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NEUTRON AND GAMMA-RAYS SENSITIVITY OF NEW FIBER-MOUNTED SCINTILLATION NEUTRON DETECTORS FOR SPENT FUEL PIN DIVERSION MEASUREMENTS

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Abstract

The verification of spent fuel represents a pressing safeguards challenge in view of the increasing amount of spent fuel and the commissioning of spent fuel repositories. In this framework, we are investigating a novel method to detect and identify missing/diverted fuel rods in a spent fuel assembly, which were replaced by dummy rods. The method is based on performing measurements with thin fiber-based scintillation neutron detectors, inserted into either the detector channel positions in the assembly, or in between the fuel pins. The neutron flux and its radial gradient are measured and compared to the ones expected to exist in an intact assembly.

In this work we report about the measurements carried out at the BR1 research reactor and the Laboratory for Nuclear Calibrations (LNK) facilities of the Belgian Nuclear Research Centre SCK CEN. At BR1 we measured the detector response in a well-known Maxwellian thermal neutron flux and determined the detector sensitivity to thermal neutrons. These measurements allowed also to assess the impact of dead time as well as the impact of gamma-rays on the detector response. Additional measurements at LNK were instrumental to study the sensitivity of the detector to different exposures with ^{137}Cs and ^{60}Co calibrated sources. The detectors were insensitive to gamma-rays up to a dose rate of 70 Gy/h.

The obtained neutron sensitivity was compared with estimations of a Monte Carlo model based on the nominal detector data. Building on previous work carried out at SCK CEN, the obtained sensitivity was used to estimate the expected count rates and measurement times with an intact 17×17 PWR fuel assembly in a spent fuel pool.

1. INTRODUCTION

The characterisation of spent nuclear fuel (SNF) assemblies via non-destructive assay (NDA) is very important for the implementation of safeguards, in addition to the safety assessment associated to its handling and disposal. Current methods for the verification of the integrity of the SNF assemblies rely on the detection of the Cherenkov light or total neutron counting when the assemblies are located in the spent fuel pool. A recently developed method relies on passive gamma emission tomography; it has the potential of an improved accuracy but the system is expensive, bulky and requires the movement of the SNF from the storage position.

An ongoing collaboration between the Belgian Nuclear Research Centre, SCK CEN, and Chalmers University of Technology focusses on studying the possibility to perform neutron measurements with movable detectors positioned either between the fuel pins or right next to the fuel assembly. If the spatial dependence of the neutron flux, and hence its gradient, is measured in several radial positions in the assembly, a comparison with the expected neutron flux distribution contains information on possibly missing pins and their positions. For this purpose, thin fibre prototype detectors available are considered.

In this framework, we characterized the neutron and gamma-ray response of two $^6\text{LiF}(\text{ZnS})$ based miniature scintillators. The neutron sensitivity was determined in a well-known thermal neutron flux as well as the sensitivity to gamma-ray. In this paper, we will first describe the measurement campaigns that were carried out and the results obtained. The results allowed defining an equivalent Monte Carlo model of the detector which was used to estimate the expected behaviour with 17×17 PWR assemblies in wet storage conditions.

2. MEASUREMENTS

2.1. Detector and electronics

The detectors that are considered are $^6\text{LiF}(\text{ZnS})$ miniature scintillators [MOR94,YAG11]. The crystals are coupled to an optical fiber serving as light guide which connects to an Hamamatsu R9880U-210 Photo-Multiplier Tube (PMT) (where the scintillation photons generate electrons) and an Hamamatsu E10679-02 D type socket assembly (which contains the voltage divider circuit), as shown in Fig. 1.

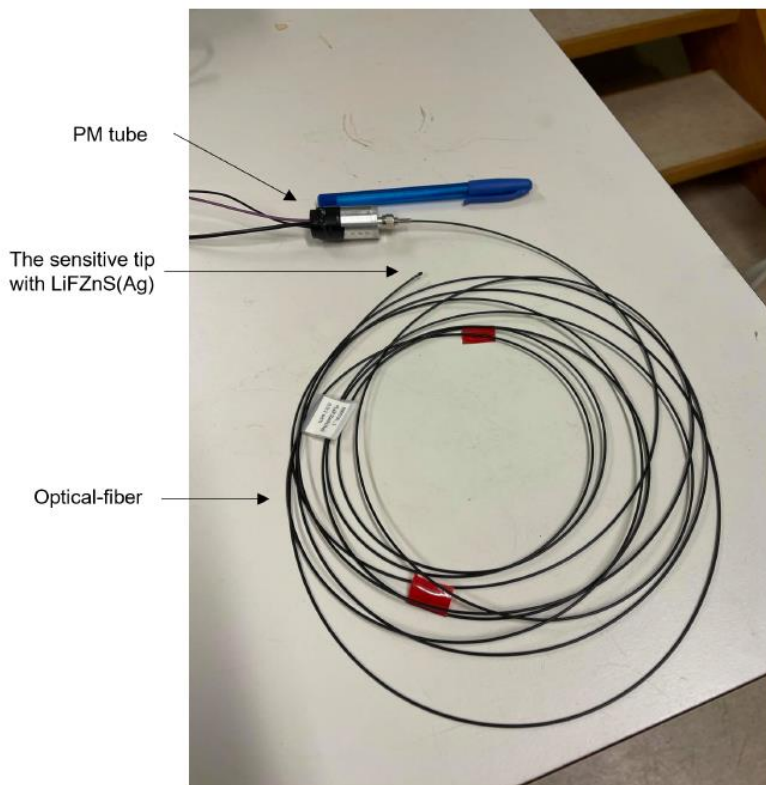


Figure 1. $^6\text{LiF}(\text{ZnS})$ miniature scintillators coupled with an optical fibre to a PMT.

The output signal of this is fed to a charge sensitive- preamplifier and then to the data acquisition system. Two detectors, with identical nominal characteristics and indicated as detector A and detector B, were measured in the same way.

The data-acquisition system used is the MCA527 module from GBS Elektronik GmbH [MCA]. It digitally processes the signal from the preamplifier. In addition it also supplies the necessary high voltage (~ 800 V) for the PMT and power for the pre-amplifier. Spectra can be acquired with the WinSPEC software package [WINSPEC]. The MCA settings were as follows: FlatTop time of $2.0\ \mu\text{s}$, Shaping Time of $3.0\ \mu\text{s}$, and Trigger Filter [+1;-2;+1]. Baseline restoring was set at 1/16. The spectra were acquired in 2048 channels. Pile-up rejection (PUR) was active. Coarse Gain (CG) 2 and Fine Gain (FG) 0.7.

2.2. Gamma-ray sensitivity measurements at LNK

Due to the large gamma-ray field associated with spent fuel assemblies, gamma-ray sensitivity is an important issue when it comes to neutron detection from spent fuel assemblies. In the Fork detector setup, fission chambers are used due to their insensitivity to gamma-rays. The use of other detectors such as ^3He require heavy lead shielding to limit the gamma-ray flux at the detector position [MEN2009].

To investigate this aspect, a set of experiments was performed in the Laboratory for Nuclear Calibrations (LNK) at SCK CEN. In the experiment, the two scintillators were exposed to ^{137}Cs and ^{60}Co sources with different dose rates between 5 mGy/h and 200 Gy/h.

The detectors did not appear to be sensitive up to a dose rate of up to about 50 Gy/h. Sensitivity to gamma-rays can be measured starting from dose rates of around 77 Gy/h. The results when using different channel thresholds are shown in Table 1.

We could estimate a sensitivity of less than 2.0×10^{-3} counts per second per Gy/h of absorbed dose. Our results indicated that detector A provides slightly higher sensitivity in comparison with scintillator B and that by increasing the threshold the sensitivity can be reduced. At a threshold of 100, no sensitivity could be measured.

TABLE 1. Gamma-ray sensitivity at different thresholds for the two detectors

Dose rate Gy/h	Detector	Sensitivity in c/s per 100 Gy/h		
		Threshold 10	Threshold 20	Threshold 100
77.2	A	1.8(1)	0.3(<1)	---
192	A	1.3(1)	0.4(<1)	---
77.2	B	0.4(1)	0.1(<1)	---
192	B	0.8(1)	0.2(<1)	---

The experiment proved that the neutron scintillators do have a sensitivity to gamma radiation.

In a spent fuel assembly the gamma dose rate can be up to 1000 Gy/h [27] which is 5 times higher than 200 Gy/h, so further testing is required to study the behaviour of the scintillators under a more intense gamma radiation field. Such dose rates can be achieved at other irradiation facilities (e.g. RITA) at SCK CEN.

2.3. Thermal neutron sensitivity measurements at BR1

To determine the detector sensitivity under well-known thermal neutron flux conditions, measurements at the BR1 reactor were performed [BR1a, BR1b]. The reactor is air-cooled and graphite-moderated. A schematic representation is given in Fig. 2.

At the top of the reactor a spherical cavity with a radius of 50 cm is available for irradiation and calibration of detection instruments under a well-defined Maxwellian thermal neutron flux. The neutron flux at the centre of the cavity is constantly monitored using a calibrated fission chamber. The two scintillators were inserted one at a time into the cavity by using an aluminium pipe. The pipe length was such that the detectors were at the centre of the cavity.

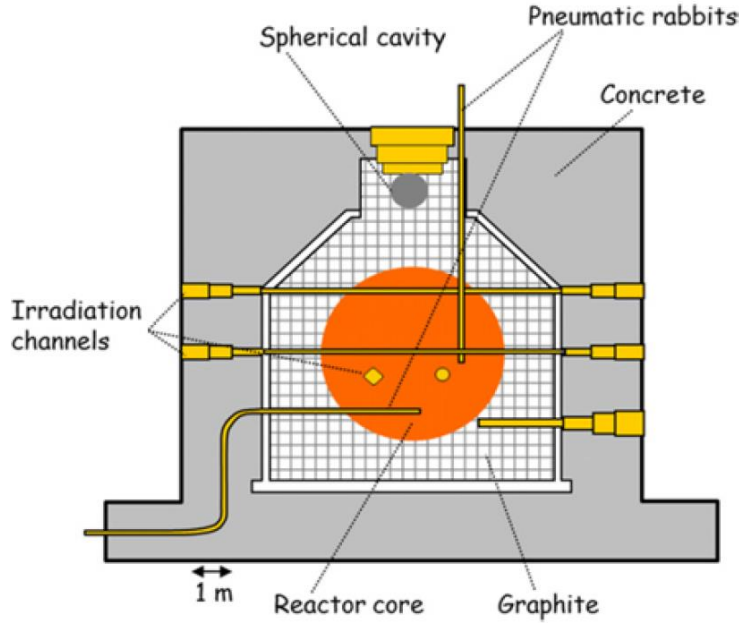


Figure 2. Schematic cross-section of the BR1 research reactor.

The neutron count rate was recorded for each scintillator, at the centre of the cavity and at different reactor power levels. The background count rate when the reactor was shut down was negligible for both scintillators. The measuring time was at least 10 minutes. The measured count rate at 1 kW nominal power was about 1200 cps with a dead time correction of about 2%. The dead time correction was estimated from the real and live time provided by the MCA527; assuming a non-paralyzable behaviour, we could estimate a dead time of about 16 μ s.

The expected dose rate at the centre of the cavity is in the order of 5 mGy/h at 1 kW nominal power [REF]. Based on the measured gamma-ray sensitivity and assuming a linear relation between dose rate and nominal power, the gamma-rays should not contribute to the measured response.

An example of the measured spectrum is shown in Fig. 3. A broad peak can be observed at about channel 100.

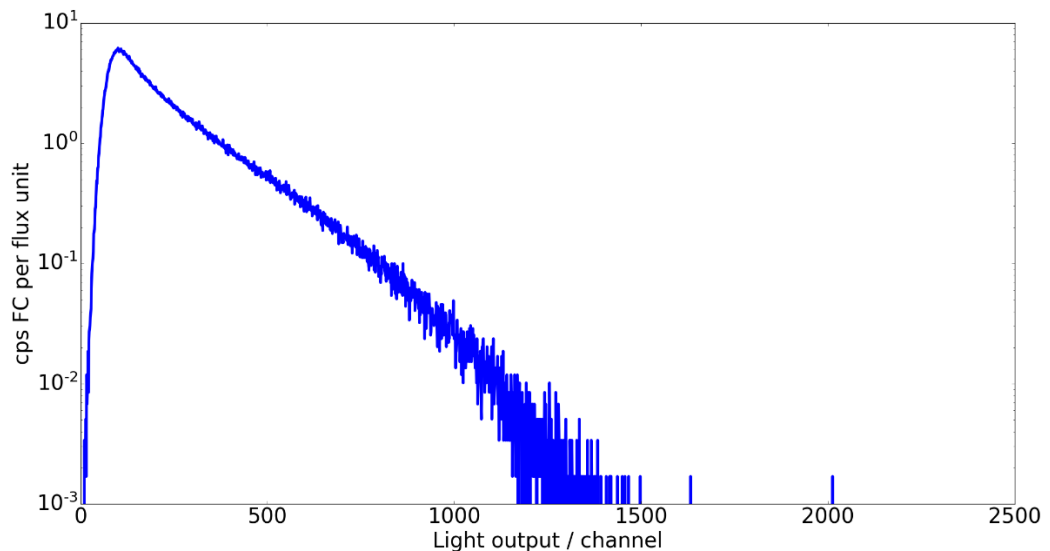


Figure 3. Example of a measured light output spectrum.

We carried out measurements in a range of nominal power between 0.1 and 30 kW for detector B and 0.1 and 3 kW for detector A. We observed that starting from a nominal power of 3 kW the shape of the spectrum starts to deviate from the one observed up to 1 kW.

The obtained sensitivity was calculated by dividing the background corrected count rate by the neutron fluence rate. The neutron fluence rate was determined from measurements with a calibrated fission chamber flux monitor [REF].

The ratio between the count rate from the neutron scintillators and the conventional thermal flux at the centre of the cavity represents the sensitivity of the scintillators.

The obtained sensitivities for the two detectors are shown in Fig. 4 when using a threshold at channel 10. Starting from 3 kW, the data for detector B deviate from the values at lower nominal power.

The measurements at 1 kW were repeated four times and exhibited a systematic uncertainty of 0.1% and 0.8% for detector A and B, respectively.

The neutron sensitivities for detectors A and B were obtained from an average value between 0.1 and 1 kW. These values are $1.25 \times 10^{-3} \text{ cm}^2$ and $1.19 \times 10^{-3} \text{ cm}^2$ with a standard deviation of 1.7% and 1.1% for detector A and B, respectively. A statistical component of 0.9% was calculated from the average of the statistical uncertainties for both detectors. The statistical uncertainty includes both the statistical uncertainty associated with the $^6\text{LiF}(\text{ZnS})$ detectors and the flux monitor but not the uncertainty of about 1% on the calibration factor for the flux monitor. A systematic bias of 1.5% and 0.7% for detector A and B, respectively, was then estimated by subtracting the statistical component from the observed standard deviation.

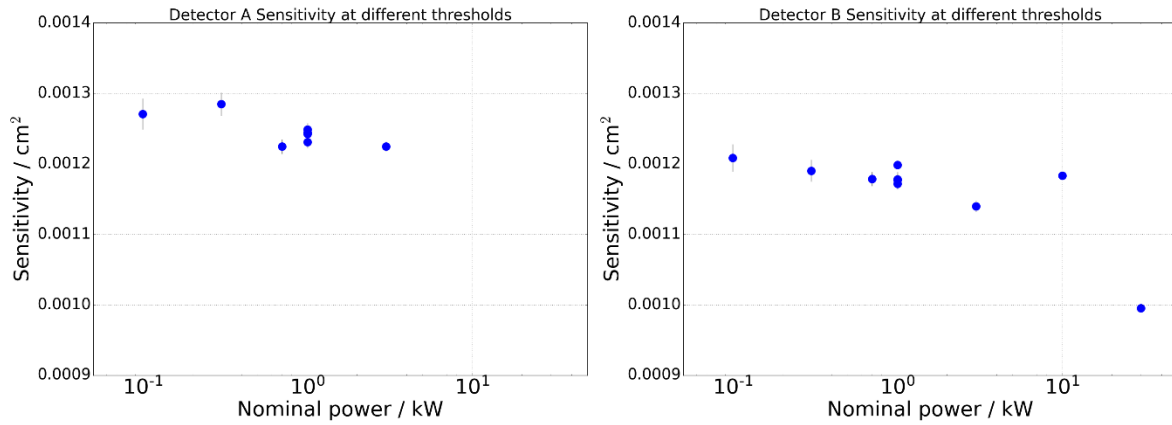


Figure 4. Neutron sensitivity at different thresholds for detector A and B.

3. SIMULATIONS

3.1. Monte Carlo model of the detector

The obtained sensitivities can be used as parameters to fine tune the Monte Carlo model of the detector. For this purpose, we determined the neutron sensitivity to a thermal flux with a Maxwell-Boltzmann distribution for different detector geometries.

We modelled the detector with MCNP [MCNP6.2] based on the information available and some assumptions. The crystal was modelled as right circular cylinder with the diameter equal to the height and a density of 3.46 g/cm^3 . The mass ratio between ZnS and LiF was 2:1. We assumed a 100% enrichment in ^6Li . The crystal was exposed to a flux of thermalized neutrons coming from the moderation of neutrons in water. The neutron sensitivity was obtained by comparing the reaction rate for the $^6\text{Li}(n,t)\alpha$ reaction in the crystal with the flux in absence of the crystal. We did not model all the processes leading to the observed signal such as light production and transport as well as the conversion to electron signal, but we implicitly assumed that each reaction corresponded to a detected signal. Due to the fact that the light may be absorbed or lost before reaching the PMT, the actual sensitivity should be lower than the obtained one.

The derived sensitivity as a function of the crystal radius is shown in Fig. 5 together with the theoretical sensitivity corresponding to an ideal point detector with the same mass of ^6Li . The theoretical limit was calculated with the methods given in [ANE2013] and using the value of 849 barn as thermal cross section for the (n,t) reaction on ^6Li .

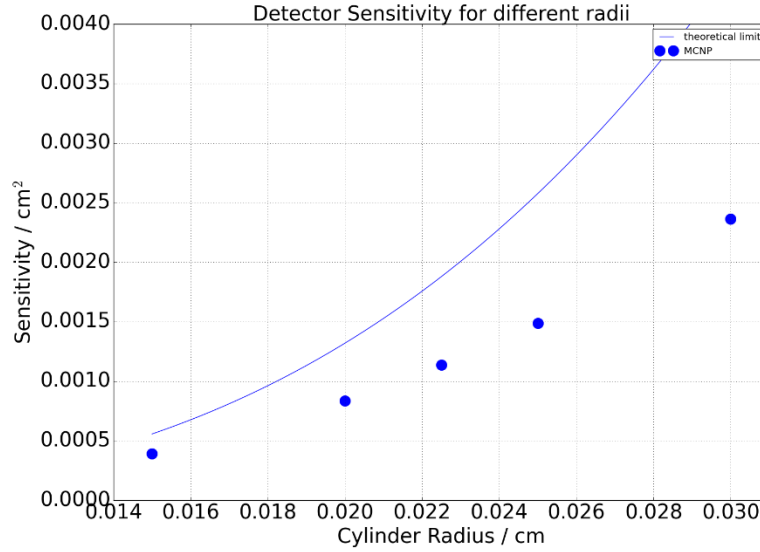


Figure 5. Simulated neutron sensitivity for cylinders as a function of the cylinder radius. The continuous line indicates the theoretical limit for the sensitivity in absence of neutron self-shielding.

Given the fact that we measured a sensitivity in the range of $1.1 \div 1.3 \times 10^{-3} \text{ cm}^2$, we took a cylinder with radius 0.225 mm as an equivalent cylinder for neutron transport simulations of a detector with a neutron sensitivity corresponding to the measured one. If we could account for the fact that not all of the (n,t) reactions result in a measurable signal and for the attenuation due to the presence of structural materials that surround the crystal, the value of the radius would be higher.

The detector response as a function of neutron energy is shown in Fig. 6. The detector response function was obtained from Monte Carlo simulations considering neutrons of different energy and with an isotropic directional distribution impinging on the crystal. The resonance at around 200 keV neutron energy in the (n,t) cross section for ^6Li can be observed.

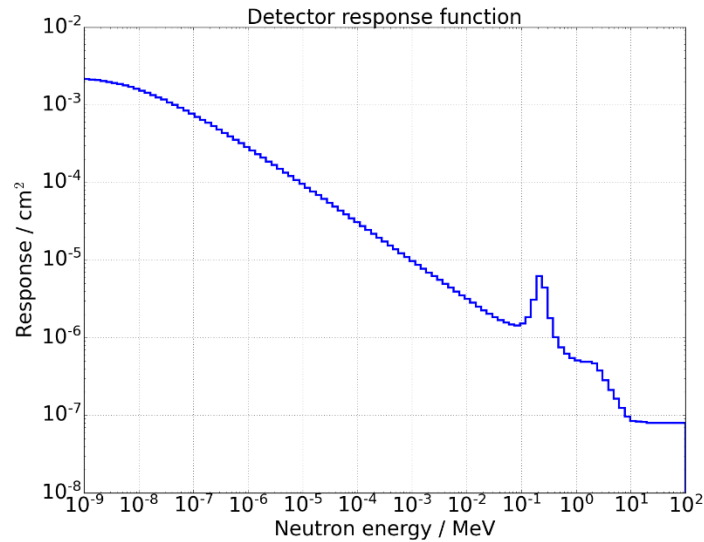


Figure 6. Detector response function as a function of neutron energy.

3.2. Spent Fuel estimate

The goal is to deploy such a detector for spent fuel measurements. Therefore, based on the obtained results, we estimate its suitability for such an application by estimating the expected neutron count rate as well as the impact of gamma radiation.

We considered a case with initial enrichment of 4.5%, burnup of 45 GWd/t_{HM} and cooling time of 5 years from the SCK CEN spent fuel library [SFL]. This case is representative of a typical case for Belgian nuclear power plants [DOEL]. The fuel was assumed to be stored under water with a boron content of 2500 ppm.

With MCNP [MCNP6.2] we modelled a 17×17 PWR LEU spent fuel assembly with a fibre detector in the instrumentation channel. The count rate was estimated from the flux tally with FM special treatment. With this treatment the fluence is multiplied by the cross section for the (n,t) reaction and the density in at/(b cm) of the considered cell. The obtained tally is per unit volume in cm³ [MCNP6.2] and therefore by multiplying it by the detector volume one obtains the counts per source particle. By further multiplying with the neutron emission (in this case 5.35×10^8 n/s t_{HM}) as explained in [OBS] we can estimate the neutron count rates being 10.0(1) cps. Assuming that a statistical precision of 0.3% would be required, we can estimate a measurement time of approximately 3 hours.

For the gamma radiation we estimated a dose rate of 500 Gy/h in the instrumentation channel.

4. CONCLUSIONS

We carried out measurements to determine the gamma-ray and neutron sensitivities of two ⁶LiF(ZnS) based miniature scintillators to be deployed for spent fuel verification. The detectors were insensitive to gamma-rays up to a dose rate of 70 Gy/h. Given the fact that in spent fuel measurement dose rates up to 1000 Gy/h are possible, additional measurements are required to study the response of the detectors above a dose rate of 200 Gy/h. We estimated a neutron sensitivity of 1.2×10^{-3} cm². A Monte Carlo model of the detector yielding the same neutron sensitivity was built. With the model a count rate of about 10 cps was estimated for a 17×17 PWR LEU spent fuel assembly with a burnup of 45 GWd/t_{HM}, 4.5 % initial enrichment and 5 years cooling time. To limit the measurement time several detectors could be deployed.

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