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In-Pile Qualification of the Fast-Neutron-Detection-System

D. Fourmentel¹, J-F. Villard¹, C. Destouches¹, L. Barbot¹, B. Geslot², L. Vermeeren³, M. Schyns³

Abstract— In order to ensure the quality and the relevance of irradiation programs in the future Jules Horowitz Reactor (JHR), the French Alternative Energies and Atomic Energy Commission (CEA) has significantly increased its research and development effort in the field of in-pile instrumentation during the last decade. Major progresses have thus been achieved in the capability to perform accurate in-pile measurements using reliable and updated techniques.

A significant part of this effort have been conducted in the framework of the Joint Instrumentation Laboratory between the CEA and the Belgian Nuclear Research Centre (SCK•CEN).

In order to improve measurement techniques for neutron flux assessment, a unique system for online measurement of fast neutron flux has been developed and recently qualified in-pile. The Fast-Neutron-Detection-System (FNDS) has been designed to monitor accurately high-energy neutrons flux ($E > 1$ MeV) in typical Material Testing Reactor conditions, where overall neutron flux level can be as high as 10^{15} n.cm⁻².s⁻¹ and is generally dominated by thermal neutrons. Moreover, the neutron flux is coupled with a high gamma flux of typically a few 10^{15} γ.cm⁻².s⁻¹, which can be highly disturbing for the online measurement of neutron fluxes.

The patented FNDS system is based on two detectors allowing the simultaneous detection of both thermal and fast neutron flux. Thermal neutrons can be measured using a Self-Powered-Neutron-Detector (SPND) or a ²³⁵U miniature fission chamber, while fast neutron detection requires a miniature fission chamber with a special fissile material presenting an energy threshold near 1 MeV, which can be ²⁴²Pu for MTR conditions. Fission chambers are operated in Campbelling mode for an efficient gamma rejection. FNDS also includes a specific software that processes measurements to compensate online the fissile material depletion and to adjust the sensitivity of the detectors, in order to produce a precise evaluation of both thermal and fast neutron flux even after long term irradiation.

FNDS has been validated through a two-step experimental program. A first set of tests was performed at BR2 reactor operated by SCK•CEN in Belgium. Two FNDS prototypes were operated in-pile during nearly 1000 hours. These tests exhibited the consistency of the measurement of thermal to fast neutron flux ratio with Monte Carlo calculations, as well as the right compensation of fissile material depletion. Then a second test was recently completed at ISIS reactor operated by CEA in France. For this irradiation, FNDS signal was compared to reference thermal and fast neutron flux measurements using activation dosimeters analyzed under COFRAC® Quality Certification. During this latter test, FNDS proved its ability to measure online the fast neutron flux with an overall accuracy better than 5%.

This paper describes the innovative features of FNDS and discusses the results of its final in-pile qualification. FNDS is now operational and is assumed to be the first and unique acquisition system able to provide an online measurement of the fast neutron flux in MTR conditions. This system will of course be used to perform spectral neutron characterization of JHR channels, but it may also be implemented in future irradiation experiments, for

a better and real-time evaluation of the fast neutron flux received by material and fuel samples.

Index Terms—Nuclear Measurements, Fission Chamber, Reactor Instrumentation

I. INTRODUCTION

FAST neutron flux ($E \geq 1$ MeV) is a key neutron parameter in nuclear reactors. Its measurement is particularly relevant to assess material damage under irradiation. In very low power research reactors, such as ZPRs (Zero Power Reactors), the fast neutron flux is usually measured on-line with specific fission chambers, containing for example ²³⁸U coating. In more powerful reactors, such as Material Testing Reactors (MTRs) or nuclear power plants (NPPs), techniques used in ZPRs are not appropriate. This is mainly due to the rapid evolution of the fissile coating under high neutron flux that reduces the sensitivity of the sensor to the ‘fast’ spectrum domain (cf. II.B). This neutron parameter is therefore not accessible on-line and is generally evaluated afterward by activation dosimetry or by Monte Carlo calculations. However, in the current context of increasing requirements for in-pile measurements, particularly for experiments in MTRs, the on-line measurement of fast neutron flux has become a major objective.

In order to ensure the quality and the relevance of irradiation programs in the future Jules Horowitz Reactor (JHR), the French Alternative Energies and Atomic Energy Commission (CEA) has significantly increased its research and development effort in the field of in-pile instrumentation during the last decade. In this context and in the framework of the Joint Instrumentation Laboratory between the CEA and the Belgian Nuclear Research Centre (SCK•CEN), a measurement system dedicated to on-line measurement of fast neutron flux has been developed.

Next paragraphs describe the innovative features of the Fast-Neutron-Detection-System (FNDS) and discuss the results of its final in-pile qualification.

II. FNDS SPECIFICATIONS

On-line measurements of fast neutron flux are based on the use of a fission chamber with a specific fissile coating, presenting an energy threshold near the fast domain ($E > 1$ MeV). If the use of such fission chamber is not a problem at very low neutron flux, it faces major difficulties under intense mixed neutron and gamma fluxes. First of all,

because of the low neutron sensitivity of this type of fission chamber, the high gamma flux of MTRs leads to a high gamma contribution to the signal. Furthermore, neutron captures on the nuclei of the fissile coating create isotopes that have a high cross-section in the thermal spectrum domain. Thus the ‘fast’ sensitivity of the detector decreases rapidly. Isotopic impurities of the coating must also be taken into account. Figure 1 illustrates this difficulty for the use of ^{242}Pu , which has a suitable response in the fast spectrum domain [1].

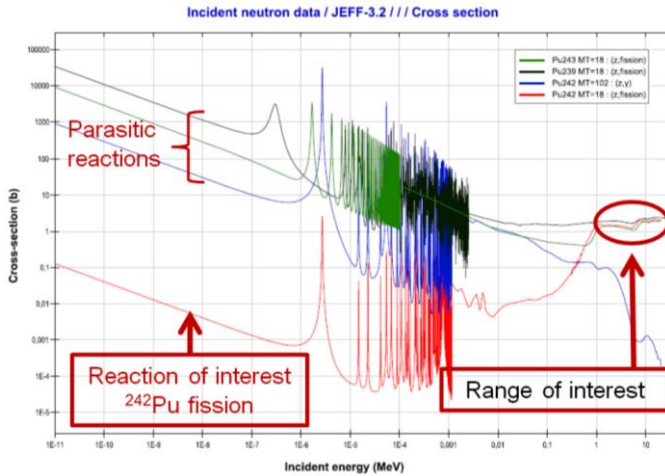


Fig. 1. Fission cross sections of a typical ^{242}Pu fissile coating showing that impurities and isotopes created by capture must be taken into account for fast neutron flux measurements

We can notice that impurities and isotopes created by neutron captures induce an increasingly high sensitivity to thermal neutrons.

A. FNDS general description

In order to compensate difficulties mentioned above, FNDS includes an acquisition system allowing measurements with fission chambers in Campbell mode. This mode allows a very efficient rejection of the gamma contribution [2] while offering a very wide dynamic range. FNDS also integrates signal processing software that firstly calculate in real time the evolution of the fissile coating of the fission chamber using an evolution code DARWIN [3], and that secondly deduce the corresponding evolution of the neutron sensitivity of the fast fission chamber. This software extracts in real-time the value of the fast neutron flux from the signal.

In order to allow a precise evaluation of thermal neutron fluence received by the ‘fast’ fission chamber, FNDS is equipped with a second detector that is sensitive to thermal neutron flux and positioned near the ‘fast’ fission chamber. This second detector may be a fission chamber with ^{235}U coating or a Self-Powered Neutron Detector (SPND).

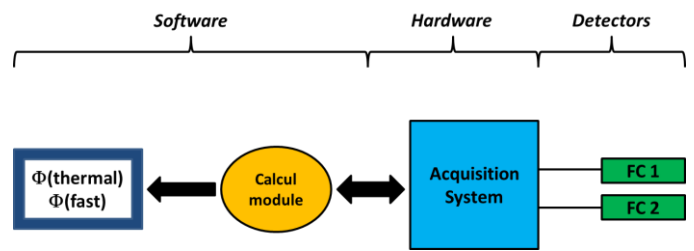


Fig. 2. Overview of FNDS components

FNDS thus consists in 2 detectors for ‘fast’ and ‘thermal’ spectrum domains, an acquisition system operating in Campbell mode and a calculation module for on-line analysis of neutron fluxes (Figure 2). These innovations were patented for the detectors [4] and for the entire measurement system [5].

B. Detectors

An important bibliography study was carried out to establish the state of the art of neutron and photonic measurements in MTRs [6, 7], and led us to select fission chamber sensors. It was shown that the most suitable fissile isotope for the estimation of the fast neutron flux is ^{242}Pu under the typical irradiation conditions of MTRs, such as OSIRIS (CEA Saclay – France), BR2 (Mol – Belgium) and JHR reactors [1, 8]. Figure 3 illustrates the reason why ^{242}Pu coating is the best choice compared to other fissile isotopes; it shows the stability of its fast neutron sensitivity at high fluence.

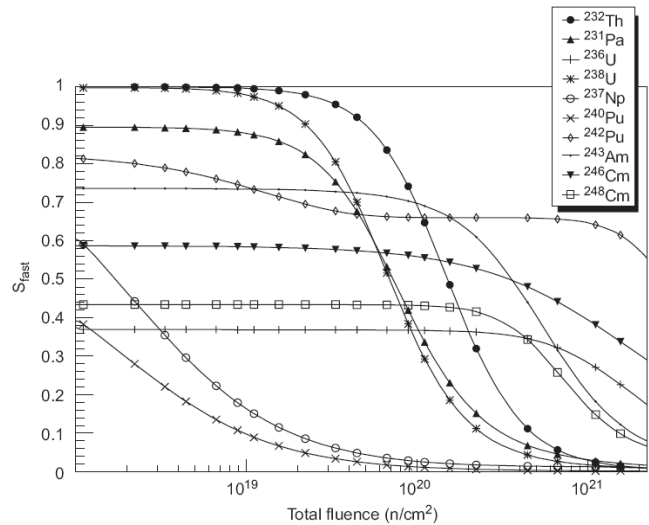


Fig. 3. Sensitivity to fast neutrons for initially pure deposits under typical BR2 neutron spectrum [3]

A typical ‘fast’ fission chamber selected for MTRs contains $100\ \mu\text{g}$ of ^{242}Pu while the typical associated ‘thermal’ detector contains $10\ \mu\text{g}$ of ^{235}U . Both sensors have a 3 mm outer diameter body and integrated mineral insulated cable.

These sensors have been developed and manufactured at the Fission Chamber Manufacturing Workshop located in CEA Cadarache.

As explained previously, the analysis of fission chamber

signal requires a precise knowledge of its isotopic composition and as well as the effective deposited mass of fissile materials. It is necessary to quantify with the best accuracy their contribution to the signal. For the in-pile qualification of FNDS (§.III), analyzes of fissile coatings give the following results:

$m(^{235}\text{U})$:	10.4 μg			
isotope:	^{234}U	^{235}U	^{236}U	^{238}U
% mass:	1.66	97.65	0.15	0.54

$m(^{242}\text{Pu})$:	98.3 μg				
isotope:	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
% mass:	0.003	0.009	0.019	0.006	99.963

As a reminder, the odd isotopes of plutonium are mostly fissile in the ‘thermal’ spectrum domain and have very high fission cross sections compare to ^{242}Pu in the ‘fast’ spectrum domain (cf. Figure 1).

C. Acquisition system

The acquisition system has 2 channels for the sensor couple. Each channel is equipped with a high-voltage power supply for polarization with adjustable voltage (typically 250 V to reach the saturation plateau of a 3 mm fission chamber). The measured low-level current signals pass through a preamplifier (manufactured by CEA Saclay), amplification stage and then are filtered and digitalized for processing with the measurement interpretation software.

There are typically 3 modes to operate fission chambers: pulse, fluctuation (or Campbelling) and current modes. In MTRs conditions, the advantage of the Campbelling mode is the radical limitation of the gamma contribution on the signal [2] compared to current mode. Indeed, MTR neutron fluxes are too high to operate such fission chambers in pulse mode.

In Campbelling mode, the measurement of the variance of low fission chamber signal with a specific bandwidth has required the development of a dedicated acquisition system. As such a system was not available from Industry; prototypes were specially designed at CEA. Studies are on-going for the industrialization of such acquisition system in a short term [9].

D. Processing software

The FNDS processing software calculates the sensitivity of each fission chamber to the thermal, epithermal and fast neutrons from specific input data and then calculates the thermal and fast neutron fluxes versus time. Input data required for the processing are the following:

- typical neutron spectrum at the location of the detectors,
- nuclear data library,
- mass and isotopic composition of the fissile coatings,
- calibration coefficient of the fission chambers (§.III.B),
- calibration coefficient of the electronic (§.III.B),
- variance measurements.

Processing software includes DARWIN code [6], developed

by the CEA, to compute fission cross sections of each isotope present in the fissile coating and to calculate the isotopic evolution of these coatings with fluence and time. The method to combine measurements and estimate fast neutron flux is the following [7, 8]:

If R_f is the fission rate of the ‘fast’ fission chamber and R_t the fission rate of the ‘thermal’ fission chamber, the following two equations can be written:

$$\begin{aligned} R_1 &= S_{th_1} \cdot \Phi_{th} + S_{\acute{e}pi_1} \cdot \Phi_{\acute{e}pi} + S_{rap_1} \cdot \Phi_{rap} \\ R_2 &= S_{th_2} \cdot \Phi_{th} + S_{\acute{e}pi_2} \cdot \Phi_{\acute{e}pi} + S_{rap_2} \cdot \Phi_{rap} \end{aligned}$$

with S_i thermal, epithermal and fast neutron sensitivity [$1/\text{n.cm}^{-2}$] and Φ_j thermal, epithermal and fast neutron fluxes [$\text{n.cm}^{-2}.\text{s}^{-1}$].

To solve this system of 2 equations with 3 unknown variables we assume that the ratio between epithermal and fast fluxes is constant during irradiation:

$$\Phi_{\acute{e}pi} = \alpha \cdot \Phi_{rap}$$

This hypothesis was verified by calculations [10]. It allows to reduce our system to 2 equations with 2 unknown variables and to establish the following equation of the fast neutron flux:

$$\Phi_{rap} = \frac{\frac{R_1}{S_{th_1}} - \frac{R_2}{S_{th_2}}}{\frac{\alpha \cdot S_{\acute{e}pi_1} + S_{rap_1}}{S_{th_1}} - \frac{\alpha \cdot S_{\acute{e}pi_2} + S_{rap_2}}{S_{th_2}}}$$

However, in a light water reactor, ^{235}U coating is predominantly (~99% initially) sensitive to thermal neutrons [6, 7] and makes it possible to measure thermal neutron flux without calculation assumptions. In this case, fast neutron flux can then be written in the following reduced form:

$$\Phi_{rap} = \frac{R_1 - S_{th_1} \cdot \Phi_{th}}{S_{rap_1} + \alpha \cdot S_{\acute{e}pi_1}}$$

III. IN-PILE QUALIFICATION OF FNDS

FNDS has been validated through a two-step experimental program. A first set of tests was performed at BR2 reactor through the FICTIONS-8 experiment. Two FNDS prototypes were operated in-pile during nearly 1000 hours. This experiment exhibited the consistency of the measurement of thermal to fast neutron flux ratio with Monte Carlo calculations, as well as the right compensation of fissile material depletion [11].

A second set of tests was then necessary to qualify the absolute fast neutron flux measurement process. The main requirement was to perform measurements in well-controlled conditions (neutron spectrum, flux gradient, etc.) in order to minimize external sources of uncertainty and bias. In this context, it was decided to carry out the EFICAF (Estimation of

Flux in Isis by Fission Chamber) experiment in ISIS reactor (CEA Saclay, France). ISIS reactor allows stable and reproducible irradiation conditions. Its neutron flux level is also sufficient to operate fission chambers in Campbelling mode. Moreover, its flexibility allowed to carry out all the measurements that were necessary for the qualification (on-line measurements and activation dosimetry measurements without external disturbance).

A. EFICAF specifications

It was decided to measure the absolute thermal and fast neutron fluxes in a single irradiation channel of ISIS core. Fission chambers were located on both sides of the maximum neutron flux height in order to receive the same neutron flux with a simple experimental device and to avoid a moving device. Reactor power was specified at 250 kW_{th}, on one hand for easy implementation of the experiment and on the other hand to minimize the fluence and the corresponding evolution of fissile materials.

For this experiment, reference neutron flux measurements were performed using activation dosimeters. They provided a precise evaluation of the neutron flux profile in the experimental channel. These dosimeters were the following:

- 7 pure iron (diameter 10 mm, thickness 100 μm),
- 7 pure nickel (diameter 10 mm, thickness 125 μm),
- 7 cobalt (AlCo1% - diameter 10 mm, thickness 100 μm) including 1 in a cadmium shield (for the epithermal ratio on cobalt).

Finally, Monte Carlo calculations were performed with TRIPOLI4 code [12] for FNDS processing software and for activation dosimeter analysis.

B. Fission chambers calibration in Campbelling mode

A method to calibrate fission chambers in Campbelling mode has been developed [13]. This calibration was carried out for FNDS sensors in MINERVE zero power reactor at CEA Cadarache.

In this calibration it is assumed that the measured signal is equal to the product of the coating fission rate, a calibration coefficient of the electronic and a calibration coefficient of the detector. The variance of the signal can then be expressed according to the following relation:

$$\sigma_V^2 = K_e \cdot K_d \cdot R$$

with: σ_V^2 : variance measurement [V²], K_e : coefficient of the electronic channel [Ω².Hz], K_d : calibration coefficient of the detector [A².Hz⁻²] and R : total fission rate of the coating [Hz].

This method was used for the first time in 2012 for the CARMEN-1 experiment led in OSIRIS reactor at CEA Saclay. It exhibited excellent results for ²³⁵U fission chamber used in Campbelling mode by comparison with rhodium SPND and activation dosimeters [14]. EFICAF experiment was therefore

the second opportunity to use this calibration method. The calibration coefficient obtained for both fission chambers is:

$$K_d = 3.75 \cdot 10^{-27} \text{A}^2 \cdot \text{Hz}^{-2}$$

We can notice that the calibration coefficient is the same for both fission chambers. Indeed, the average charge of a fission product between the electrodes is mostly similar for both coatings. This calibration coefficient value also depends on the signal bandwidth. It is therefore important to characterize the bandwidth of the analogue-to-digital processing for an appropriate use of the calibration coefficient. Details about the calibration are given in [13, 15].

According to the relation between fission rate 'R' and variance measurements σ_V^2 there is a second calibration coefficient K_e to be calculated. K_e links the current signal delivered by the fission chambers to the corresponding variance [V²] for each measurement channel.

The calibration coefficients of the electronics for the 2 measurement channels of FNDS were the following:

$$K_{e(\text{channel1})} = 5.45 \cdot 10^{15} \Omega^2 \cdot \text{Hz}$$

$$K_{e(\text{channel2})} = 4.91 \cdot 10^{15} \Omega^2 \cdot \text{Hz}$$

C. Monte Carlo and experimental results

Activity measurements of the EFICAF dosimeters were carried out at the MADERE platform of CEA Cadarache under COFRAC[®] Quality Accreditation [16]. Table 1 below summarizes the neutron flux results calculated by TRIPOLI4 code and measured by activation dosimetry at the locations of the two fission chambers.

	TRIPOLI* (n.cm ⁻² .s ⁻¹)	AlCo 1% (n.cm ⁻² .s ⁻¹)	C/E		
Thermal neutron flux	1,04E+12	1,00E+12	3,3%		
	TRIPOLI* (n.cm ⁻² .s ⁻¹)	Fe (n.cm ⁻² .s ⁻¹)	C/E	Ni (n.cm ⁻² .s ⁻¹)	C/E
Fast neutron flux	4,74E+11	4,58E+11	3,4%	4,58E+11	3,4%

C/E : TRIPOLI4/Dosimeter type -1 [%]

The results of TRIPOLI4 calculations and dosimetry measurements are in good agreement to assess neutron fluxes values given by FNDS.

Thermal and fast neutron fluxes measured by FNDS are compared with values given the TRIPOLI4 code and activation dosimetry respectively in Table 2 and Table 3.

	CF U235 (n.cm ⁻² .s ⁻¹)	(TRIPOLI/CF U235)-1	(AlCo 1%/CF U235)-1
1st measurements	9.34E+11	11.0%	7.5%
2nd measurements	9.15E+11	13.3%	9.7%

These results confirm the good agreement for absolute thermal neutron flux using Campbelling mode operation and the corresponding calibration process [14].

TABLE III

FAST NEUTRON FLUX EVALUATIONS AND DEVIATIONS BETWEEN FNDS AND TRIPOLI4 CALCULATIONS AND BETWEEN FNDS AND FE, NI MEASUREMENTS

	CF Pu242 ($\text{n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$)	(TRIPOLI/CF Pu242)-1	(Fe-Ni/CF Pu242)-1
1st measurements	4.39E+11	7.9%	4.4%
2nd measurements	4.40E+11	7.7%	4.1%

The deviation between FNDS fast neutron flux measurements and dosimetry results is mainly covered by the uncertainties of the activation dosimetry measurements (typically 4% for fast neutron flux evaluation). The results of this experiment confirm the quality of the on-line fast neutron flux measurements at a few $10^{11} \text{ n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$ ($E > 1 \text{ MeV}$) with this type of miniature fission chambers and therefore confirm FNDS capability to measure in real-time the absolute thermal and fast neutron fluxes with an excellent accuracy.

IV. CONCLUSION

EFICAF experiment demonstrated the ability of FNDS to measure on-line thermal and fast neutron fluxes. Deviations between fast neutron flux measured by FNDS and activation dosimetry results are below 5%. These results confirm the excellent accuracy of the analysis method using a couple of ^{235}U and ^{242}Pu fissile coatings and special calibration in Campbelling mode.

FNDS is now operational and is assumed to be the first and unique acquisition system able to provide an on-line measurement of the fast neutron flux in MTR conditions. This system will of course be used to perform spectral neutron characterization of JHR channels, but it may also be implemented in future irradiation experiments, for a better and real-time evaluation of the fast neutron flux received by material and fuel samples.

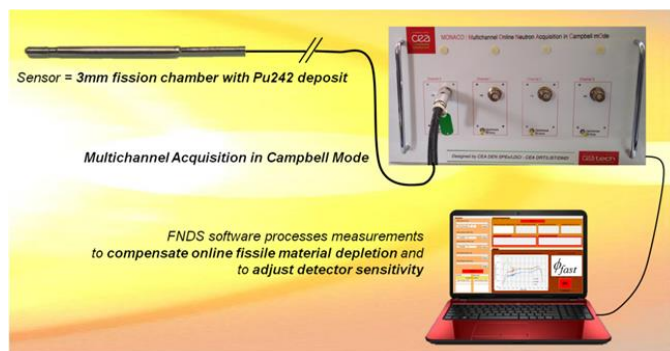


Fig. 4. Illustration of updated FNDS based on the Multichannel Acquisition System MONACO [9].

Further studies are on-going to update the FNDS acquisition system using a multi-channel and multi-mode acquisition system called MONACO (Multichannel Online Neutron Acquisition in Campbell mOde) [9,17]. Studies are also carried out to assess the uncertainty associated with neutron flux analysis calculations, mainly for the calibration coefficients of the detectors. Finally, additional tests will

shortly be performed to evaluate FNDS ability to measure also the epithermal neutron flux.

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