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Detection of fuel pins diversion with the self-indication neutron resonance densitometry technique

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Abstract

The verification of spent fuel assemblies is among the activities conducted during a safeguards inspection, and several non-destructive assay techniques are being developed to improve the accuracy of existing methods. Among other techniques, the self-indication neutron resonance densitometry (SINRD) relies on the passive neutron emission from the spent fuel assemblies. Previous research conducted at SCK•CEN found that the optimal configuration was obtained with the fuel kept in air and surrounded by a polyethylene slab.

The SINRD technique was proposed mainly for the direct quantification of the ^{239}Pu mass in spent fuel, whereas this contribution is focused on the potential to detect the diversion of fuel pins from a spent fuel assembly. First, the detector responses of several fission chambers placed in the guide tubes of a PWR 17x17 fuel assembly were calculated with the Monte Carlo code MCNPX. Different fissile material coatings (e.g. ^{239}Pu, ^{238}U) were taken into account to consider detectors mostly sensitive to thermal and fast neutrons. In addition, the response to ionization chambers was modelled for the detection of gamma-rays. Fuel assemblies with material compositions corresponding to different initial enrichment, burnup, and cooling time were modelled to evaluate the sensitivity of the detector responses to the fuel irradiation history.

The detector responses were calculated also for several diversion scenarios where fuel pins from a complete fuel assembly were replaced with dummies made of stainless steel. The diversions ranged from 15% to 50% of the total pins. The detector responses obtained from the diversion cases were compared to the values for the complete fuel assemblies to determine the capability of SINRD to detect the diversion of fuel pins. Promising results were obtained by combining the responses of the different detector types.

Keywords: SINRD, spent fuel, NDA, partial defect, Monte Carlo

1. Introduction

A technical objective of nuclear safeguards is to ensure the detection of the diversion of nuclear material from peaceful applications to the manufacture of nuclear weapons (IAEA, 1972).

Safeguards inspections are carried out by the International Atomic Energy Agency (IAEA) in the countries signatories of the Non-Proliferation Treaty (NPT) (IAEA, 1970). Since most of nuclear material placed under safeguards is in the form of spent fuel, the verification of this material is of major interest for the IAEA (IAEA, 2013).

However, the measurement of spent fuel presents many challenges due to its very high radiation emission and decay heat. Currently the spent fuel verification is performed with non-destructive assay (NDA) techniques such as the digital Cherenkov viewing device (DCVD) (Chen et al., 2003), (Chen et al., 2009), (Branger et al., 2014), the spent fuel attribute tester (SFAT) (Arit et al., 1995), (Honkamaa et al., 2003), and the Fork detector (Rinard et al., 1988), (Borella et al., 2011). In addition, many other NDA techniques are under development to improve the accuracy of the verification (Tobin et al., 2011).

This contribution is focused on the capabilities of the self-indication neutron resonance densitometry (SINRD) (Menlove et al., 1969) for the detection of diversion from a spent fuel assembly. The basic principle of SINRD is described with the Monte Carlo models used in the study. Then the overview of the simulations is given, considering both complete fuel assemblies and diversion scenarios, and the capabilities of SINRD for this application are discussed. Finally the conclusions are presented with an outlook on future research.

2. Description of the SINRD technique

The self-indication neutron resonance densitometry is a non-destructive assay technique that relies on the spontaneous neutron emission of spent nuclear fuel (LaFleur, 2011), (LaFleur et al., 2015), (Rossa et al., 2015), (Rossa, 2016).

The basic principle of SINRD is described in Figure 1. The total cross-section for neutron-induced reaction of ^{239}Pu is plotted with the transmission of a neutron flux through samples containing different percentages of ^{239}Pu. The transmission values were calculated with Monte Carlo simulations considering a sample of ^{239}Pu with density and dimensions equal to a PWR fuel pin. It is evident from the figure that the attenuation of the neutron flux is related to the amount of ^{239}Pu in the sample. The SINRD technique aims at measuring the attenuation of the neutron flux around the 0.3 eV region due to the presence of ^{239}Pu in the fuel assembly. The neutron detection

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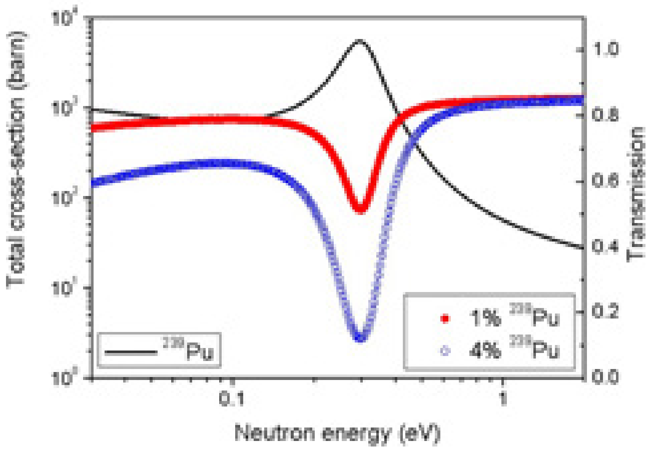


Figure 1: Total cross-section of ^{239}Pu and transmission of a neutron flux through samples containing ^{239}Pu. The cross-section values were obtained from the ENDF/B-VII.0 data library, whereas the transmission was calculated with Monte Carlo simulations.

in the 0.3 eV region is enhanced by using a fission chamber with ^{239}Pu as fissile material, according to the self-indication principle (Fröhner et al., 1966).

The neutron flux in the 0.3 eV energy region is estimated with SINRD by taking the difference between the neutron counts of two ^{239}Pu fission chambers. One detector is covered by a thin Gd filter, whereas the other detector is covered by a Cd filter. These materials were chosen because they have a cutoff in the neutron absorption below and above 0.3 eV, respectively.

In addition, the thermal neutron flux and fast neutron flux were estimated in this work by calculating the response of a bare ^{239}Pu fission chamber and ^{238}U fission chamber, respectively.

The approach for the study of the SINRD technique was extended in this paper by calculating the response of ionization chambers for the detection of gamma-rays. The multiple insertion of neutron and gamma-ray detectors in a fuel assembly was proposed for the PDET detector (Ham et al., 2009), (Ham et al., 2015), and can be beneficial also for the SINRD technique.

3. Model developed for the study

3.1 Monte Carlo model

The capability of SINRD for the detection of partial defects was investigated in this article with Monte Carlo simulations. The Monte Carlo code MCNPX v.2.7.0 (Pelowitz, 2011) was used to simulate a PWR 17x17 fuel assembly stored in air and surrounded by a 12-cm slab of polyethylene to ensure neutron moderation. The model of the fuel assembly is shown in Figure 1. The fuel geometry chosen for the simulation contains 264 fuel pins and 25 guide tubes. These are used for the insertion of control rods during the reactor operation and provide enough room for neutron or gamma-ray detectors once the fuel is discharged.

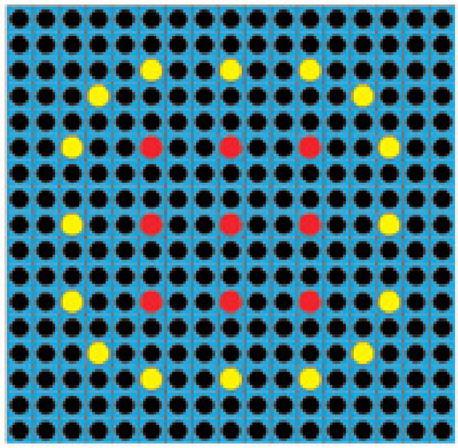


Figure 2: MCNPX model of the fuel assembly used for the study. The fuel pins are depicted in black, the peripheral guide tubes in yellow, and the central guide tubes in red.

The measurement setup chosen for this study can be representative of an encapsulation plant where spent fuel with long cooling time is verified before the final disposal (Park et al., 2014).

3.2 Calculation of the neutron detectors counts

The total counts of the neutron detectors (N_{bare}) were calculated with Formula (1) as the product between the coefficient C_N, the incoming neutron flux (φ_N) and the microscopic cross-section (σ_{DET}) of the active material in the detector itself. The coefficient C_N was calculated with Formula (2) as the product between the amount of fissile material in the detector (n_{fiss}), the total neutron emission from the spent fuel assembly (N_E), and the measurement time (t). The Photonis CFUE43 fission chamber (Photonis, 2017) was taken as reference design, but the active length was increased to 2 m to obtain a fissile material mass of 263.89 mg (Rossa, 2016). This choice was made to maximize the count rate, whereas the technical feasibility of such detector will be included in future work. The total neutron emission was taken from the reference spent fuel library (Rossa et al., 2013), whereas the measurement time was set to one hour.

The neutron flux (φ_N) and the microscopic cross-section (σ_{DET}) included in Formula (1) are a function of the incoming neutron energy E_N. The neutron flux was obtained from the MCNPX simulations and accounts also for the multiplication effect. The cross-section values were taken from the ENDF/B-VII.0 nuclear data library (Chadwick, 2006) and averaged over 600 logarithmically-interpolated energy bins between 10^{-9} and 20 MeV. The fission cross-sections of ^{239}Pu and ^{238}U were used to model detectors sensitive mainly to thermal and fast neutrons, respectively.

$$N_{bare} = C_N \int_{E_N} \varphi_N(E_N)\, \sigma_{DET}(E_N)\, dE_N \quad (1)$$

$$C_N = n_{fiss} N_E t \quad (2)$$

The presence of a thin Gd or Cd filter around the ^{239}Pu fission chamber was accounted for with Formula (3), where n_{fil} and σ_{fil} are the atom density and cross-section of the filter.

$$N_{fil} = C_N \int_{E_N} \varphi_N\left(E_N\right)\ \sigma_{DET}\left(E_N\right)\ e^{-n_{fil}\sigma_{fil}(E_N)}\ dE_N \qquad (3)$$

The neutron counts in the energy region close to 0.3 eV were calculated as the difference between the counts of two fission chambers, one covered by a Gd filter and one covered by a Cd filter.

The uncertainty of the neutron counts for the different detectors was estimated as the square root of the corresponding neutron count.

3.3 Calculation of the gamma-ray detectors response

The gamma-ray detector response (P) was calculated with Formula (4) as the product between the coefficient C_P, the gamma-ray flux (φ_P) and the response function of the detector (f_{DET}). The coefficient C_P is the product between the total photon emission from the spent fuel assembly and the measurement time. The total photon emission was taken from (Rossa et al., 2013) and the measurement time was set to one hour as for the neutron measurements.

The photon flux (φ_P) and response function (f_{DET}) were obtained from MCNPX simulations and are function of the incoming gamma-ray energy E_γ. The photon flux was calculated in the guide tubes with the model of the spent fuel described in Section 3.1, whereas the response function was obtained by modelling the detector alone as an aluminum cylinder filled with nitrogen. The transport of both photons and electrons was simulated to obtain the response function (f_{DET}), which was calculated as the energy deposition tally (F6 type) in the gas-filled cavity. The energy range of the source term was divided into 23 bins from 50 keV to 5 MeV, and separate simulations were performed defining the source with a uniform histogram distribution over a single energy bin.

$$P = C_P \int_{E_\gamma} \varphi_P\left(E_\gamma\right)\ f_{DET}\left(E_\gamma\right)\ dE_\gamma \qquad (4)$$

The statistical uncertainty of the gamma-ray detector response was neglected since ionization chambers are normally operated in current mode and reach a stable signal well within the considered measurement time.

4. Overview of the performed simulations

4.1 Complete fuel assemblies

The simulations performed for this study considered both complete fuel assemblies and assemblies with diverted pins. In the case of a complete fuel assembly the fuel pins are identical in material composition and source strength, and these characteristics were taken from the reference spent fuel library (Rossa et al., 2013), (Borella et al., 2015). The sensitivity of the detector responses to the fuel irradiation history was evaluated by considering fuel assemblies with material composition and source strength corresponding to:

- initial enrichments: 2.0, 2.5, 3.0, 3.5, 4.0, 4.5, and 5.0%;
- burnup: 5, 10, 15, 20, 30, 40, and 60 GWd/$t_{HM.}$

The range of initial enrichment and burnup was chosen to represent the majority of operating conditions of current PWR reactors and it is in line with previous research (Trellue et al., 2010), (Borella et al., 2015).

4.2 Diversion scenarios

In the diversion cases the fuel pins were replaced by dummies made of stainless steel with the same dimensions of a fuel pin. The diversion scenarios are shown in Figure 3 and the replaced pins were between 50% and 15% of the fuel pins in a fuel assembly. The diversion scenarios were symmetrical since it resulted from previous work as the most challenging pattern to detect (Sitaraman et al., 2009), (Rossa, 2016). Nevertheless, it is worth noting that dummy pins placed in the outer section of the assembly may be easy to detect by visual inspection due to the optical alteration of the spent fuel pins through irradiation.

For the simulations with the diversion scenarios the fuel pins had a material composition and source strength corresponding to fuel with the following:

- 2% initial enrichment and 30 GWd/t_{HM} burnup;
- 3.5% initial enrichment and 10, 30, or 60 GWd/t_{HM} burnup;
- 5% initial enrichment and 30 GWd/t_{HM} burnup.

In all simulations included in this contribution the fuel pins had a cooling time of 5 years.

5. Results

5.1 Complete fuel assemblies

For each simulation in this study the detectors responses calculated according to the approach described in Sections 3.2 and 3.3 were normalized to the value obtained in the central guide tube. In addition, the guide tubes were divided for this study into 16 peripheral and 9 central guide tubes depending on the geometrical location in the fuel assembly. The two groups are identified in Figure 1 by different colors. The average detector responses were calculated in the two guide tubes groups.

The average detector responses obtained in the cases with complete fuel assemblies were used to establish a reference band associated to each type of detector response (i.e. thermal neutrons, resonance region neutrons,

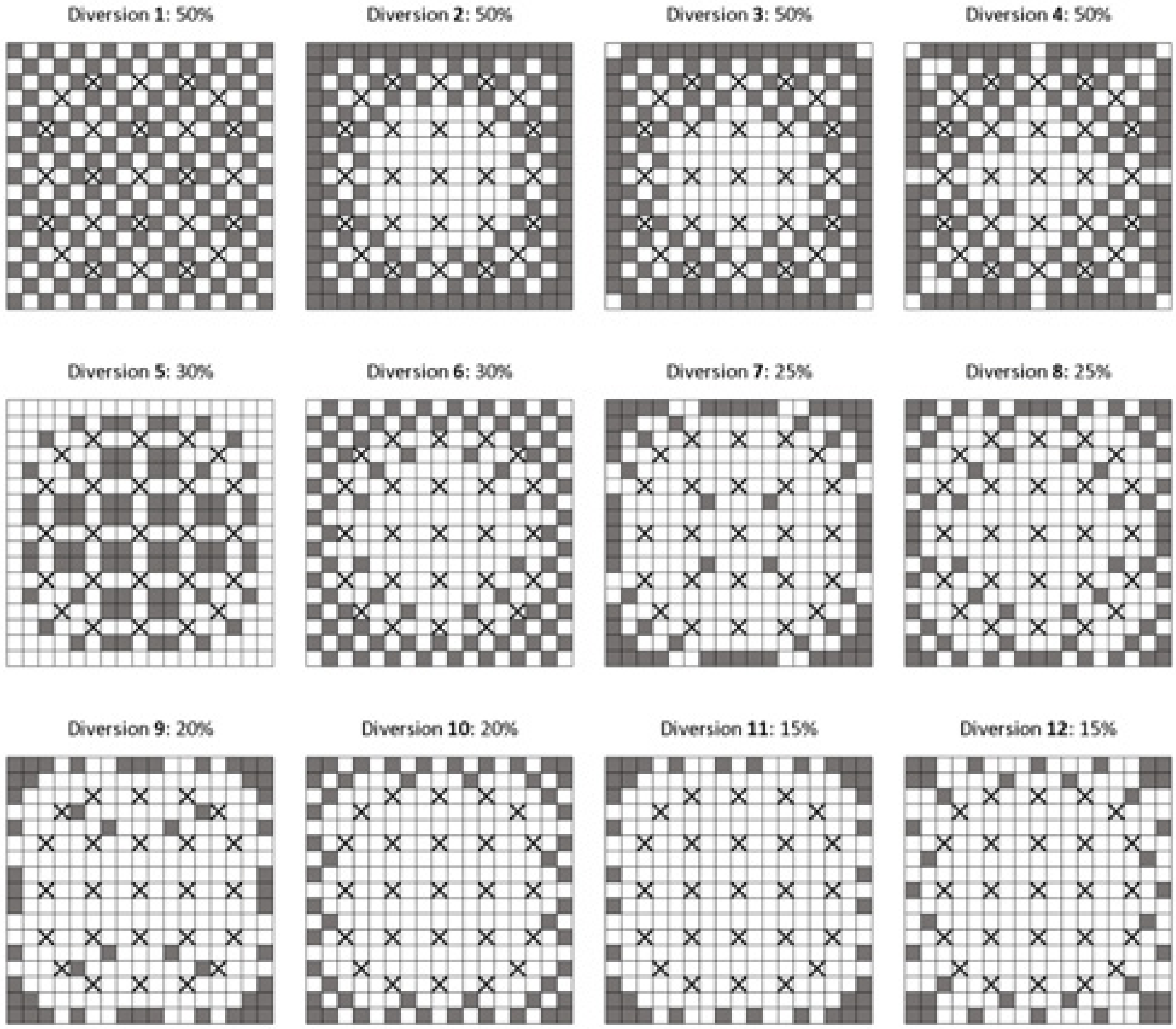


Figure 3: Overview of the diversion scenarios. The fuel pins are depicted in white, the dummy pins in grey, and the guide tubes with crosses.

fast neutrons, gamma-rays). The low and high boundaries are reported in Table 1 for the nine central guide tubes and for the sixteen peripheral guide tubes. In order to obtain the low boundaries for the neutron detectors in Table 1, the minimum detector responses obtained in the whole set of complete fuel assemblies were further decreased by the 1-σ value to account for uncertainty. Similarly the high boundaries were obtained by increasing by 1-σ the maximum values obtained for each detector type. The boundaries for the gamma-ray detector were taken as the minimum and maximum detector responses obtained in the whole set of complete fuel assemblies.

Both boundaries for thermal and resonance region neutrons are lower for the central guide tubes compared to the peripheral guide tubes, whereas the opposite occurs for fast neutrons and gamma-rays. In general the width of the reference band is larger for peripheral guide tubes compared to central guide tubes, and it is significantly larger for neutron than for gamma-ray detectors.

	Central guide tubes		Peripheral guide tubes	
	Low boundary	**High boundary**	**Low boundary**	**High boundary**
Thermal neutrons	1.153	1.263	1.695	2.298
Resonance region neutrons	1.122	1.271	1.576	2.290
Fast neutrons	0.862	1.090	0.802	0.977
Gamma-rays	0.985	0.987	0.904	0.912

Table 1: Normalized detector responses calculated for the complete fuel assemblies for thermal neutrons, neutrons around the 0.3 eV region, fast neutrons, and gamma-rays. The low and high boundaries are given for the nine central guide tubes and the sixteen peripheral guide tubes.

5.2 Diversion scenarios

The average detector responses obtained for the diversion scenarios were compared to the reference bands shown in Table 1, and the values that fell outside these bands signaled possible diversion cases. Figures 4-7 show the normalized detector responses calculated for the diversion scenarios for the different detector types. The results are the average values for the nine central guide tubes and for the sixteen peripheral guide tubes.

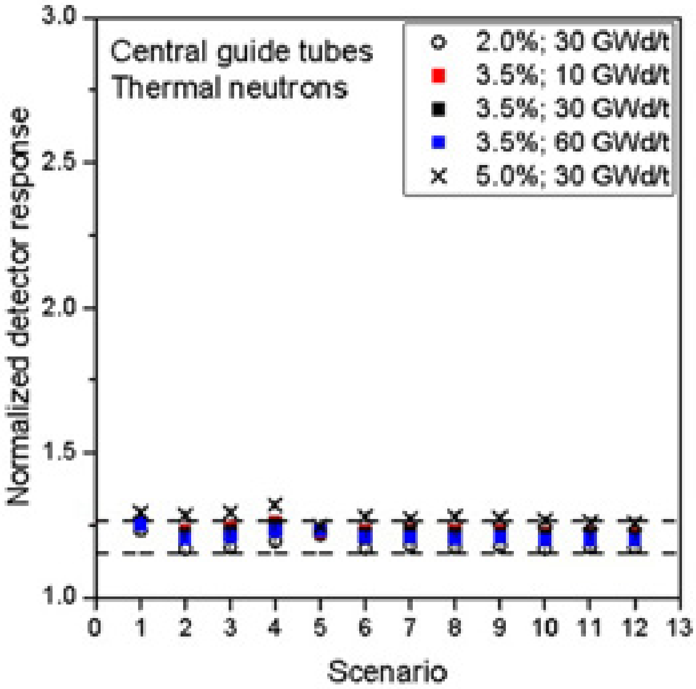
The results for the thermal neutron detectors show that most of the diversion cases fall within the reference band of the complete fuel assemblies. Only for diversion with fuel with 5% initial enrichment the detector responses are above the high boundaries both for central and peripheral guide tubes.

Considering the detectors measuring neutrons around the 0.3 eV resonance region (Figure 5), most of the diversions with fuel assemblies with initial enrichment of 5% fall outside the reference band. In addition, also some scenarios with 50% of replaced pins from fuel with 3.5% initial enrichment and burnup larger than 30 GWd/t have values above the reference band.

The results for the fast neutron detectors show that for all diversion scenarios the average values for the central guide tubes are within the reference band. The average detector responses for the peripheral guide tubes are lower than the reference band for some scenarios with 50% and 30% of replaced pins. In all cases there is not a significant difference due to the fuel irradiation history.

Figure 7 shows that for all diversion scenarios the average responses of the gamma-ray detectors are outside the reference band of the complete fuel assemblies. The peripheral guide tubes are the most affected by the replacement of fuel pins. As in the case of fast neutron detectors, the irradiation history of the fuel assembly does not influence significantly the results. By comparing the different detector types, the gamma-ray detectors show a larger variation due to the fuel pins diversion compared to the neutron detectors.

The reference bands reported in the figures in this section were calculated for fuel assemblies with uniform composition. Variation of burnup among pins due to core loading strategies might have a noticeable impact on the bands and needs to be investigated.



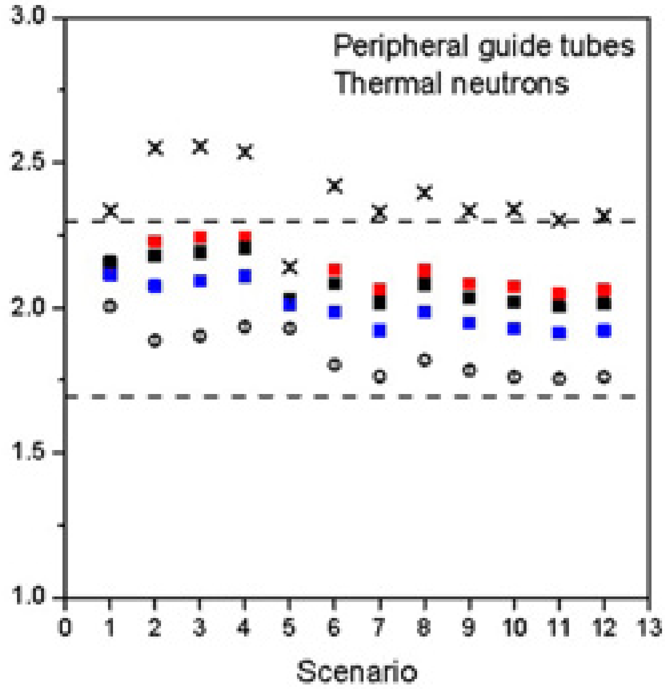


Figure 4: Normalized detector responses for thermal neutrons in the different diversion scenarios. The average value for central guide tubes (left), and peripheral guide tubes (right) are shown. The lower and upper boundaries for the cases with complete fuel assemblies are also reported.

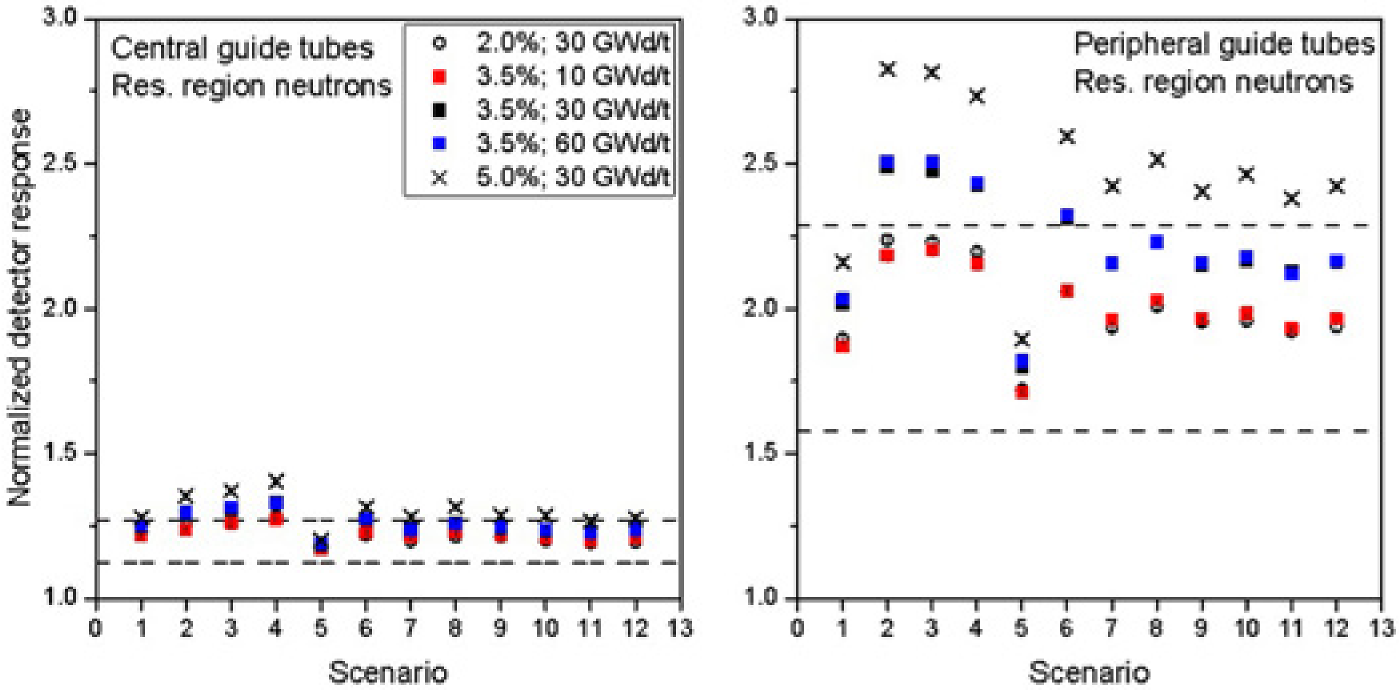


Figure 5: Normalized detector responses for neutrons around the 0.3 eV resonance region in the different diversion scenarios. The average value for central guide tubes (left), and peripheral guide tubes (right) are shown. The lower and upper boundaries for the cases with complete fuel assemblies are also reported.

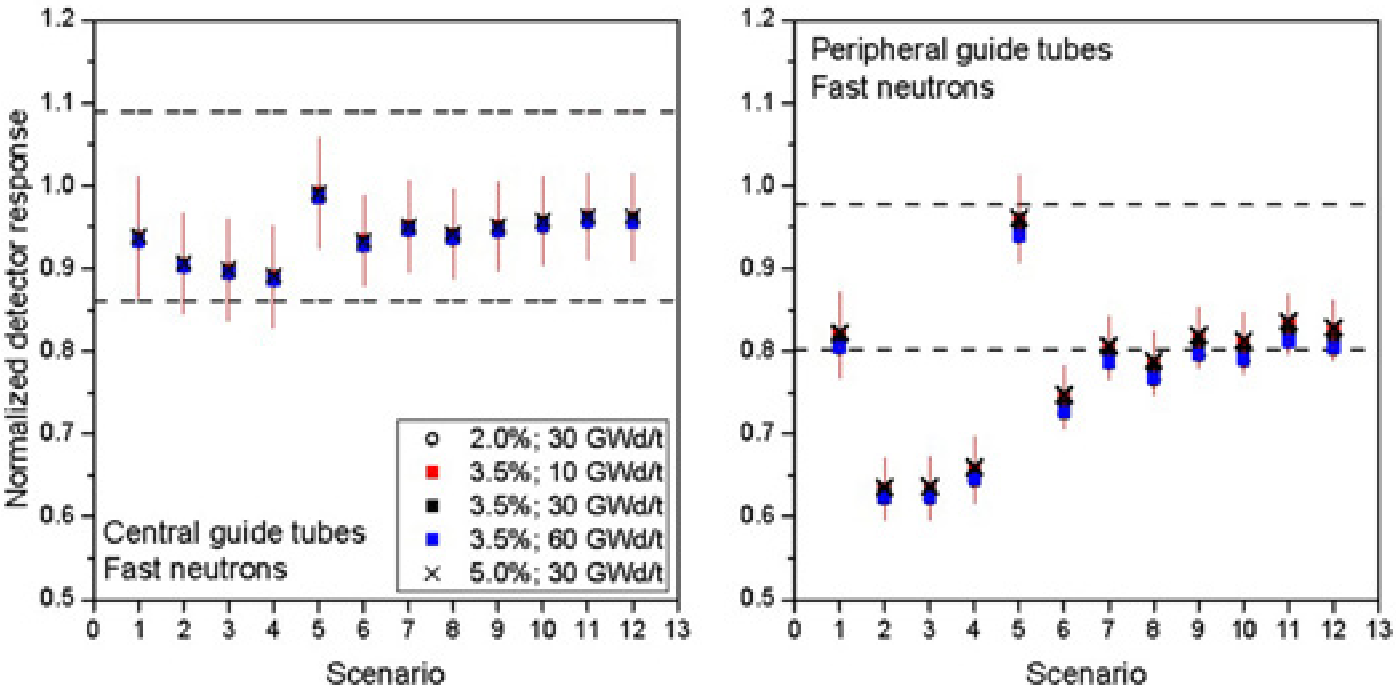


Figure 6: Normalized detector responses for fast neutrons in the different diversion scenarios. The average value for central guide tubes (left), and peripheral guide tubes (right) are shown. The lower and upper boundaries for the cases with complete fuel assemblies are also reported.

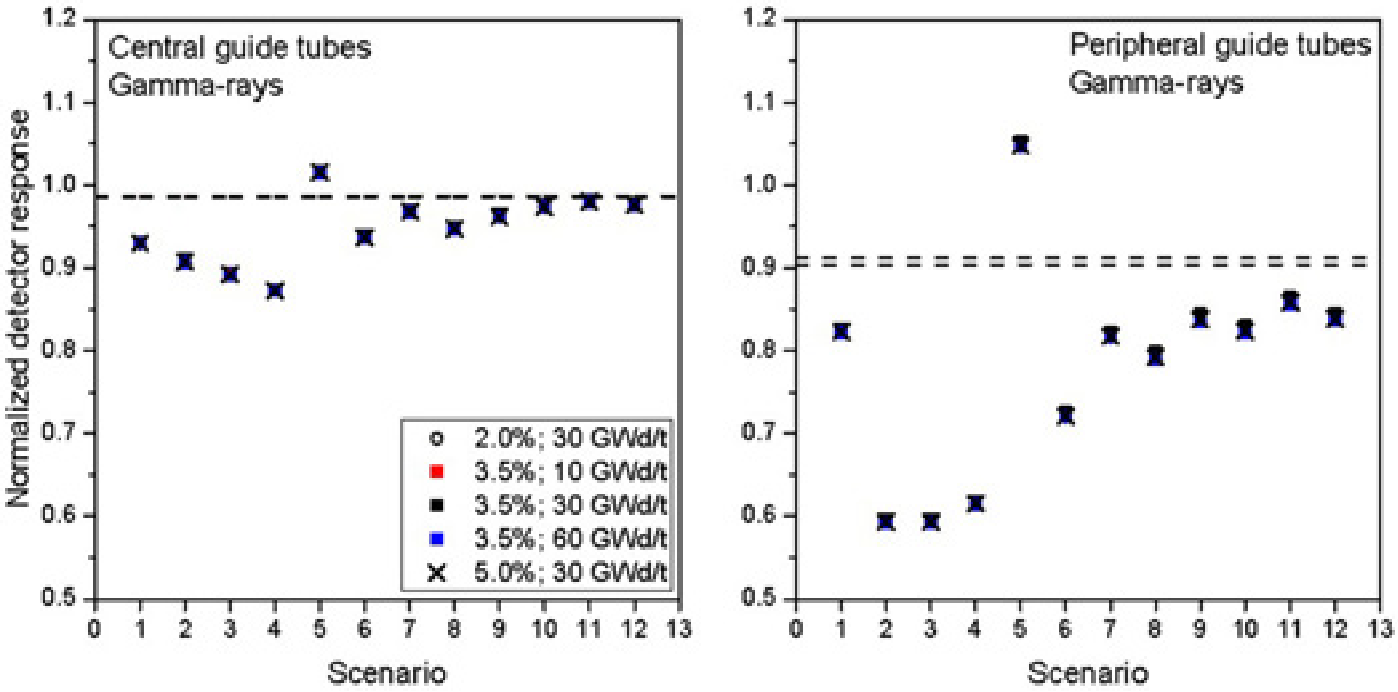


Figure 7: Normalized detector responses for gamma-rays in the different diversion scenarios. The average value for central guide tubes (left), and peripheral guide tubes (right) are shown. The lower and upper boundaries for the cases with complete fuel assemblies are also reported.

6. Conclusions

The capabilities of SINRD to detect the substitution of metal pins for fuel pins from a complete assembly were investigated in this paper. The neutron and gamma-ray fluxes were estimated in the guide tubes of a PWR fuel assembly to mimic the use of multiple detectors during the measurements. The response of detectors sensitive mainly to thermal neutrons, neutrons with energy around 0.3 eV, fast neutrons, and gamma-rays were considered.

A first series of simulations concerned complete fuel assemblies with different irradiation histories to estimate the influence of initial enrichment, burnup, and cooling time on the detector responses. The results from the complete fuel assemblies were used to identify a reference band of values obtained for complete fuel assemblies. The values calculated for the diversion scenarios were then compared with the reference bands of each detector type, and the values that fell outside the reference bands were an indication of diversion.

The peripheral guide tubes in almost all scenarios were the most affected by the fuel pins diversion and were outside the reference bands for multiple detector types. For all diversion scenarios the average gamma-ray detector response for the guide tubes was outside the reference band, whereas for some diversion cases with 50% and 30% of replaced pins also the values for neutron detectors were outside the reference bands. Overall the gamma-ray detectors showed a larger change due to the diversion of pins compared to the neutron detectors.

Future work will continue the assessment of the SINRD technique for the detection of fuel pins diversion by considering additional diversion scenarios and other approaches to the data analysis. The comparison among different NDA techniques for the same diversion scenarios is also foreseen.

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