Nuclear Engineering and Technology 52 (2020) 1022–1028

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Conceptual design of neutron measurement system for input accountancy in pyroprocessing

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ARTICLE INFO

Article history:
Received 14 November 2018
Received in revised form
4 November 2019
Accepted 5 November 2019
Available online 6 November 2019

Keywords:
Nondestructive assay
Nuclear safeguards
Nuclear material accountancy
Spent-fuel assembly

ABSTRACT

One of the possible options for spent-fuel management in Korea is pyroprocessing, which is a process for electrochemical recycling of spent nuclear fuel. Nuclear material accountancy is considered to be a safeguards measure of fundamental importance, for the purposes of which, the amount of nuclear material in the input and output materials should be measured as accurately as possible by means of chemical analysis and/or non-destructive assay. In the present study, a neutron measurement system based on the fast-neutron energy multiplication (FNEM) and passive neutron albedo reactivity (PNAR) techniques was designed for nuclear material accountancy of a spent-fuel assembly (i.e., the input accountancy of a pyroprocessing facility). Various parameters including inter-detector distance, source-to-detector distance, neutron-reflector material, the structure of a cadmium sleeve around the close detectors, and an air cavity in the moderator were investigated by MCNP6 Monte Carlo simulations in order to maximize its performance. Then, the detector responses with the optimized geometry were estimated for the fresh-fuel assemblies with different ^{235}U enrichments and a spent-fuel assembly. It was found that the measurement technique investigated here has the potential to measure changes in neutron multiplication and, in turn, amount of fissile material.

1. Introduction

As of the end of 2016, a total of ~7100 tU of spent fuel from commercial pressurized water reactors (PWRs) are being temporarily stored in pools at four nuclear-power-plant sites in Korea [1]. Two considered options for spent-fuel management in Korea are direct disposal to a geological repository or recycling of useful resources by pyroprocessing incorporated with a sodium-cooled fast reactor (SFR). The advantages of recycling are (1) burning of long-lived isotopes in a SFR and (2) separation of the high-heat elements from the others, which enables significant reduction of the volume of high-level radioactive waste [2–6].

Because a pyroprocessing facility handles spent nuclear fuels, various safeguards technologies, which enable us to detect diversion of significant quantities of nuclear material in a timely manner, to deter such diversion by the risk of early detection, and to declare the amount of nuclear material as accurately as possible, should be developed. According to the IAEA [7], nuclear material accountancy (NMA) is defined as a safeguards measure of fundamental importance. Although various techniques for NMA have already been developed in the more than four decades of relevant history, they are mostly for the purposes of aqueous reprocessing. There are many significant differences between aqueous reprocessing and pyroprocessing in terms of process type, condition, and material composition of product materials; indeed, no IAEA safeguards technology or criteria for pyroprocessing have yet been firmly established [8].

Destructive analysis (DA) can provide the most precise measurement of the amount of nuclear material in a sample; however, there are two concerns: (1) the representativeness of the sample, especially for a pyroprocessing facility (due to the lack of an input-accountancy tank), and (2) the significant time required for analysis or additional process (resulting in limited facility throughput). The non-destructive assay (NDA) technique has the potential to address these concerns. For example, because nuclear materials emit useful neutron signatures that penetrate bulk samples and container

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https://doi.org/10.1016/j.net.2019.11.007

walls, passive neutrons have the potential to measure large and high-density samples with less concern for sample homogeneity.

In two of our previous studies [9,10], a new, total-neutron-measurement-based hybrid concept for NMA of the final product in pyroprocessing (i.e., U/TRU ingots) was proposed and tested. NMA of input material should be considered to be as important as that of output materials. Additionally, accurate NMA of input material can help to determine shipper-receiver difference (SRD) more accurately. Note that a spent-fuel assembly has a more complex isotopic composition and less neutron multiplication than a U/TRU ingot, which difference can result in a different detector response.

At an aqueous reprocessing facility, the spent-fuel assembly is chopped and dissolved in acid at an input accountancy tank (IAT). Then, solution samples are taken for DA to establish input accountancy. However, there is no such IAT in a pyroprocessing facility; hence, several options for measurement point in a head-end area could be considered in order to declare the amount of input material: spent fuel assembly, extracted rods, mechanically declad fragments, or homogenized powder after oxidation process on the chopped rod cut. If one can measure the amount of nuclear material in a spent fuel assembly by NDA method, it would be beneficial in terms of the reduced number of process steps by eliminating a mixing process for homogenization and a pelletizing process and the reduced time for head-end process resulting in increased facility throughput. One of the possible strategies for input accountancy could be as follows: the amount of nuclear material introduced into a pyroprocessing facility could be determined by measuring a spent fuel assembly with NDA method if it can provide a reasonably low uncertainty. The amount of nuclear material going into the main process of pyroprocessing (i.e., oxide-reduction process) could be determined by a representative sampling method on fragments and DA on the samples. The oxidation process on the cladding hull could minimize the amount of residual material and the recovered material is also going to the main process. A decladding process should be designed to minimize the amount of holdup and an appropriate method needs to be applied for holdup measurement.

Many attempts to develop NDA techniques for assaying nuclear materials in a fresh- and a spent-fuel assembly have been made [11–13]. Especially, various types of neutron measurement systems have been designed, built, and tested on actual spent fuel assemblies through the Next Generation Safeguards Initiative (NGSI) project [14]. For example, a Passive Neutron Albedo Reactivity (PNAR) [15] instrument was tested in Japan. A Californium Interrogation Prompt Neutron (CIPN) [16] and Self-Interrogation Neutron Resonance Densitometry (SINRD) [17] instruments were tested in Korea. In addition, a Differential Die-away Self-Interrogation (DDSI) [18] and Differential Die-Away (DDA) [19] instruments were tested in Sweden. These systems are all in the development/demonstration stage and there is, at the moment, no consensus on which one is the most promising technique to quantify the plutonium content in a spent fuel assembly. Combining two or more techniques might be the feasible solution [20]. In the present study, a neutron measurement system, as one of the possible options, was designed for input material accountancy. To this end, the effect of various parameters was evaluated, and the detector response was estimated, for fresh- and spent-fuel assemblies by MCNP6 Monte Carlo simulations [21]. The aims of this study were limited to (1) understanding the detector response depending on the detection geometry and (2) evaluating the feasibility of a neutron measurement technique to quantify the fissile material content in a nuclear fuel assembly. The engineered design needs to be done in order to develop and deploy a practical system and it is not the scope of this study.

2. Hybrid concept for plutonium accountancy

The proposed hybrid concept is based on the total neutron measurement technique with fast-neutron energy multiplication (FNEM) and passive neutron albedo reactivity (PNAR) methods. This is a kind of passive neutron self-interrogation method according to which the FNEM and PNAR methods use the relationship between the amount of fissile material and neutron multiplication. The difference between the two methods is the energy of neutrons that induce fission events in the sample to be measured. That is, the FNEM and PNAR methods measure a signature related to the degree of induced fission rates by fast and thermal neutrons, respectively. In the FNEM method, a two-ring structure of neutron detectors is applied to measure the change in average neutron energy, which has a correlation with the degree of induced fission rates by fast neutrons and, in turn, the total amount of fissile material. On the other hand, in the PNAR method, the removable cadmium (Cd) structure around the sample is applied to measure the change in neutron populations between the cases with and without Cd. This change is correlated with the degree of induced fission rates by thermal neutrons and, thus, with the total amount of fissile material [22]. The hybrid concept combines two methods, FNEM and PNAR, to enhance the signature in the measurement of the amount of fissile material. The present system has the advantage that the detection system uses totals neutrons rather than coincidence neutrons, so the detectors can have less intrinsic efficiency than is required for the neutron coincidence systems. Also, the interrogation neutrons come from the spent-fuel assembly and no external neutron source is required (i.e., a self-interrogation technique). The PNAR systems have these intrinsic advantages; however, the present hybrid concept of FNEM × PNAR introduces a new physics signature based on fast neutron multiplication that compliments the thermal-neutron based signatures and provides a larger signature ratio. This is a concept paper, and many of the details that would be required for implementation would be expected in future papers.

Fig. 1 shows the initial detector design with the hybrid concept described above for measurement of the amount of fissile material in a spent-fuel assembly. For the FNEM measurement, two ^{3}He detectors of 2.54 cm (D) × 22 cm (H) dimensions with 4 atm ^{3}He filling pressure were positioned close to and far from the sample, respectively, so that the count rate ratio between the far and close detectors (i.e., ring ratio) can be measured. For the PNAR measurement, removable Cd was positioned between the fuel assembly and neutron moderator system so that the Cd ratio, which is the count-rate ratio between cases with and without Cd, can be measured. Each FNEM and PNAR signal was normalized to the unity for the no-fissile-material case for which the ^{235}U was replaced with ^{238}U. The FNEM, PNAR, and hybrid signal of FNEM × PNAR were calculated using the following equations:

$$\text{FNEM signal} = \frac{\textit{Total count of far detector without Cd}}{\textit{Total count of close detector without Cd}} \tag{1}$$

$$\text{PNAR signal} = \frac{\textit{Total count of far detector without Cd}}{\textit{Total count of close detector with Cd}} \tag{2}$$

$$\text{FNEM} \times \text{PNAR signal} = \text{normalized FNEM signal} \times \text{normalized PNAR signal.} \tag{3}$$

The measurement precision (i.e., fractional uncertainty) for the far or close detectors was assumed to be $\sigma_{Far,\ Close} = 1/\sqrt{Singles\ counts}$. The FNEM and PNAR signals were calculated by Eqs. (1) and (2) with error propagation of

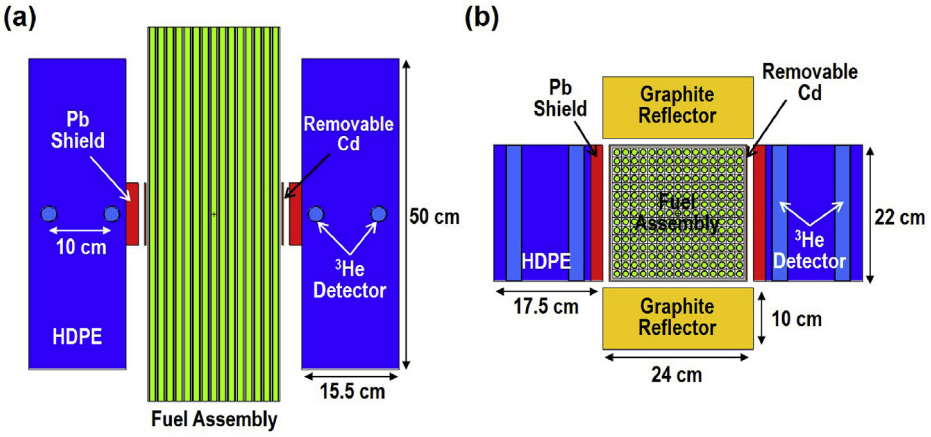

Fig. 1. Schematic of initial hybrid-concept detector design for measurement of amount of fissile material in spent-fuel assembly: (a) side view and (b) top view.

$\sigma_{FNEM,\,PNAR} = \sqrt{\sigma_{Far}^2 + \sigma_{Close}^2}$. Then, the FNEM and PNAR signals were normalized to the unity for each no-fissile-material case with error propagation of $\sigma_{normFNEM,\,normPNAR} = \sqrt{\sigma_{with\ fissile}^2 + \sigma_{without\ fissile}^2}$. Finally, the FNEM × PNAR signal was calculated from the normalized FNEM signal multiplied by the normalized PNAR signal (i.e., Eq. (3)) with error propagation of $\sigma_{FNEM\times PNAR} = \sqrt{\sigma_{normFNEM}^2 + \sigma_{normPNAR}^2}$.

In order to shield the intense gamma-rays from fission products, especially ^{137}Cs and ^{154}Eu, a 2-cm-thick lead shield, which can reduce the intensity of 662-keV gamma-rays from ^{137}Cs by ~92%, was applied. Two sides of the assembly have detector pods to measure the signature, and the other two sides have graphite reflectors to increase the induced fission reactions through neutron reflection. The thickness of the reflectors is 10 cm. In order to shield the detectors from neutrons coming from the upper and lower parts of the assembly, the height of the high-density polyethylene (HDPE) moderator was extended to 50 cm, and the removable Cd, reflectors, and gamma-ray shields are of 10 cm height. The overall weight of the detection system would be ~53 kg with the current conceptual design. The 50-cm-high HDPE moderator and the Pb shield weighs ~33 kg and ~10 kg, respectively. The weight of the system could be reduced by an engineered design; that is, the height of the HDPE moderator can be reduced by using a combination of the HDPE and a Cd layer because the role of the upper and lower parts of the HDPE in the current design is to shield the background neutrons. However, a detector housing and a moving mechanism for the removable Cd should be added for practical use which results in increasing the overall weight. The removable Cd could be designed to be inserted and removed with guide attachments to the detector body. Another factor that needs to be addressed by an engineered design is a relatively low melting temperature of the HDPE moderator. Possible solutions would be (1) applying a high-melting-temperature polymer (e.g., PEEK having a melting temperature of 343 °C [23]) on a surface layer of the HDPE, (2) using a fan to circulate air, and (3) limiting measurement time to a few minutes.

It was assumed that the fuel assembly has 15 × 15 rods with zirconium cladding, a pellet density of 10.48 g/cm^3, and a height of 60 cm. Although the full length of a fuel assembly is about 400 cm, it was reduced to 60 cm in our simulation model in order to reduce the MCNP calculation time. In the simulation, the primary neutrons were sampled using a spontaneous fission (SF) capability, i.e., PAR=SF on the SDEF card in the MCNP6 code. The detection probability of each ^{3}He detector was calculated using the pulse height tally F8 with a coincidence capture (CAP) treatment. The time gate keyword of GATE did not used because it is a kind of the total neutron counting system.

Initially, a fresh PWR fuel assembly with 5 wt% ^{235}U enrichment was simulated rather than considering spent-fuel material compositions, in order to evaluate the feasibility of this measurement concept without the complexities of spent-fuel assemblies' material compositions. The 5 wt% enrichment was chosen to provide good statistics for the comparison of the variables and it is the maximum fissile content in commercial fresh fuel assemblies. The typical fissile content in spent fuel would be 1–2% and the detector response could be estimated by interpolating from the response curve. It was necessary to limit this conceptual design study to the simple case of fresh fuel so that the detection geometry could be optimized and the signature magnitude could be studied. The considered parameters were inter-detector distance, source-to-detector distance, neutron-reflector material, the structure of the Cd sleeve around the close detectors, and the air cavity within the moderator. The FNEM and PNAR signals were evaluated depending on the parameters listed above and the optimized design was determined when the FNEM × PNAR signal was maximized. After evaluating the effects of various parameters on the FNEM and PNAR signals, the spent-fuel material composition was applied to estimate the detector response with the optimized design.

3. Effects of various parameters

3.1. Inter-detector distance (IDD)

The effect of the distance between the close and far detectors, which is to say the inter-detector distance (IDD), was examined first. To this end, the IDD was changed from 5 to 20 cm at 5-cm intervals, as shown in Fig. 2. The other parameters, including source-to-detector distance, neutron-reflector material, the structure of the Cd sleeve around the close detectors, and moderator structure were fixed as an initial design. The singles counts were estimated for 100 h measurement due to the low neutron emission rate of fresh fuel (i.e., ~1 × 10^3 neutrons/s). Note that the neutron emission rate of spent fuel should be higher than that of fresh fuel by more than 4 orders of magnitude; hence, the same measurement precision can be achieved by a few tens of seconds of measurement. Fig. 3 shows that the FNEM, PNAR, and FNEM × PNAR signals increased with increasing IDD; however, the degree of improvement was reduced. Obviously, with increasing IDD, the

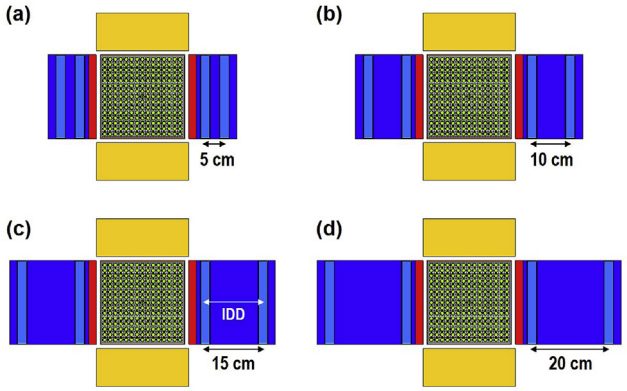

Fig. 2. Simulation geometries for different inter-detector distances: (a) 5, (b) 10, (c) 15, and (d) 20 cm.

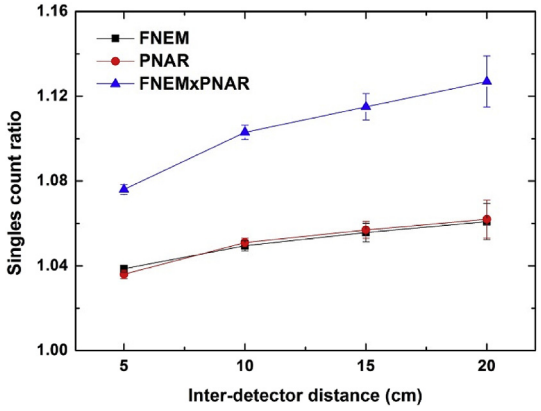

Fig. 3. FNEM, PNAR, and FNEM × PNAR signals as functions of inter-detector distance.

detection efficiency of the far detector will be decreased, resulting in a longer measurement time required for the same measurement precision. Interestingly, the FNEM and PNAR signals were determined to be about the same for each case and it is considered to be caused by chance. The optimal IDD was determined to be 10 cm.

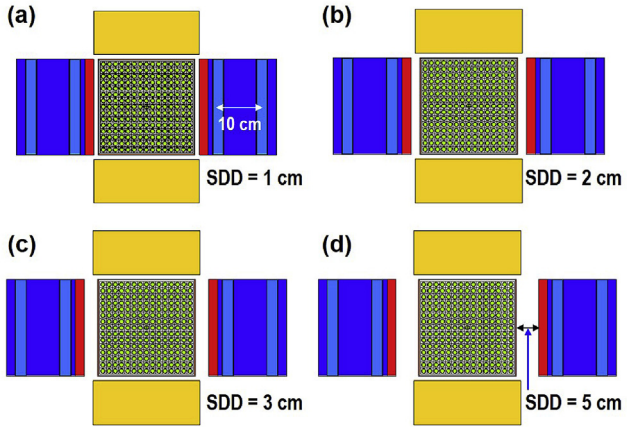

Fig. 4. Simulation geometries for different source-to-detector distances: (a) 1, (b) 2, (c) 3, and (d) 5 cm.

3.2. Source-to-detector distance (SDD)

The second parameter investigated was the source-to-detector distance (SDD). The signal changes were estimated as a function of the SDD (1, 2, 3, and 5 cm) with the fixed IDD of 10 cm. Fig. 4 shows the simulation geometries. In this simulation, the position of the reflectors was fixed, while the position of the detection system (i.e., gamma-ray shield, HDPE moderator, and ^{3}He detectors) was changed. The FNEM, PNAR, and FNEM × PNAR signals decreased with increasing SDD, as shown in Fig. 5. This was due mainly to the fact that the neutron multiplication was reduced as the SDD was increased. A close SDD is also beneficial in terms of the detection efficiency. Again, the FNEM and PNAR signals showed very close value and no physics reason was identified at this moment. The optimal SDD was determined to be 1 cm.

3.3. Neutron-reflector material

The effect of the material of the side neutron reflector was investigated. For this, the material was changed, from the graphite of the initial design, to HDPE, steel, and tungsten. The IDD and SDD were fixed to the optimal values of 10 cm and 1 cm, respectively. The estimated signals with measurement precisions ($\pm 1\sigma$) are listed in Table 1. The highest signal was obtained for the case of HDPE. Again, this was due mainly to the fact that neutron multiplication with the HDPE reflector showed the highest value, 1.12. As a result, the optimal material of the side neutron reflector was determined to be HDPE.

3.4. Cadmium sleeve

A 2-mm-thick Cd sleeve around the close detectors was installed to tune their response. The structure of the Cd sleeve was varied from a full cylinder to a partial cylinder on the back side of the close detectors, which covers 360 (i.e., full cylinder), 270, 180, and 0° (i.e., no Cd sleeve). Fig. 6 shows the detection geometry for the half-cylinder case (i.e., 180°). As the angle covering the close detector was increased, the average incident neutron energy that induces the fission events increased, resulting in an increased FNEM × PNAR signal. On the other hand, the induced fission rate decreased with the Cd sleeve as a consequence of the reduction of the number of thermal neutrons returning back to the fuel region, resulting in reduced neutron multiplication and, in turn, a reduced FNEM × PNAR signal. These results indicated that there is a trade-off with respect to the FNEM × PNAR signal: the changes in the average incident neutron energy have a positive effect, whereas the

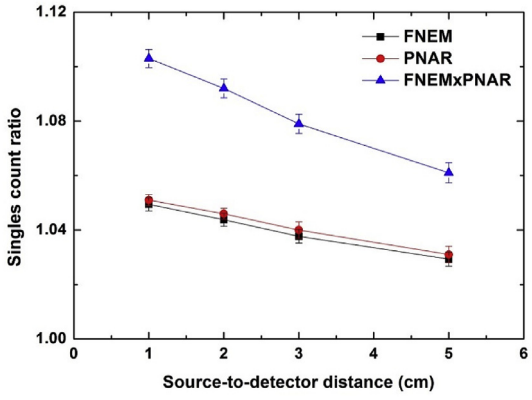

Fig. 5. FNEM, PNAR, and FNEM × PNAR signals as functions of source-to-detector distance.

Table 1
FNEM, PNAR, and FNEM × PNAR signals as functions of neutron-reflector material.

	FNEM	±1σ	PNAR	±1σ	FNEM × PNAR	±1σ
Graphite	1.050	0.002	1.051	0.002	1.103	0.003
HDPE	1.071	0.002	1.080	0.003	1.157	0.003
Steel	1.050	0.002	1.051	0.002	1.104	0.003
Tungsten	1.044	0.002	1.047	0.002	1.093	0.003

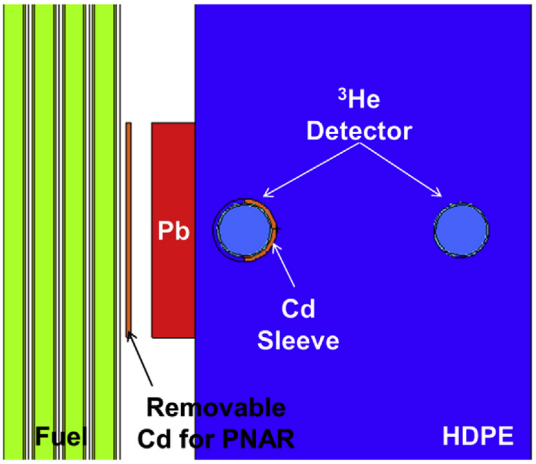

Fig. 6. Simulation geometry for different structures of cadmium sleeve around close detectors (half-cylinder case).

Table 2
FNEM, PNAR, and FNEM × PNAR signals for different structures of cadmium sleeve around close detectors.

Cd sleeve angle	FNEM	±1σ	PNAR	±1σ	FNEM × PNAR	±1σ
0	1.071	0.002	1.080	0.002	1.157	0.003
90	1.091	0.003	1.085	0.003	1.184	0.004
180	1.131	0.003	1.089	0.003	1.231	0.004
270	1.170	0.004	1.093	0.004	1.278	0.005
360	1.023	0.005	1.072	0.005	1.096	0.007

changes in the neutron multiplication have a negative effect, with the increase of the Cd sleeve angle. The simulated signals with the measurement precisions (±1σ) for the different structures of Cd sleeve are listed in Table 2. The FNEM × PNAR signal changed from 1.096 to 1.278 according to the Cd sleeve structure. The optimal structure, that which showed the highest signal, covered the close detectors to 270°.

3.5. Air cavity in moderator

The last parameter studied was the air cavity installed within the HDPE moderator. The HDPE is a good moderator for slowing down of neutrons through elastic scatterings; however, some amounts of neutrons can be absorbed by the moderator before they contribute to the signal or return to the fuel assembly. So, it was expected that the FNEM × PNAR signal could be tuned to the air cavity within the HDPE moderator. To investigate the effect of the air cavity, its size was varied from 0 to 5 cm (Fig. 7). The simulation results showed that there was no significant gain from the installation of the air cavity, as shown in Fig. 8. This was due mainly to the fact that the changes in neutron multiplication and average incident neutron energy inducing fission events were again affected in opposite ways, and thus canceled in the FNEM × PNAR signal. In fact, the detection efficiency was affected by the air cavity in a

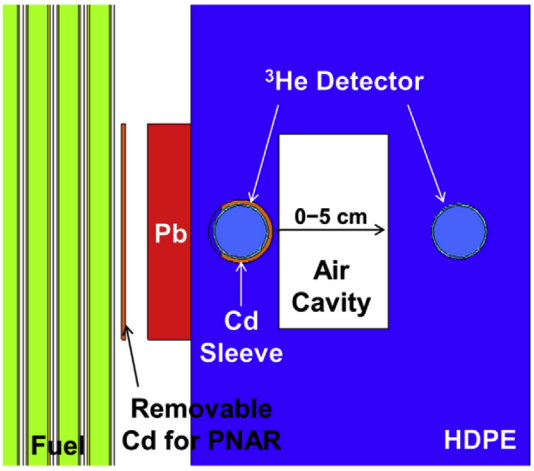

Fig. 7. Simulation geometry for different sizes of air cavity in HDPE moderator (5-cm case).

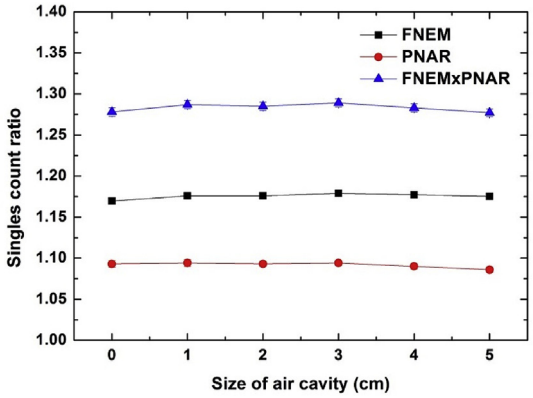

Fig. 8. FNEM, PNAR, and FNEM × PNAR signals as functions of size of air cavity.

negative and positive way for the close and far detectors, respectively. It is worthwhile to note that the changes in detection efficiency of the close and far detectors were canceled when the signal was normalized to the unity for each no-fissile-material case (i.e., the 0 wt% enrichment case for which the ^{235}U was replaced with ^{238}U) [9]. The 0 wt% enrichment case was used for the normalization to illustrate the change in response with respect to the fissile content because it removes the response from the induced fission reactions in the ^{235}U. Because there was no significant gain with the air cavity, we decided to remove it from our optimized design for ease of manufacture.

3.6. Estimation of detector response

Based on the optimized detection geometry determined above, the signals were estimated with different ^{235}U enrichments of a fresh-fuel assembly ranging from 0 to 5 wt%. The detection efficiencies of the close and far detectors were 0.09% and 0.13%, respectively, for the fresh-fuel assembly with 5 wt% enrichment. The expected count rate was only ~1 counts/s; hence, the measurement precision was calculated for 100 h measurement. The FNEM, PNAR, and FNEM × PNAR signals increased with increasing enrichment, as shown in Fig. 9. The neutron multiplication increased from 1.04 to 1.12, resulting in a 27.8% signal enhancement. Although this measurement system can provide the signature of the relationship between enrichment (= amount of fissile

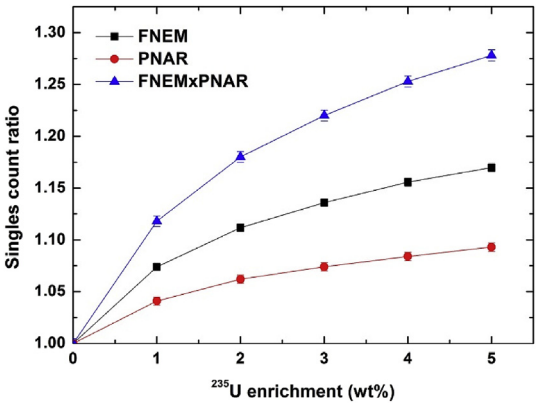

Fig. 9. FNEM, PNAR, and FNEM × PNAR signals of optimized detection geometry as functions of ^{235}U enrichment range of 0–5 wt%.

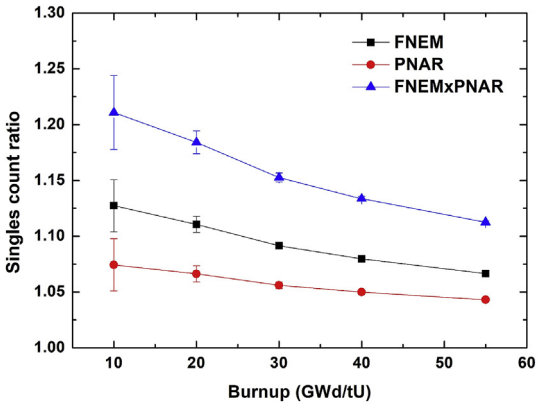

Fig. 10. FNEM, PNAR, and FNEM × PNAR signals as functions of burnup range of 10–55 GWd/tU with initial enrichment of 4.5 wt% and cooling time of 10 years. Measurement precision was calculated for 100 s measurement.

material) and signal, it is not practical for a fresh-fuel assembly, because, due to extremely low neutron emission rate, a measurement time of 100 h is required for ~0.5% measurement precision.

The surrounding material was changed from air to water in order to estimate the signal change for the fresh-fuel assembly with 5 wt% enrichment. The simulation result showed that the FNEM, PNAR, and FNEM × PNAR signals increased due to the increased multiplication when the assembly was surrounded by water (Table 3). As expected, the PNAR signal was larger than the FNEM signal because the PNAR is based on the thermal neutron multiplication. The signal enhancement of the FNEM × PNAR signal was about 210% from 1.278 to 2.686. Although a sophisticated engineered design is required for underwater applications, it would be beneficial in terms of the signature intensity.

In order to estimate the signal for a spent-fuel assembly with different burnups, the isotopic compositions of the spent fuels were calculated by the OrigenARP code [24]. The spent fuel considered here has a burnup range of 10–55 GWd/tU with an initial enrichment of 4.5 wt% and a cooling time of 10 years. The calculated isotopic compositions in the case of 55 GWd/tU burnup were 93.9 wt% of uranium, 1.2 wt% of plutonium, 1.7 wt% of rare-earth elements, and 3.2 wt% of the rest elements (e.g., minor actinides, noble metals, and alkali metals). The calculated neutron multiplication decreased with increasing burnup from 1.12 to 1.09. The expected count rates for the close and far detectors, in the case of 55 GWd/tU burnup, were ~6.5 × 10^4 and ~1.1 × 10^5 counts/s, respectively; hence, the measurement precision of 0.1% could be obtained for about 100 s of measurement. However, in the case of 10 GWd/tU burnup, the expected count rates for the close and far detectors were significantly reduced to ~7.6 × 10^1 and ~1.2 × 10^2 counts/s, respectively, due to the reduced neutron emission rate of the spent-fuel assembly. The estimated FNEM × PNAR signals for the spent-fuel assemblies, as shown in Fig. 10, decreased with increasing burnup from 1.211 to 1.113, which was normalized to the no-fissile-material (^{235}U, ^{239}Pu, and ^{241}Pu) case. For the no-fissile-material case, ^{235}U, ^{239}Pu, and ^{241}Pu, which are the major fissile isotopes in the spent fuel considering the induced fission cross section and the amount, were removed and the removed mass was converted into the remaining isotopes while maintaining the original mass. In the case of measuring an actual spent-fuel assembly, the measured FNEM × PNAR signal would be normalized to a dummy fuel assembly with depleted uranium for the no-fissile-material case. According to Fig. 9, the FNEM × PNAR signal for 55 GWd/tU burnup case was expected to be ~1.15, as correspondent to the ^{235}U enrichment of 1.4 wt%, because the fraction of fissile material (^{235}U, ^{239}Pu, and ^{241}Pu) for the spent fuel was about 1.4 wt%. This indicated that there was an approximately 3% signal reduction due to the neutron absorbers (Sm and Gd) in the spent fuel.

Table 3
FNEM, PNAR, and FNEM × PNAR signals for different surrounding materials (air and water).

Surrounding material	FNEM	±1σ	PNAR	±1σ	FNEM × PNAR	±1σ
Air	1.170	0.004	1.093	0.004	1.278	0.005
Water	1.522	0.004	1.765	0.005	2.686	0.006

4. Summary

In the present study, a neutron measurement system for nuclear material accountancy of a spent-fuel assembly was designed by Monte Carlo simulations. A spent-fuel assembly contains different amount of fissile material, resulting in different neutron multiplications. So, the system was designed to measure the signature of neutron multiplication. Two measurement techniques, FNEM and PNAR, were incorporated into the system. FNEM and PNAR are sensitive to induced fissions by fast and thermal neutrons, respectively. In order to optimize the measurement system, the effects of various parameters were investigated, including inter-detector distance, source-to-detector distance, neutron-reflector material, the structure of the cadmium sleeve around the close detectors, and the air cavity in the moderator. Then, with the optimized detection geometry, the detector responses for the fresh-fuel assemblies with different ^{235}U enrichments were estimated. The neutron multiplication of the fresh-fuel assemblies was varied from 1.04 to 1.12 according to the enrichment, resulting in signal increases up to 27.8%. On this basis, the measurement system's sensitivity to neutron multiplication was confirmed.

In order to estimate the detector response for a spent-fuel assembly with different burnups, the isotopic compositions of the spent-fuels assembly with a burnup range of 10–55 GWd/tU with 4.5 wt% initial enrichment and 10 years cooling time were calculated by the OrigenARP code. Then, the detector response was estimated by MCNP6 simulations. The expected count rate for the close and far detectors, in the case of 55 GWd/tU burnup, was about 10^4–10^5 counts/s with a measurement precision of 0.1% for 100 s measurement. The estimated FNEM × PNAR signals for the spent-fuel assemblies decreased with increasing burnup due to the reduced neutron multiplication. Note that a spent-fuel assembly has a burnup distribution in the axial and radial directions, resulting in the different isotopic compositions depending on the

position. As a result, it was expected that axial scanning and measurement on the 4 sides of an assembly should be beneficial for error minimization. The effects of various combinations of spent-fuel conditions in terms of burnup, initial enrichment, and cooling time should be focus of a follow-up study evaluating the applicability of the measurement system to a spent-fuel assembly.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This study was supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (NRF-2018R1D1A1B07043515). This study was also supported by National Research Foundation of Korea (NRF) funded by the Ministry of Science and ICT (NRF-2017M2A8A5015084) and by research funds for newly appointed professors of Chonbuk National University in 2018.

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