It is operational for 5 days per week at a power of 700kW for a maximum of 8 hours a day. The core is cooled by force air convection and its temperature is kept well below 200°C.

Due to this multiple purpose activity, the reactor activity was not driven specifically for our tests. The temperature inside irradiation channels was not constant. Nevertheless, it gave us the opportunity to perform a long term irradiation test for 150 days.

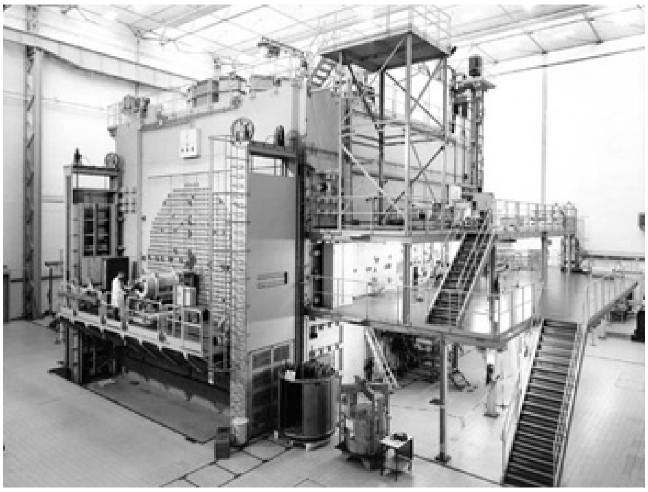

Fig. 9 View of the BR-1 reactor

For material irradiation, several irradiation channels are available to place test devices into the reactor core. For our experiment, a dozen of piezoelectric cells were linked to a multiplexed network analyser with specific coaxial wires made of mineral insulator and external metallic tube to minimize induced irradiation effect on connections. Theses devices were fixed into an aluminium box with dosimeter targets for measurement of the total fluence at the end of this experiment campaign. Inner temperature was also continuously monitored using a thermocouple classically used for irradiation test.

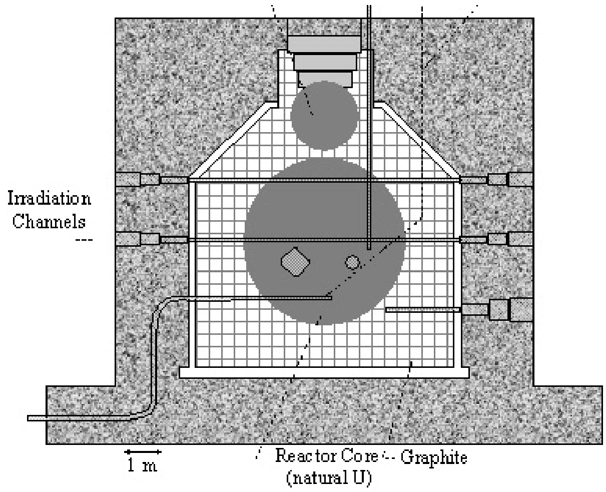

Fig. 10 Schematic structure of the reactor

	Fast neutron	epithermal	Thermal neutron	gamma
Estimation after 50 days	$5.9\ 10^{19}$ n/m^2	$3.9\ 10^{19}$ n/m^2	$6.3\ 10^{20}$ n/m^2	150 kGy
Measurement for 150 days	$1.6\ 10^{20}$ n/m^2	$1.1\ 10^{20}$ n/m^2	$1.6\ 10^{21}$ n/m^2	

Table 1: received neutron fluence

As for the previous gamma irradiation campaign, each piezoelectric device was checked sequentially by the measurement of its electrical impedance versus frequency in order to detect irradiation induced effects. Thus, the resonance frequency and resonance amplitude were monitored with the inner temperature with a repetition period of one minute during 50 days. Then, the devices under test stayed in the reactor a hundred days more. The measurement system was re-plugged for two days before the complete dismantling of this experiment. It gave us a total number of 14000 acquisitions for each channel (corresponding to "acquisition number*"on some graphs).

The complete set of data is presented on the two following figures for two piezoceramics in free mode.

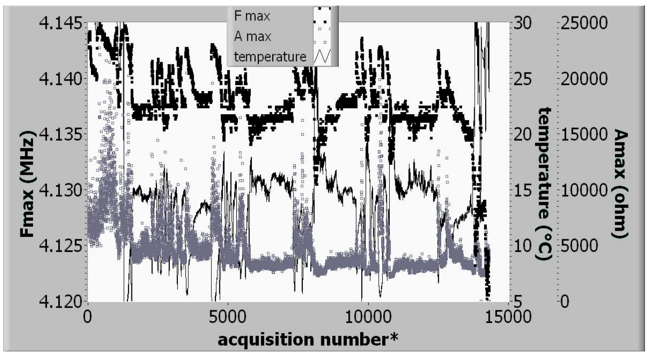

Fig. 11 Neutron irradiation test for a 0,6mm thin PZT46 piezoelectric cell in free mode

This PZT46 sample has experienced very large variations of its resonance amplitude whereas the maximal resonance frequency variation has not exceeded 0,8% (cf. Fig; 11). Nevertheless, an overall decrease of -0.5% and -50% is established respectively for F_{max} and A_{max}. Transitory effects are visible for F_{max} with a 0.2% amplitude variation.

For the PZ27 sample, transitory effects are smaller and F_{max} increases first rapidly and this variation next slows down. This increase reaches +0.4% at the end of irradiation

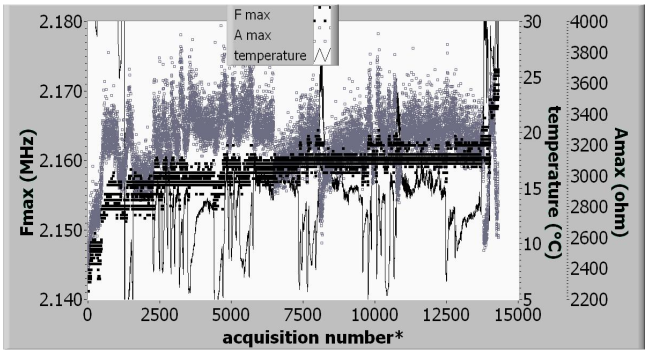

Fig. 12 Neutron irradiation test for 1mm thick PZT27 cell in free mode

Due to irradiation effect and to operation of the reactor air cooling system, the inner temperature inside the box containing the piezoelectric cells changes in a somewhat difficult way to analyse it. So, to investigate the correlation of these thermal drifts with direct irradiation effects, all the previous data have been displayed versus temperature instead of acquisition number.

For the PZT46 sample, there is a constant linear correlation of F_{max} variation versus temperature during irradiation test with a decreasing rate of roughly 0.6kHz/°C which is very much larger than the pure thermal dependence of this material simply placed into a thermostated room (cf. Fig 14). Variations of A_{max} are not monotonous with temperature during irradiation test whereas a global increasing rate of 21kHz/°C was found from pure thermal tests. Irradiation effects seem to have completely changed this material behaviour.

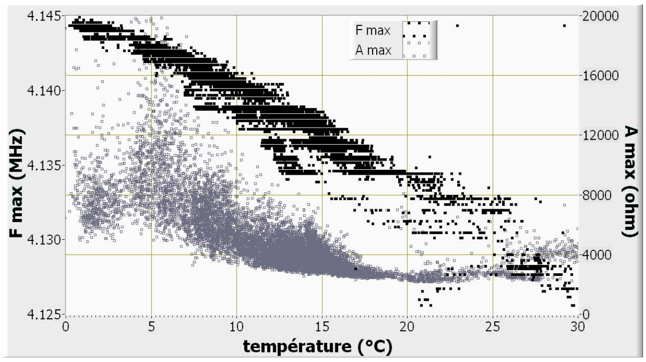

Fig. 13 Correlation of the electrical resonance parameters to temperature variation during irradiation campaign for the 0.5mm thin PZT46 piezoelectric cell in free mode

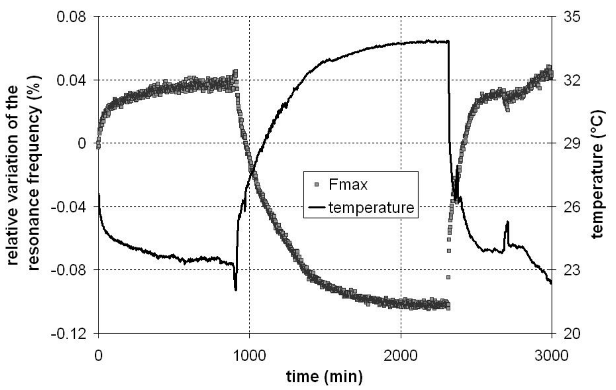

Fig. 14 Temperature dependence of the resonance frequency F_{max} for the 0,5mm thin PZT46 piezoelectric cell

For PZT27, F_{max} increased slightly with temperature during irradiation test (cf. Fig 15) whereas it decreased for pure thermal tests (cf. Fig 7). By comparison of figures 15 and 6, the thermal dependence of A_{max} is found also completely different between irradiation and pure thermal tests.

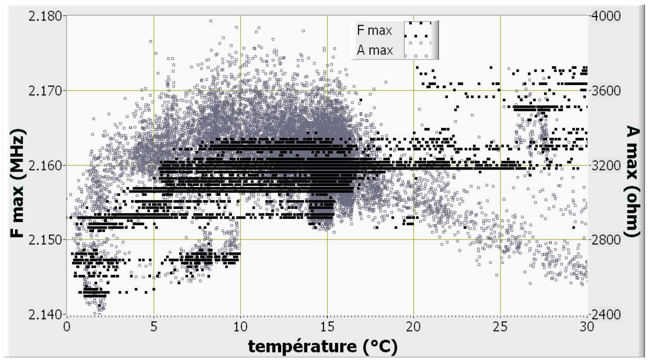

Fig. 15 Correlation of the electrical resonance parameters to temperature variation during irradiation campaign for the 1mm thick PZT27 piezoelectric cell in free mode

4 Conclusion

Electrical impedance variations are difficult to explain without more details of the piezoceramics structures. Nevertheless, this experimental work has shown the reliability of these materials to gamma irradiation up to 1.5MGy. For the resonance frequency and resonance amplitude, the variation is respectively smaller than one percent and 10 percents for the most sensitive tested piezoceramics. Nevertheless, an almost complete recovery of the initial resonance parameters rapidly happens. Gamma fluence rate variations also induced transitory effects.

For the neutron irradiation campaign, the tested piezoelectric cells cumulated a fluence of $1.6.10^{21}$n/cm^2 (for thermal neutron) for 150days of irradiation. Irreversible variations have been measured but variations of the resonance frequency remained generally smaller than 1%. The amplitude resonance was more largely reduced but the piezoelectric tested materials remained operational.