

Innovative in-pile instrumentation developments for irradiation experiments in MTRs

J-F. VILLARD

CEA SACLAY, F-91191 Gif-Sur-Yvette Cedex, France

E-Mail: jean-francois.villard@cea.fr

Abstract

In-pile measurement is a key parameter for current and future neutron irradiation experiments in Material Testing Reactors.

An ambitious program is managed by French CEA (Commissariat à l'Energie Atomique) with the aim of developing innovative in-pile instrumentation for MTRs irradiation experiments. The scope of these studies includes:

- radiation measurements, for instance neutron flux and gamma heating; these parameters are crucial to improve MTR irradiation monitoring.
- measurements of physical parameters inside the irradiation rigs, for example temperature, sample dimensions, fission gas release, etc.

An overview of this program is presented, illustrated by details about some of the recent developments, such as:

- sub-miniature fission chambers for on-line fast neutron flux measurements,
- high-temperature thermocouples for long-term in-pile experiments,
- acoustic measurements of fission gas release in PWR fuel rods.

Introduction: current in-pile instrumentation

For more than 50 years, neutron irradiation experiments have been using in-pile instrumentation to monitor and measure in-situ major physical parameters. The most typical in-pile instrumentation currently implemented in MTRs around the World is summarized in table 1.

Measurements	Examples of usual in-pile instrumentation
Fuel or material samples temperature	Thermocouples with metal sheath and mineral insulation : Below 1100°C : - Ni-Cr / Ni-Al (K type) - Ni-Cr-Si / Ni-Si (N type) Above 1100°C : - W-Re alloys (C type)
Fuel rod or material sample dimensions	Elongation sensors and diameter gauges : - LVDT based sensors - Strain gauges based sensors - Hyper frequency resonant cavities
Fission gas release in fuel rods	- Fuel rod internal pressure sensors [1] + fuel temperature measurements (centreline thermocouple) - Fission products laboratory analysis
Neutron flux	- Self Powered Neutron Detectors - Fission Chambers - Activation foil dosimeters (post-irradiation analysis)
Gamma heating	- Calorimeters - Gamma thermometers

Table 1 : Most typical in-pile instrumentation currently implemented in MTRs

This instrumentation was mainly developed more than 20 years ago, and the majority of research and development programs about in-pile instrumentation had been clearly reduced since the beginning of the Eighties.

Nevertheless some recent issues highlighted that the need for very fine and various in-pile measurements is becoming more and more acute. These new requirements for high-quality instrumentation are due on one hand to constant improvement in modeling and simulation, and on the other hand to the specific needs of emerging irradiation programs, relating for example to fuel or material characterization for HTR, VHTR, Gen IV systems, fusion, etc. As a consequence, innovative measurements are a strong necessity for upcoming in-pile experiments.

Overview of recent CEA's R&D studies for in-pile instrumentation

For some years, the CEA (Commissariat à l'Energie Atomique) is managing an ambitious program called "INSNU" to develop and qualify innovative in-pile instrumentation for MTRs irradiation experiments.

The global chart of this project is given figure 1. It summarizes all the technical items which are in the scope of these studies. This document has been established by joining all current and prospective needs pointed out not only by research reactors operators but also by CEA customers for irradiation experiments.

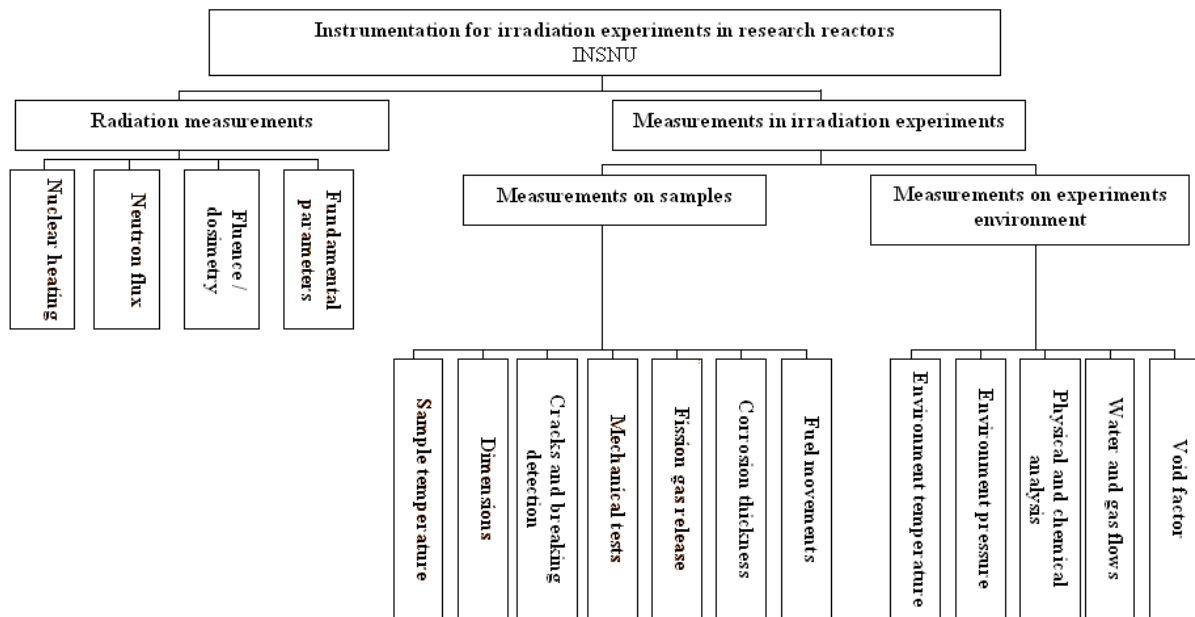


Figure 1: global chart of in-pile instrumentation needs for future irradiation experiments

The capability to operate this variety of in-pile measurements is a crucial challenge for the quality of upcoming neutron irradiation experiments in current and future MTRs.

To improve the capabilities of this project, CEA and SCK-CEN (Belgian Nuclear Research Centre) intend to team up and join efforts for research on advanced MTR instrumentation systems. A joint laboratory dedicated to in-pile instrumentation will carry on some of the future developments, particularly in the following domains:

- neutron flux measurements,
- gamma flux measurements,
- samples dimension measurements,
- fission gas release measurements.

As a matter of fact, each instrumentation developed in the INSNU project is mainly intended for existing Material Testing Reactors located in CEA or SCK-CEN Research Centers, such as OSIRIS in Saclay (France) or BR2 in Mol (Belgium), but they are also designed to be implemented in projected neutron irradiation experiments of the future Jules Horowitz Reactor, which is planned to be in operation in Cadarache (France) by 2014.

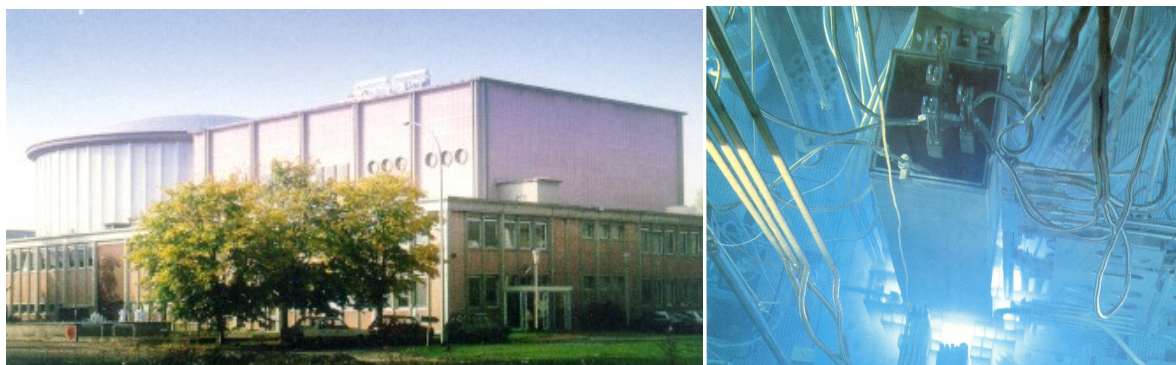


Figure 2: external and internal views of CEA's OSIRIS reactor (70MW MTR located in SACLAY, near PARIS, France)

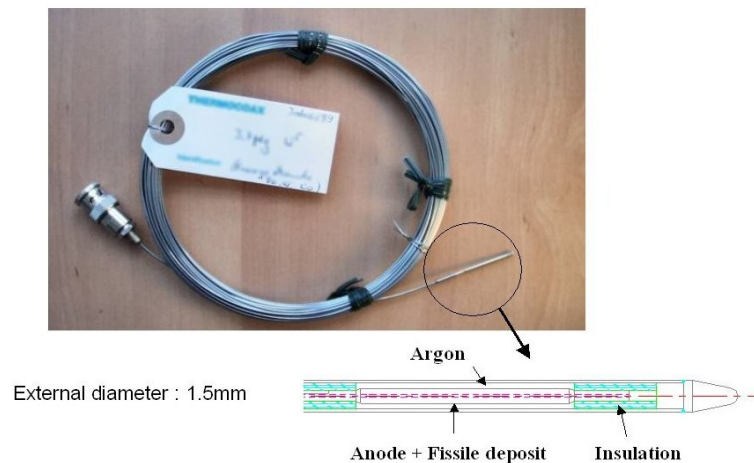
An overview of several on-going developments carried out by CEA for in-pile instrumentation is given below.

Sub-miniature fission chambers for neutron flux measurements

For decades, in-pile neutron flux measurements have been operated using Self Powered Neutron Detectors, which signals are generally correlated with post-irradiation analysis of activation foil dosimeters.

An important improvement has been recently achieved in this domain, with the development of CEA's sub-miniature fission chambers for in-pile measurements of high thermal neutron fluxes (up to 4×10^{18} n/m².s). These 1.5mm external diameter sensors, containing a ²³⁵U fissile deposit, were qualified in the SCK•CEN reactor BR2 in Mol (Belgium) between 2001 and 2004. This product has now been industrialized and is manufactured by PHOTONIS Company under the name "CFUZ53". More details about this program are given in [2].

*Figure 3:
1.5 mm sub-miniature fission chamber*



In addition, a new model of sub-miniature fission chambers dedicated to the measurement of fast neutron fluxes ($E > 1\text{MeV}$) has been studied and developed by CEA. As there is at this time no other sensor available to give online measurement of the fast neutron flux, this innovative fission chamber, based on a ²⁴²Pu fissile deposit, is very promising for all irradiation programs related to in-pile material radiation damage investigations.

Advanced prototypes of these "fast neutron flux fission chambers" have been manufactured and optimized. They are about to be irradiated for in-pile qualification campaigns in the OSIRIS and BR2 reactors during 2005 and 2006.

High temperature thermocouples for long-term irradiations

The necessity to carry on reliable and long-term in-pile high-temperature measurements has become particularly relevant for scientific programs relating on one hand to fission reactors studies (HTR, VHTR, Gen IV systems) and on the other hand to characterization of materials for nuclear fusion (ITER). The expected samples temperature for these long-term experiments ranges from 1100 to 1600°C.

Such measurements however encounter a strong limitation due to existing high-temperature thermocouples inability to withstand long term irradiations. Indeed in-pile high-temperature measurements (above 1100°C) are usually carried out using C type or S type thermocouples, respectively made of Tungsten-Rhenium and Platinum-Rhodium alloys. But when used under

irradiation, these thermocouples are altered by an important drift of their electrical signal mainly due to the transmutation of some of their elements. For example, submitted to significant thermal neutron flux, Rhenium and Tungsten are rapidly transformed into Osmium. These transmutations change the initial chemical composition of these couples, causing a permanent alteration of their thermoelectric response. This important drift requires proper compensation for future high-temperature irradiation programs.

Facing this problem, different scientific teams in the 1980's have studied some improvements of high-temperature thermocouples under long-term irradiation [3] [4], but these developments had not been completed.

In this scope, and following the results of previous studies, a new type of thermocouple for high-temperatures in-pile measurements has been developed and industrialized by CEA, in collaboration with THERMOCOAX Company [5]. This sensor is made of thermoelectric elements based on Molybdenum and Niobium, shielded by mineral insulator and appropriate metallic sheath. Both Mo and Nb have very low neutron absorption cross sections. As a consequence, this thermocouple -theoretically hardly affected by transmutations- should not therefore exhibit significant drift due to irradiation, even for enduring experiments.

The first phase of this development concerned the high-temperature compatibilities of different materials usable for sheath, insulators and wires, including tests of different diameters wires. The tests were managed in a high temperature furnace, in the range from 1000 to 1600°C, in pure helium atmosphere and in contact with graphite. The most significant materials tested are listed in Table 2.

The tests show strong reactivity between the majority of these materials at 1600°C; the most suitable associations were Mo and Nb wires with HfO₂ insulation and Nb or Ta sheath.

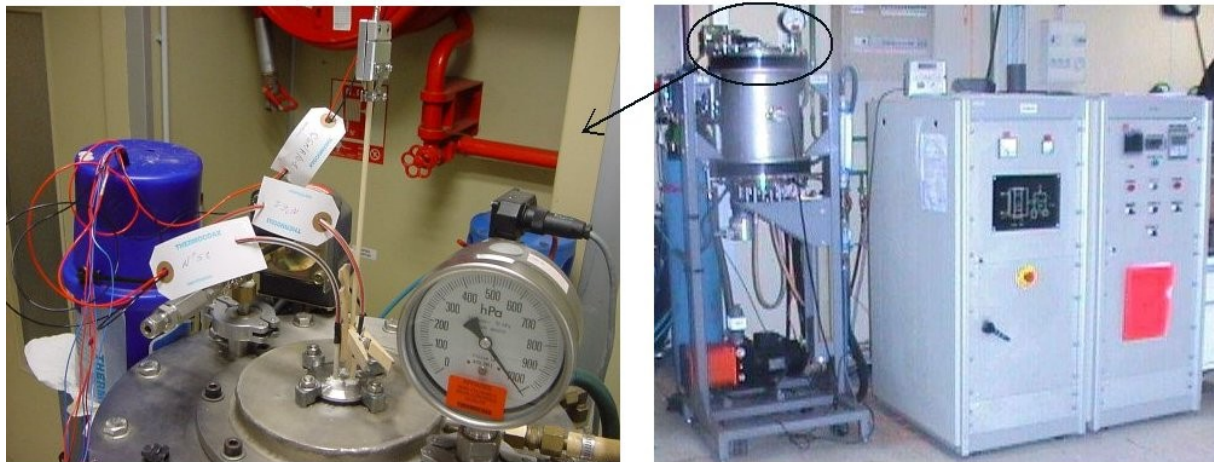


Figure 4: detailed and general views of the 2000°C THERMOCOAX furnace used for high-temperature studies



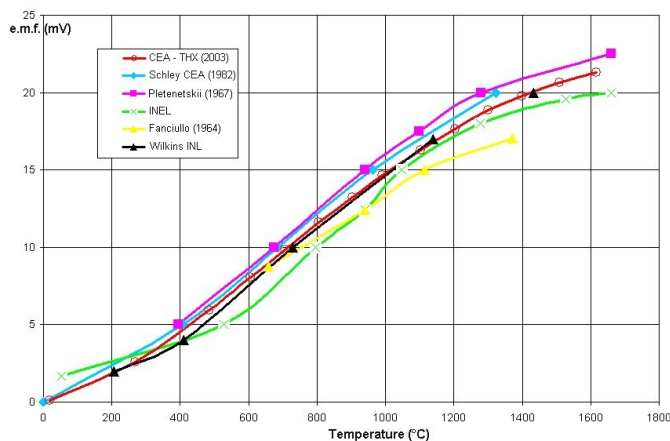
Figure 5: sheath materials samples

Wires	Mo	Nb
Insulators	Al ₂ O ₃	HfO₂
Sheath	Ta	Mo-Re 50% alloy
	Ti	Mo
	Re	Nb

Table 2: materials tested for high-temperature compatibilities

The second phase of the development was the definition of the thermoelectric response of the new Mo-Nb couple. This was established by using a very accurate calibration process in the temperature range from ambient to 1600°C. The result is shown in Figure 6. The curve obtained during this calibration is very consistent with previous results, for example from CEA and INL. The electromotive force of the Mo-Nb couple in the intended temperature range is about 14 $\mu\text{V}/^\circ\text{C}$, which is of the same order as the e.m.f. evolution of standard high-temperature thermocouples, such as C or S types. Interpolation polynomials have been calculated for low and high temperatures.

*Figure 6:
Thermoelectric response of the
Mo-Nb couple (last results in red),
compared with previous results from
various laboratories*



The next step concerned the thermal stabilization of these thermocouples. The objective was to determine the appropriate thermal treatment to be operated during the sensor fabrication, in order to minimize the thermal drift for high temperature use.

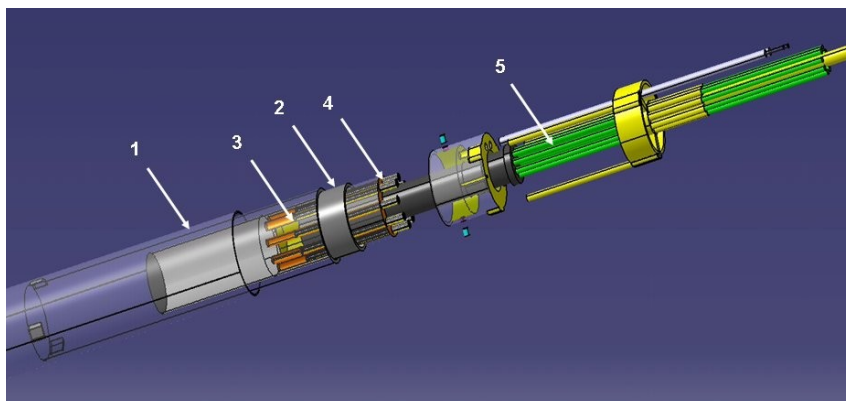
The efficiency of the selected treatment is finally studied during on-going enduring high-temperature out-of-pile tests. This 5000 hours duration campaign is planned to be finished at the end of year 2005. Preliminary results show a good behavior of these thermocouples, with a residual thermal drift lower than 0.02°C/h at 1100°C.

Other kind of standard thermocouples, such as K, N and C types, are also tested in this high-temperature endurance test, with the objective to get a better knowledge of their long-term behavior in the above thermal conditions.

The last phase of the development will be the in-pile high-temperature qualification, which is scheduled during 2006 and 2007 in the OSIRIS reactor. The principle is to measure the signal of both standard high-temperature and new Mo-Nb thermocouples during the irradiation, in order to determine their possible drifts as a function of the neutron fluence.

So a specific and long-term neutron irradiation experiment called “THERMEX” is under preparation in CEA (see Figure 7). During irradiation, the reference temperature will be regularly established by using a pure copper fixed point fusion device, and will be compared with the temperature measured by the thermocouples [6].

*Figure 7:
exploded view of
THERMEX experiment
device:
stainless steel capsule (1),
graphite barrel (3) filled
with pure Copper (2),
additional electric heating
elements (4),
thermocouples (5)*



After qualification process ended, these promising thermocouples will be available as industrial sensors from THERMOCOAX Company.

Fission gas release acoustic measurements

In-situ measurement of fission gas release inside nuclear fuel rods is of major interest for irradiation programs in Material Testing Reactors. This parameter is particularly relevant for scientific expertise on PWR fuel rods and to update data bases for nuclear safety. As fission gas release kinetics is a relevant indicator of the state of nuclear reactions, its measurement is a key issue for burn-up increase studies.

The intended application is based on acoustic measurements and uses a high-frequency echography technique [7]. The principle is to implement a piezoelectric sensor measuring the acoustic waves velocity in the gas contained inside the fuel rod. This sensor can generate a high-frequency acoustic pulse in a cylindrical cavity directly connected to the plenum of the fuel rod, and then measures the echoes due to the propagation of the acoustic waves in the cavity (see Figure 8).

It has been proved that in these conditions the acoustic waves velocity is directly dependant of the fraction of fission products in the helium gas [8]. So this online measurement could directly give the fraction of the fission gas released during the irradiation.

As this technique is relatively easy to implement and requires no mechanical parts, its in-pile application is really promising for future irradiation programs.

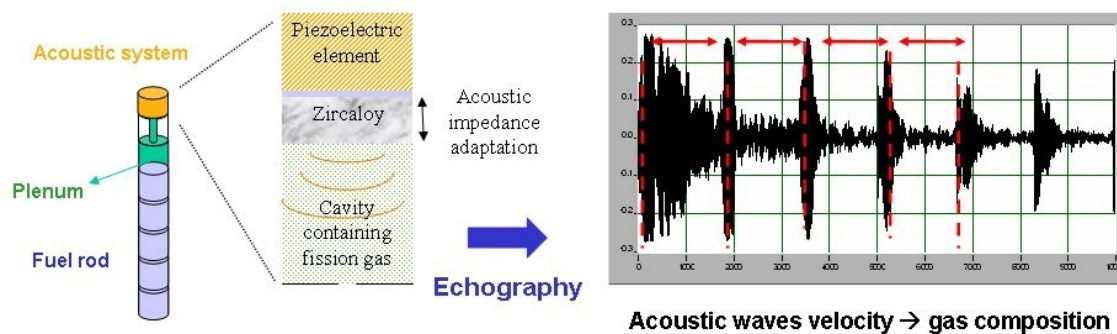


Figure 8: principle of the in-pile fission gas release acoustic measurement

These studies, initiated by EDF (Electricité de France) and LAIN (Interfaces and Nanophysics Analysis Laboratory), are carried out since 2003 through a scientific collaboration between CEA and LAIN.

In a first phase of the development, the feasibility to measure He-Xe mixture composition has been validated on an out-of-pile bench test. A preliminary design of the fission gas release sensor has been manufactured and tested, in order to ensure a sufficient propagation of ultrasonic waves inside the gas cavity, solving the problem of the small acoustic impedance of gas compared to solid one, which induces a strong insertion loss.

The signal processing system has also been developed in order to properly and automatically extract the He-Xe mixture composition parameter.

In parallel, piezoelectric sensors have been successfully tested in 2004 under pure gamma radiation up to 1.5 MGy [9], and will be irradiated in a mixed neutron-gamma environment during 2005 in the SCK•CEN reactor BR1. These tests aim at measuring the effects of irradiation on piezoelectric sensors, in order to select the most appropriate materials and to determine optimal position of the sensor in future irradiation experiments.

In the coming years, an optimized integration of the acoustic assembly on fuel rods will be designed and tested, with the goal to reach in the middle term in-pile qualification of this innovative and promising fission gas release sensor.

References

[1] High accuracy sensor for online measurement of the fuel rod internal pressure during irradiation experiments

JF. VILLARD, G. LEMAITRE, JM. CHAUSSY, F. LEFEVRE - 7th International Topical Meeting on Research Reactor Fuel Management (RRFM 2003), Aix-en-Provence (France), March 2003

[2] In-pile CFUZ53 sub-miniature fission chambers qualification in BR2 under PWR conditions

C. BLANDIN, S. BREAUD, JM. LAURENS, L. ORIOL, L. VERMEEREN, M. WEBER – 10th International Group on Research Reactors (IGORR) meeting, Gaithersburg (USA), September 2005

[3] Thermocouples for Measurements under Conditions of High Temperature and Nuclear Radiation

R. SCHLEY, G. METAUER - Temperature: Its Measurement and Control in Science and Industry, Vol 5 Part 2, 1109-1113 (1982)

[4] Low Cross-Section Mo-Nb Thermocouples for Nuclear Applications: the State-of-the-Art

S.C. WILKINS and al. - 5th Symposium on Space Nuclear Power Systems, Albuquerque (USA), January 1988

[5] Characterisation of a thermocouple for high temperature measurement under irradiation - Definition and properties of the Molybdenum Niobium thermocouple

S. FOURREZ, G. BAILLEUL, M. KOLLEN, JF. VILLARD - 12th International Metrology Congress, Lyon (France), June 2005

[6] High temperature measurement needs for irradiation experiments in Material Testing Reactors - Development of a new type of thermocouple and interest of high temperature fixed points for its characterisation and in-pile qualification

JF. VILLARD, S. FOURREZ - Physikalisch-Technische Bundesanstalt Seminar on High-temperature fixed-points for industrial and scientific applications, Berlin (Germany), April 2005

[7] Determination of the composition of a gas mixture in a nuclear fuel rod by an acoustic method

MF. NARBAY, D. BARON, G DESPAUX, JM SAUREL - Journal of the British Institute of Non-Destructive Testing (INSIGHT), vol 42, 603-605 (2000)

[8] An ultrasonic sensor for pressure and fission-gas release measurements in fuel rods for pressurised water reactors

JY. FERRANDIS, G. LEVEQUE, F. AUGEREAU, E. ROSENKRANTZ, D. BARON, JF. VILLARD - 9th International conference on CANDU fuel, Belleville (Canada), September 2005

[9] PZT Materials under Gamma irradiation

F. ALGABER, JY. FERRANDIS, F. AUGEREAU, JF. VILLARD - 4th European Workshop on Piezoelectric Materials, Montpellier (France), July 2004