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Original Article

Isotopic Fissile Assay of Spent Fuel in a Lead Slowing-Down Spectrometer System

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ABSTRACT

A lead slowing-down spectrometer (LSDS) system is under development to analyze isotopic fissile content that is applicable to spent fuel and recycled material. The source neutron mechanism for efficient and effective generation was also determined. The source neutron interacts with a lead medium and produces continuous neutron energy, and this energy generates dominant fission at each fissile, below the unresolved resonance region. From the relationship between the induced fissile fission and the fast fission neutron detection, a mathematical assay model for an isotopic fissile material was set up. The assay model can be expanded for all fissile materials. The correction factor for self-shielding was defined in the fuel assay area. The corrected fission signature provides well-defined fission properties with an increase in the fissile content. The assay procedure was also established. The assay energy range is very important to take into account the prominent fission structure of each fissile material. Fission detection occurred according to the change of the Pu239 weight percent (wt%), but the content of U235 and Pu241 was fixed at 1 wt%. The assay result was obtained with 2~3% uncertainty for Pu239, depending on the amount of Pu239 in the fuel. The results show that LSDS is a very powerful technique to assay the isotopic fissile content in spent fuel and recycled materials for the reuse of fissile materials. Additionally, a LSDS is applicable during the optimum design of spent fuel storage facilities and their management. The isotopic fissile content assay will increase the transparency and credibility of spent fuel storage.

1. Introduction

The accumulation of nuclear spent fuel is currently a significant issue in Korea. The storage capacity of spent fuel will soon reach the maximum level; therefore, spent fuel policies and technology must be developed in the near future. One option for spent fuel treatment is the pyroprocessing technology in Korea to reuse the fissile materials and reduce the high level and long lived waste volume. At the Korea Atomic Energy Research Institute (KAERI), a pyroprocess which

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extracts the fissile material from used fuel to fabricate fuel rods for a sodium fast reactor (SFR) was developed. In the used fuel, ~1.4% of the fissile material still exists [1], depending on the initial enrichment and burnup. The pyroprocess produces a transuranium (TRU) mixture with some fission products. The uranium-TRU mixture is used to fabricate the fuel for the SFR. Therefore, before fabricating the fuel rod, verification of the isotopic fissile content must be carried out to ensure the quality of the fabricated fuel for safety and economics purposes.

A lead slowing down spectrometer (LSDS) is under development at the KAERI for analysis of the isotopic fissile material contents in used fuel [2,3]. The advantage of the LSDS is that it induces direct fissile fission and can analyze the isotopic fissile content in near real time. The isotopic fissile content is obtained by measuring the direct fission neutrons from each fissile material. The fission measurement has a correlation with the content of the fissile materials. Each fissile material, U235, Pu239, and Pu241 in this case, has different fission characteristics with respect to the neutron energy. Therefore, if a continuous source of neutron energy can be produced, the dominant fission by each fissile material can be measured and applied for a content assay. Additionally, the source neutron must be intense to overcome the neutron background from the curium spontaneous fission in the used fuel. Usually, a used fuel has a very complex radiation background.

In order to produce the source neutron, one section electron linear accelerator was chosen with a multiple layered tantalum target. Thus, a cost-effective, size compactness, and easy maintenance system was considered for neutron generation. Moreover, a linear accelerator offers the advantage of being able to increase the accelerating energy simply by adding an accelerating column when necessary. In the target, from an (e, γ)(γ, n) reaction, a source neutron is produced at an approximate energy of 0.5 MeV. The source neutron slows down in the lead medium and a continuous neutron spectrum is obtained.

In the designed device [3], the neutron spectrum and resolution were analyzed and the neutron resolution was found to be fine in the lead medium, with an energy range of keV~eV. From the relationship between the fissile fission and the fission measurements, a mathematical assay model for the isotopic fissile content was developed based on the assumption that fast fission neutrons only come from fissile content. A correction factor for self-shielding was developed with respect to slowing down energy of the source neutron by the Monte Carlo method. The correction factor is a very important parameter to increase the accuracy of the content assay. The assay sensitivity was performed in the fuel rod by changing the Pu239 enrichment in the range of 1~10 weight percent (wt%). The fuel rods use U235, Pu239, Pu241, and U238 as the base materials.

The energy deposition by an electron was calculated in the Ta (Tantalum) target layers, Be (Beryllium), and in the lead medium. For a practical throughput value for this assay system, the highly irradiated fuel assembly that arises from the high radiation levels could be assayed. The calculation of the effective shielding was done by applying an additional shield cover around the spectrometer to decrease the total dose in the system. The development of a lead spectrometer must be pursued in order to minimize assay errors. These types of error can be reduced by selecting the proper assay energy range for the dominant type of fission by each fissile material.

The technology development for an accurate fissile content assay can be applied in various areas using nuclear spent fuel. These include the optimum designs of storage sites for spent fuel and efforts to maximize the burnup credit and utilization of the existing fissile materials in the spent fuel. Additionally, an improvement of the fissile content assay will play an important role in international transparency and credibility in in the area of spent fuel management.

2. Lead spectrometer

2.1. *Fissile assay model*

In the designed lead spectrometer, the source neutron slows down in the lead medium and the neutron energy from eV to keV enters the fuel assembly. The fission neutrons induced by fissile materials are measured at surrounding fission chamber detectors. Based on the assumption that there is a linear relation between the induced fission and the fissile mass, a linear detection model was set up to obtain the content of the isotopic fissile material [4,5]. The assay model has a property in that there is linearity between detection and fissile fission. The detected signal involves information of the fission from U235, Pu239, and Pu241 by the interrogation source neutron. The detector signal is expressed as shown below:

$$y_i = k\varepsilon \left[\nu_{1i}N_1 < \sigma_{f,1}\phi >_i + \nu_{2i}N_2 < \sigma_{f,2}\phi >_i + \nu_{3i}N_3 < \sigma_{f,3}\phi >_i\right] \quad (1)$$

where y_i is the detector signal at channel i (the slowing down time), k is the normalization constant (accounting for the geometric effect and the number of fission neutrons entering the detector), and ε is the detector efficiency. Here, the numbers 1, 2, and 3 represent the fissile materials, U235, Pu239, and Pu241, respectively, and N is the fissile mass. ν is

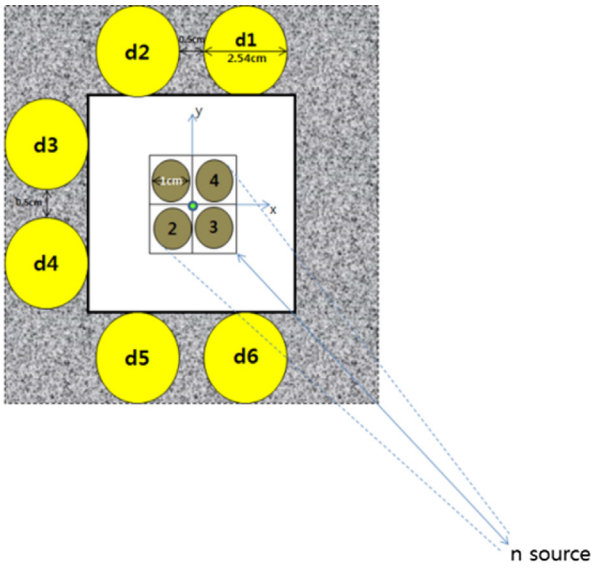

Fig. 1 – Configuration of the detector, assay area, and fuel rod for fission neutron detection.

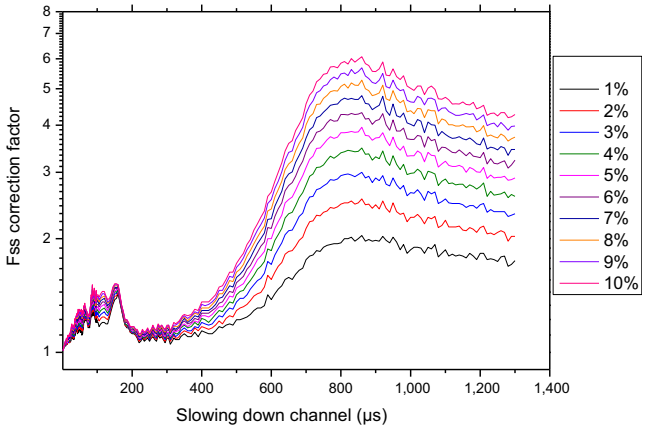

Fig. 2 – Correction factor for the amount of Pu239 (Pu241: 1 wt% used). Fss, self-shielding factor.

the average neutron yield by the fission of each fissile material, σ_f is the fission cross section, and ϕ is the source neutron intensity entering the fuel rod. Therefore, $<\sigma_{f,1}\phi>_i$ represents the fission reaction rate according to the source neutron at channel i. Therefore, for each fissile material, the fission rate by the source neutron at channel i is expressed as:

$$<\sigma_{f,1}\phi>_i = \frac{\int_i \sigma_{f,1}\phi\, dt}{\int_i dt}. \quad (2)$$

The detector collects the fast fission neutrons through the fissile material. The detector discriminates the fission neutrons from the source neutrons and the spontaneous fission neutrons according to the spent fuel. The detector is located one inch above the fuel assay area. The overall configuration of the detector, the assay area, the fuel rod, and the source neutron is shown in Fig. 1. In the figure, the numbers 1, 2, 3, and 4 represent the fuel rods, while d1, d2, d3, d4, d5, and d6 represent the detectors. The source neutron is located at a distance of 40 cm in the diagonal direction, as shown in Fig. 1. The detection signal is expressed as:

$$\int_{S_{det}} \int_{E_1}^{E_2} \sigma_f \varnothing_f(r,E,t)\,dE\,dA, \quad (3)$$

where $\varnothing_f$ is the fission neutron flux entering the detector area (S_{det}) and σ_f is the fission cross-section of the detector material. Therefore, $\sigma_f\varnothing_f$ represents the response rate by the incoming neutrons in the detector material.

In the assay model of Eq. (1), a self-shielding factor (F_{ss}) must be taken into account to enhance the linearity between the fissile fission and the mass [6]. Due to the competition of the specific reaction, a fission reaction occurs in the compound. Usually, when a large volume of nuclear material or many fuel rods are analyzed, the self-shielding factor increases. Therefore, it is a very important factor in content assays. By applying the self-shielding factor, the detector signal improves in terms of linearity and provides a better assay result. The data analysis model was developed using a regression analysis over the assay energy range to obtain the fissile content. The error between the measured (y_i) and calculated values is defined at the i^{th} channel. By minimizing the error, the content matrix (U235, Pu239, and Pu241) is obtained.

The measurement is conducted with respect to the neutron slowing down time, after the source neutron burst. The relationship between the slowing down time and the energy in the lead medium was determined and the parameters were obtained for system operation. The relationship is expressed as shown below:

$$E = \frac{k}{(t+t_0)^2}. \quad (4)$$

Here, t_0 (0.67 μsec) and k (167033 eV- μsec^2) are the values for the LSDS system.

2.2. *Correction factor*

In the LSDS system using a source neutron, most of the absorption of the source neutron comes from the capture and fission properties. Fission is a competition reaction, and an induced fission neutron is used for a content assay. Therefore,

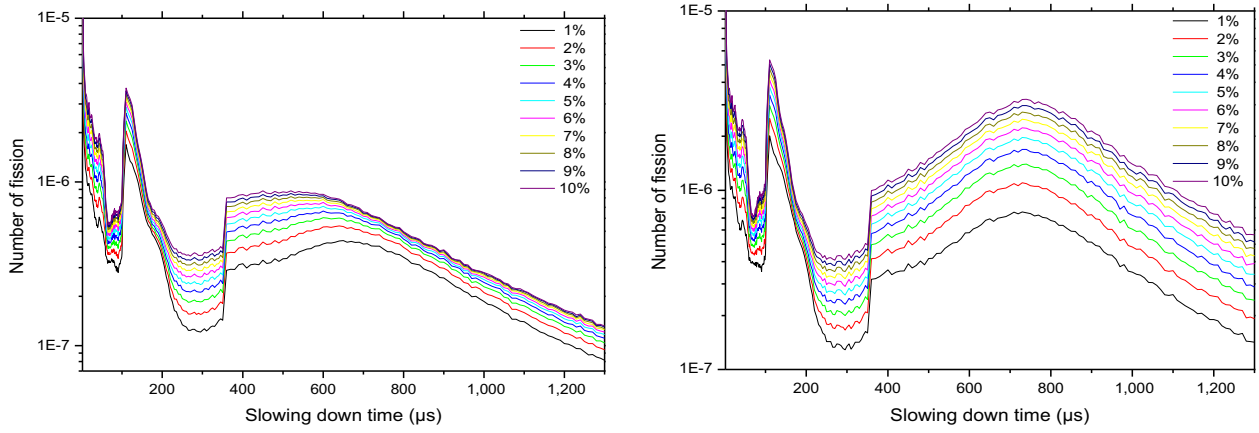

Fig. 3 – Difference in the fission signal before and after the correction (Pu239).

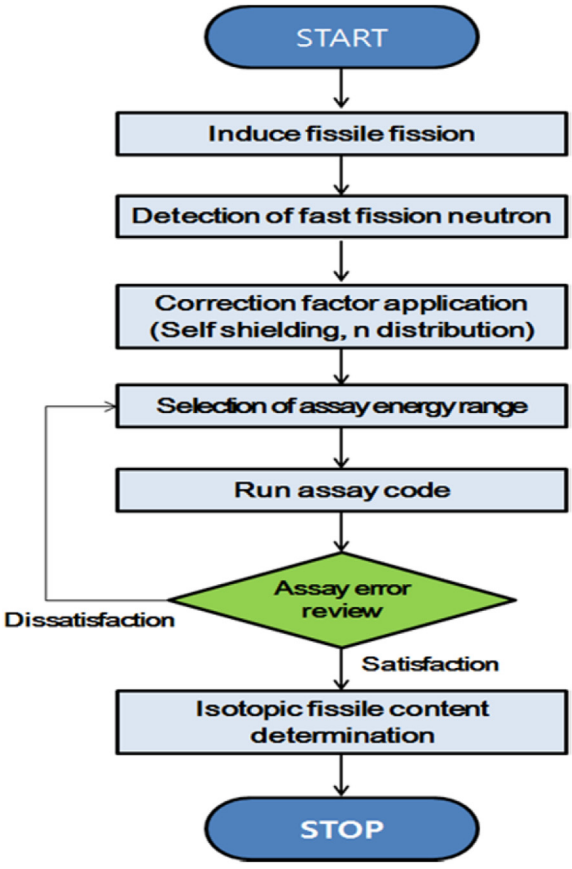

Fig. 4 – Procedure of the isotopic fissile assay.

the correction factor for fissile fission from the absorption reaction is required, in order to improve the linearity of the fissile fission rate. An underestimated fission rate gives rise to a lower fissile content. The correction factor (F_{ss}) is simply expressed as:

$$\text{Correction factor} = \frac{\Phi_s}{\Phi_f}, \quad (5)$$

where Φ_s is the source neutron flux arriving at the fuel and Φ_f is the neutron flux in the fuel.

In the final products achieved through the pyroprocess using spent fuel, TRU is the major material. The content of plutonium is relatively high, with Pu239 accounting for a particularly large portion. Therefore, when such a TRU is used for the fabrication of a fuel rod for an SFR, the correction factor based on Pu239 is evaluated. Normally, the content of Pu241 in spent fuel is very low. Therefore, in the simulation, 1 wt% of Pu241 is used.

Fig. 2 shows the correction factor calculated using Eq. (5) with a change of 1 wt% to 10 wt% of Pu239 in the fuel rod, according to the MCNP code [7]. The evaporation source neutron and the volume tally at the fuel rod were used in the code simulation. The correction was calculated over all slowing down time channels. The result shows that as the weight of Pu239 increases, the correction factor increases. The figure does not show a great difference at the initial time, but after 400 μseconds, a difference rapidly appears. Therefore, for a large amount of plutonium in the assay, the correction is a very important factor for a lengthy time period. In addition, it increases rapidly after a detection time of 600 μseconds. Pu239 has a very high fission property at 0.3 eV. Therefore, at around 800 μseconds, the correction factor reaches its largest value. The correction factor at 800 μseconds is nearly six times larger than at 200 μseconds. Such a correction will contribute to an accurate assay.

Fig. 3 shows the normalized fission event in the fuel rod before and after the correction factor is applied based on the change (1~10 wt%) in the Pu239 content. As shown in the figure, before the correction, the fission rate according to an increase in the amount of Pu239 does not increase linearly. In highly enriched cases, the increment becomes relatively small as compared to that of low enrichment. After 600 μseconds, even though the content of Pu239 increases, the result shows that the fission rate does not match the corresponding enrichment increase. However, after the correction is applied, the fission rate shows a linear increment with respect to the amount of the increase. Therefore, the corrected fission signal will give an accurate assay result. However, in an energy range with considerable neutron absorption after 800 μseconds, the assay result must be verified for accuracy, even when a correction is applied. An energy range with less absorption but which includes a dominant fission structure is recommended for a fissile assay.

2.3. *Fissile content assay*

Based on the assay model suggested in Eq. (1), an isotopic fissile assay was performed using the geometry shown in Fig. 1. In the system, the induced fissile fission neutrons are detected by the surrounding threshold detectors (in a U238 fission chamber), using the MCNP code [7]. Fission neutron detection has a direct relation to the amount of the fissile material. The detection signal in each channel is the sum of the fission by U235, Pu239, and Pu241. Fig. 4 shows the procedure of the isotopic fissile assay. After correcting the fission

Table 1 – Isotopic fissile content assay (U235:1%, Pu241:1%, Pu239 changed).

Mixed (%)	1-1-1	1-2-1	1-3-1	1-4-1	1-5-1	1-6-1	1-7-1	1-8-1	1-9-1	1-10-1
U235	1.01	1.00	1.04	1.00	1.03	0.99	1.01	1.06	1.15	1.23
Error	0.09	0.10	0.12	0.10	0.11	0.11	0.11	0.09	0.12	0.12
Pu239	1.05	1.82	3.10	4.01	5.09	5.94	7.04	8.00	9.26	10.01
Error	0.02	0.03	0.05	0.03	0.04	0.04	0.05	0.04	0.05	0.06
Pu241	1.15	1.21	1.18	1.24	1.24	1.28	1.27	1.31	1.28	1.29
Error	0.02	0.02	0.03	0.02	0.02	0.02	0.02	0.02	0.02	0.02

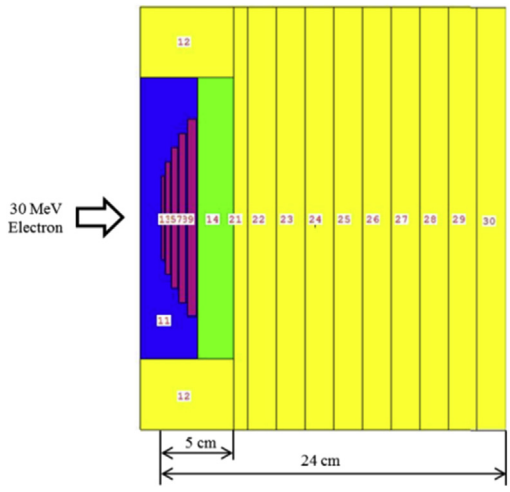

Fig. 5 – Model for the simulation of the electron energy deposition.

signal, the assay range was selected to include each dominant fissile fission signature. According to an analysis of the assay error, the final assay range is selected.

In the assay simulation [4,7], the combination of uranium and plutonium exists in a fuel rod. However, the overall content of U235 and Pu241 was fixed at 1% in the mixture, and the content of Pu239 was changed only in the range of 1–10 wt%. The amount of Pu239 is dominant in SFR fuel. From the assay, the accuracy was approved for uranium and plutonium. In addition, the assay sensitivity was examined. Pu239 has a giant fission property around 0.3 eV, which is distinguished from those of other fissile materials. Table 1 shows a summary of the fissile content assay result from the model. In the table, the mixture order is as follows: U235, Pu239, and Pu241.

The assay was conducted in the entire energy region and in the subenergy region by considering their characteristic resonance fission energy. In the low enriched Pu239 case, the content was obtained within ~3% error, while for the highly enriched Pu239 case, an accurate assay was obtained with ~2% error. However, the Pu241 assay showed relatively low accuracy. In the entire assay, the accuracy of the content for Pu241 is not as good as that for Pu239. However, in the products after the pyroprocess, the content of Pu241 is much lower than that of Pu239. For the U235 assay, a reasonable result was obtained, but at 9 wt% and 10 wt% of Pu239, the content accuracy of U235 was relatively poor. The fission structures of Pu239 and U235 are much different in the entire slowing down energy range (0.1eV ~ 1 keV). However, Pu239 and Pu241 have similar fission structures.

2.4. *Energy deposition by electrons*

To induce fissile fission in an LSDS system, an intense neutron generation system is required. Electrons are a proper option to produce the source neutrons by hitting the target material. However, to produce intense neutrons, high electron energy and current levels are typically necessary. The electrons generate neutrons through an (e, γ)(γ, n) reaction at the target and arrive at the lead medium after the target. Lead has a relatively low melting temperature. Therefore, the energy deposition at each layer of the target and the lead medium was simulated along the electron beam pathway.

Fig. 5 shows the target geometry and lead medium position [8]. The target has five Ta layers with a Be layer at the end. Be is used to maximize the beam utilization. Lead is positioned after the Be material. Ten positions were selected in the lead medium to examine the energy deposition characteristics. Fig. 6 shows the neutron and electron spectrum at each target layer, at Be, and at different lead locations. In the first and second layers of the Ta target, greater intensity of the neutrons and electrons was obtained. At the Be plate, a decrease in the neutron and electron flux occurred because the electron lost its energy in the Ta layers along the beam path. Therefore, the neutron intensity becomes relatively low at the Be layer. After the Be layer, there is still an electron beam path, but the neutron generation is very low.

Table 2 shows the energy deposition at the each position according to 30 MeV incident electrons. Along the beam path, the electrons and produced radiations deposited their energy at the target and lead medium. From the table, the electrons deposited more energy than the neutrons and gamma rays. However, the energy deposited by the neutrons and gamma rays is not negligible. At the second layer of the Ta target, most

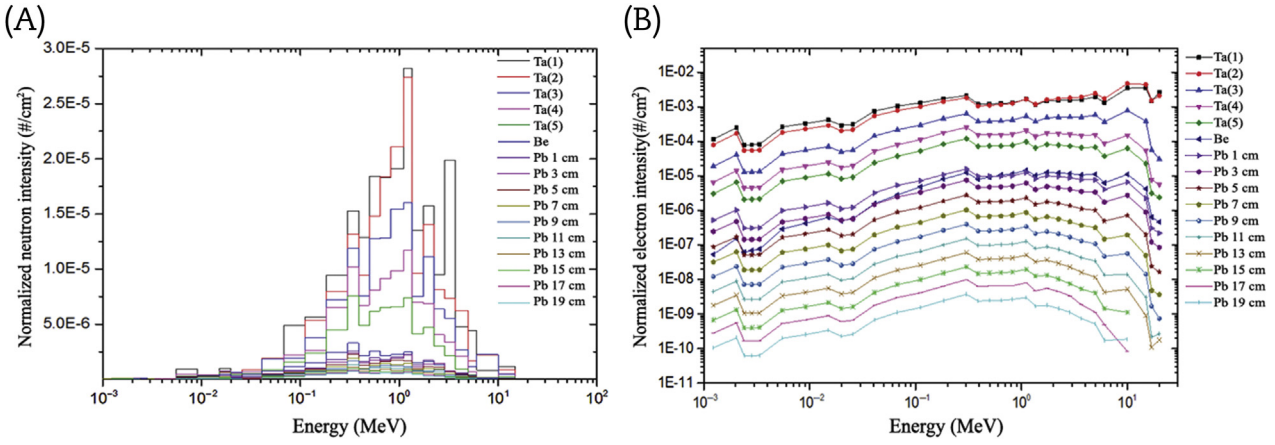

Fig. 6 – Neutron and electron spectrum at each position. (A) Neutron spectrum. (B) Electron spectrum. # means number of electrons.

Table 2 – Energy deposition by an electron beam (30 MeV) at the target layer and the lead location.

Material		Energy deposition (W)		
		Electron	Neutron & gamma	Total
1st (Ta)		405.8	93.1	498.9
2nd (Ta)		706.3	269.8	976.1
3rd (Ta)		329.5	259.6	589.1
4th (Ta)		201.5	200.3	401.8
5th (Ta)		152.3	147.2	299.5
Be (reflector)		29.3	20.7	50.1
Pb	1 cm	99.5	94.2	193.7
	3 cm	93.1	81.8	174.9
	5 cm	32.9	27.5	60.5
	7 cm	12.0	9.7	21.7
	9 cm	4.5	3.5	8.1
	11 cm	1.6	1.3	3.0
	13 cm	0.6	0.5	1.1
	15 cm	0.2	0.19	0.4
	17 cm	0.1	0.0	0.1
	19 cm	0.0	0.0	0.0

of the energy deposition occurs. At all target areas, a large amount of energy was accumulated, therefore, a cooling system may be necessary in order to prevent melting. In the lead, after a thickness of 3 cm, energy deposition decreases drastically, and after 17 cm, energy deposition overall no longer exists. Information pertaining to energy deposition is therefore very important for neutron production and material integrity.

2.5. *Effective shielding by covering*

The shielding calculation for the LSDS facility was done using a combination of concrete and borax [9], while also considering the economics and optimization. In addition to the facility shielding calculation, a covering material is applied around the spectrometer to capture leaked neutrons and to reduce the dose rate to the area outside the facility. In the spectrometer, there are several intense neutrons: source neutrons, fissile fission neutrons, and spontaneous fission neutrons generated by the spent fuel. Therefore, if the neutrons around the spectrometer are captured, the dose rate outside the facility will decrease. This will lead to a decrease in the facility wall thickness. Almost all gamma rays are shielded in the lead medium.

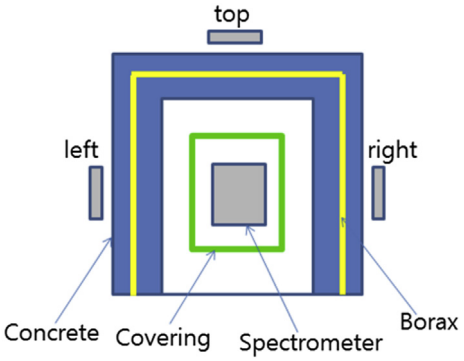

Fig. 7 – Covering geometry around the spectrometer.

Table 3 – Dose rate outside of the facility wall when applying the cover.

Material and Thickness	Dose rate (μSv/hr)			Shielding rate(%)		
	Left wall	Right wall	Top wall			
No covering	4.48E-02	3.49E-02	4.18E-02	–	–	–
HDPE-Borax (5 cm)	1.32E-02	1.11E-02	1.08E-02	70.5	68.2	74.2
HDPE-Borax (10 cm)	5.04E-03	5.80E-03	4.86E-03	88.8	83.4	88.4

HDPE, High Density Polyethylene.

Table 4 – Gap effect between the cover and the spectrometer.

Material and thickness	Gap (cm)	Dose rate (μSv/hr)		
		Left wall	Right wall	Top wall
HDPE-Borax (5 cm)	50	1.32E-02	1.11E-02	1.08E-02
	60	1.19E-02	1.09E-02	1.11E-02
	70	1.14E-02	1.08E-02	1.14E-02

HDPE, High Density Polyethylene.

Different thicknesses of the shielding material and different geometries were applied around the spectrometer. Fig. 7 shows the geometries of the cover (with a length of 250 cm on one side) and the spectrometer. As shown in Fig. 7, the gap between the cover and the spectrometer surface is 50 cm to reduce reflection into the spectrometer. Table 3 shows the calculated dose rate on the surface of the wall outside the facility, as specified in Fig. 7, when the cover is applied. When the 5-cm thick HDPE-borax covers the spectrometer, the dose rate decreases by 70% compared to that with no cover case, and it decreases by 88% at 10 cm. Therefore, the application of a cover around the spectrometer will contribute to a decrease in the wall thickness, which is related to the economics of the system.

Table 4 presents the gap effect between the cover and the spectrometer. However, it was shown that the gap did not have a significant effect in the simulation. Table 5 shows the result of the shielding effect by the height difference of the cover. The spectrometer has a cubical geometry (170 cm × 170 cm × 170 cm). Therefore, the cover height starts at 170 cm. Although the height exceeds 170 cm, the influence on the facility wall shielding is not great.

Table 5 – Height effect of the cover for shielding.

Material and thickness	Covering height (cm)	Dose rate (μSv/hr)		
		Left wall	Right wall	Top wall
HDPE-Borax (5 cm)	170	1.32E-02	1.11E-02	1.08E-02
	175	1.29E-02	1.09E-02	1.07E-02
	180	1.31E-02	1.09E-02	1.06E-02
	185	1.29E-02	1.07E-02	1.05E-02
	200	1.28E-02	1.05E-02	9.78E-03
	220	1.24E-02	1.02E-02	9.44E-03

HDPE, High Density Polyethylene.

3. Results and discussion

In the designed LSDS system [3], a mathematical isotopic fissile assay model was setup. The model feasibly describes the relationship between the isotopic fissile fission and fast fission neutron detection. The assay model can be expanded to an all fissionable material content assay. The relationship between the energy and the time was well defined in the spectrometer and the parameters worked well to express the dominant fission structures. The energy resolution in the lead was high enough to distinguish the dominant fissile fission. The assay energy range was determined to be from 0.1 eV to 1 keV, which involves the fission structures of the fissile material.

For a fissile content assay using the induced fission neutron in an LSDS system, the correction factor for self-shielding was defined in the fuel. The correction factor provides a linear relationship between the fissile fission and the fast neutron detection. The corrected fission represents a well-organized fission spectrum with respect to the slowing down energy. The correction plays an important role in ensuring an accurate fissile content assay. The fissile assay procedure was established. The content assay was conducted based on the procedure. In the fuel rod, the content of Pu239 was changed from 1 wt% to 10 wt%. For the low enrichment Pu239 cases, the content was obtained within ~3% error while for the highly enriched Pu239 cases, an accurate assay was obtained with ~2% error. However, a relatively poor assay result was obtained for Pu241. For the assay of U235, a reasonable assay result was obtained up to 8 wt% of Pu239 in the mixed fuel rod. However, at 9 wt% and 10 wt% of Pu239 in the rod, the assay result on U235 was poor.

In the additional shielding calculation when applying a cover around the spectrometer, the introduction of the cover was found to reduce the dose rate by 70% at the wall outside the facility, with 5 cm thickness of an HDPE-borax material. The covering length (250 cm), height (170 cm), and thickness (5 cm) were decided from the shielding calculation.

4. Conclusion

From a simulation, accurate isotopic fissile contents of U235, Pu239, and Pu241 were obtained. LSDS is a very promising technique for assaying the isotopic fissile content in used fuel and in recycled materials for the reuse of fissile materials. However, the source neutrons must be intense enough to obtain detection statistics to overcome the spent fuel background and to compensate for the decrease in neutron capturing by the fuel. Finally, direct fissile content assay technology will make a very useful contribution with regard to the fuel cycle to improve safety and economics and will enhance the transparency and credibility of spent fuel utilization and management efforts.

Conflicts of interest

All authors have no conflicts of interest to declare.

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