DEVELOPMENT OF LEAD SLOWING DOWN SPECTROMETER FOR ISOTOPIC FISSILE ASSAY

YONGDEOK LEE*, CHANG JE PARK†, SANG JOON AHN and HO-DONG KIM
Korea Atomic Energy Research Institute
*1045 Daedeok-daero, Yuseong-gu, Daejeon 305-353, Korea
† Gwang Gae To Building 1003C, 209 Neungdong-ro, Gwangjingu, Seoul 143-747, Korea
*Corresponding author. E-mail : parkcj@sejong.ac.kr

A lead slowing down spectrometer (LSDS) is under development for analysis of isotopic fissile material contents in pyro-processed material, or spent fuel. Many current commercial fissile assay technologies have a limitation in accurate and direct assay of fissile content. However, LSDS is very sensitive in distinguishing fissile fission signals from each isotope. A neutron spectrum analysis was conducted in the spectrometer and the energy resolution was investigated from 0.1eV to 100keV. The spectrum was well shaped in the slowing down energy. The resolution was enough to obtain each fissile from 0.2eV to 1keV. The detector existence in the lead will disturb the source neutron spectrum. It causes a change in resolution and peak amplitude. The intense source neutron production was designed for ~E12 n's/sec to overcome spent fuel background. The detection sensitivity of U238 and Th232 fission chamber was investigated. The first and second layer detectors increase detection efficiency. Thorium also has a threshold property to detect the fast fission neutrons from fissile fission. However, the detection of Th232 is about 76% of that of U238. A linear detection model was set up over the slowing down neutron energy to obtain each fissile material content. The isotopic fissile assay using LSDS is applicable for the optimum design of spent fuel storage to maximize burnup credit and quality assurance of the recycled nuclear material for safety and economics. LSDS technology will contribute to the transparency and credibility of pyro-process using spent fuel, as internationally demanded.

KEYWORDS : Fissile Assay, Slowing Down, Neutron Source, Fuel Cycle, Nuclear Material Utilization, Resolution, Radiation Measurement

1. INTRODUCTION

A optimum design of LSDS is performed for analysis of isotopic fissile content in pyro processed material and spent fuel at KAERI [1,2,3]. An isotopic fissile assay in spent fuel and recycled fuel is very difficult in a real application because of intense radiation background and weak radiation emission from fissile materials. Therefore, a direct use of emitting radiation from isotopic fissile material is very hard in the material content assay. Normally, an indirect mechanism is used for the content assay of fissile material [4]. However, an indirect methodology has a limitation in obtaining the accurate fissile content. In addition, error propagation by several steps in an indirect way results in increasing errors. Therefore, to obtain the direct signal from isotopic fissile material, an external radiation source is recommended.

The pyro processed material has a spent fuel property as well even though several fission products are extracted in the process. LSDS with an external neutron source is the most feasible technology to get a direct fission signal from isotopic fissile in the intense radiation background and to analyze the content of isotopic fissile material [5,6]. Moreover, LSDS does not need the burnup history information or burnup code help in a fissile assay. Also, a lead spectrometer itself has a shielding property of intense gamma emission from spent or pyro material. There are several reasons why the accurate fissile contents must be analyzed in spent fuel and pyro material:

1) Internationally, IAEA wants to verify the plutonium content at the storage site
2) Nationally, nuclear material approval in the shipping and receiving of spent fuel
3) For safe and economical reuse of fissile in spent fuel
4) For optimum storage design and maximum burn up credit application
5) For public acceptance of storage site and effective storage management.

The accurate content of fissile is very important data in reuse of fissile material and storage design of spent fuel. For the LSDS system operation, several calculations were done on the neutron spectrum analysis [7].
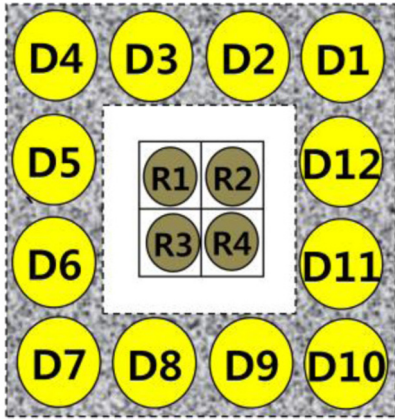
Fig. 1. Fuel and Detector Configuration.

The neutron spectrum influence by detector material and detector impurity in the spectrometer was examined. The fission characteristics were investigated in the slowing down neutron energy. Based on the designed geometry, the neutron energy resolution was investigated from 0.1eV to 100keV. The neutron detection efficiency was analyzed for multi layer cases and the detection sensitivity was done using U238 and Th232 threshold chamber. To obtain the proper neutron source, the neutron production mechanism was decided and the yield was calculated with the different incident electron energy. Also, a target activation analysis was performed.

Additionally, the advanced fissile assay technology will contribute to an increase in the transparency and international credibility of the reuse of fissile materials in future nuclear energy system development [8].

2. LEAD SLOWING DOWN SPECTROMETER

The lead spectrometer needs an external neutron source [9,10,11]. A source neutron slows down its energy by interaction with the lead medium. Therefore, a continuous slowed down neutron energy is obtained in the lead medium. The slowed down neutron finally enters into the nuclear fuel area and induces fission from fissile materials. An individual fissile material has its own fission characteristic below the unresolved resonance energy region. Therefore, if such a characteristic fission is well detected, it represents an amount of isotopic fissile material because the characteristic fission occurs directly from the fissile isotopes. In the lead spectrometer, very complex radiations exist. The fissile fission signal must be distinguished from the complex radiation field. A fission chamber is the right choice for screening the fast fission neutrons [1]. A fission chamber has a threshold property and it is not sensitive to intense gamma background. An electron linear accelerator was selected to produce an intense source neutrons. The neutrons are produced in the optimum designed Tantalum target, by $(e, \gamma)(\gamma, n)$ reaction.

2.1 Neutron Spectrum Analysis

In the lead spectrometer, the source neutron spectrum analysis was done at the fuel area. The source neutron spectrum represents a peak energy change and broadening when interacting with the lead and nuclear materials. The energy distribution was fitted to a Gaussian form. The detector is located in the first layer above the fuel. The fuel area is a 2x2 type assembly, as shown in Fig. 1. The presence of the detector in the lead will disturb the neutron spectrum. The U238 fission chamber was selected and the spectrum influence by the detector impurity was analyzed at 2, 4, and 6ppm of U235 as well. Fig. 2 shows the spectrum at the selected slowing down neutron energy, from 70keV to 0.4eV, in the case of no detectors and detectors with impurities. As shown in Fig. 2, the shape is consistent in all cases and the peak energy occurs at the same slowing down energy. The spectrum effect by detector is not severe. However, the detector existence gives a different energy resolution and peak amplitude change. The amplitude of the no detector case is generally higher than that of the case with a detector at all neutron energies. At higher energy, i.e., 70keV, the figure shows little difference, but, as the peak energy becomes lower, the peak gap between no detector and detector including impurity becomes larger. Specially, at 1.3keV, 110eV, 1.5eV and 0.47eV, the peak amplitude decreases by 11%, 17%, 18%, and 20%, respectively.

2.2 Energy Resolution

In the lead spectrometer, the resolving power of the neutron is an important parameter for the LSDS system operation. When the neutron shape follows a Gaussian distribution, the resolution is measured by the full width at half maximum.

$$R = (\frac{\Delta E}{E})_{FWHM} \qquad (1)$$

where E is the mean energy of the neutron distribution

The energy resolution was analyzed at the slowing down neutron energy. The resolution is important in getting individual isotopic fissile fission. Fig. 3 shows the energy resolution in the lead, from 1MeV to 0.1eV. The result shows a well-organized spectrum, with a ~0.3 resolution, from 3keV to 3eV. However, above 3keV and below 3eV, the resolution starts broadening. At 0.3eV, Pu239 has a big absorption resonance. Therefore, the resolution at 0.3eV is ~0.57. Therefore, by considering energy resolution, the expected assay energy range is approximately from 0.3eV to 20keV.

The inserted nuclear fuel produces a resolution function, which is different from that for homogeneous
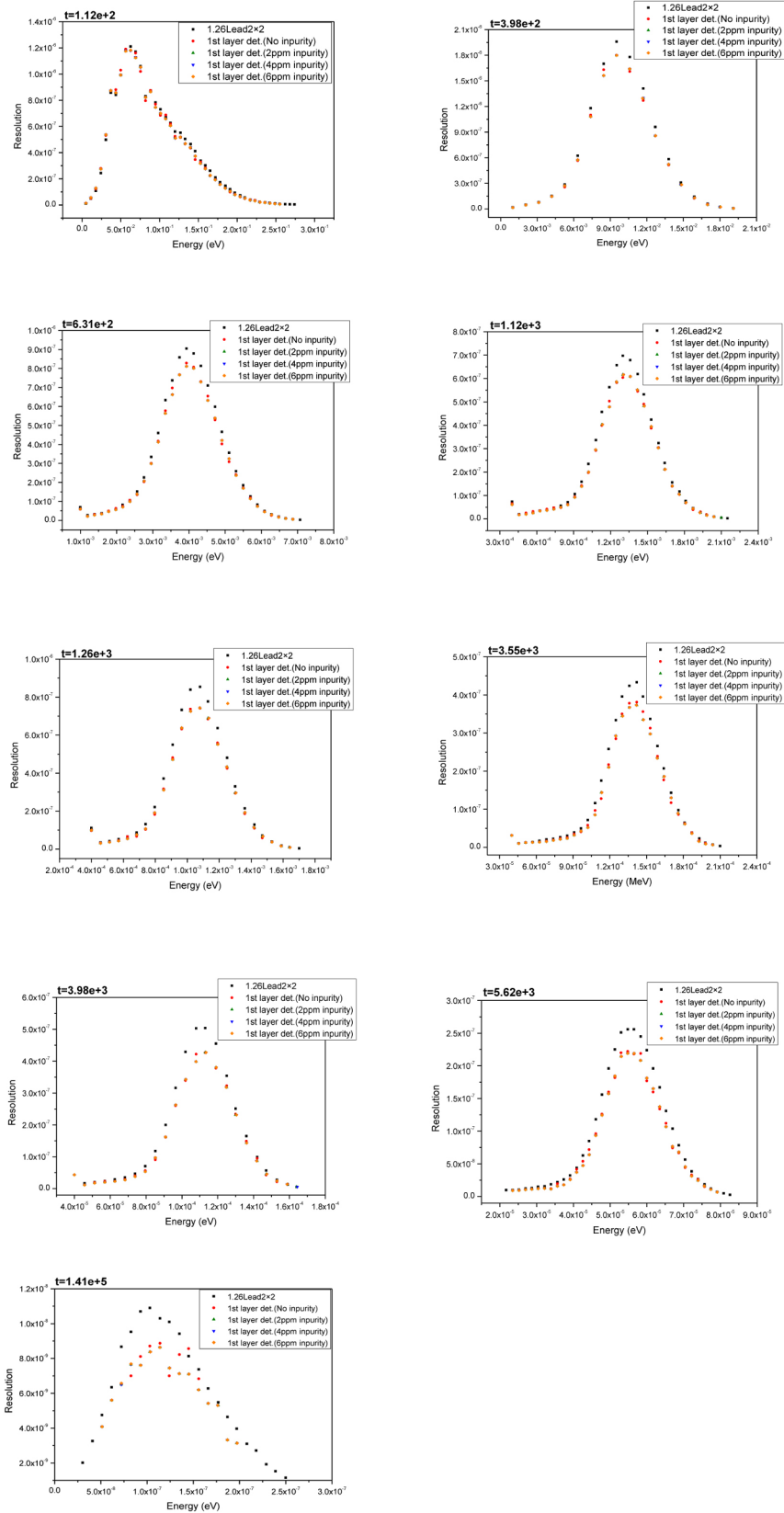
Fig. 2. Neutron Spectrum Effect by Detector Existence in the Lead (1st Layer Detectors).

lead. The energy spectrum inside the fuel is distorted by strong absorption and fission reactions by fissile and fertile materials. The energy resolution was investigated from 0.1eV to 100keV. Fig. 4 show the energy resolution of fuel when the fuel is in the assay area. From 1eV to 10keV, the result is very similar to that of lead, even though there is a slight fluctuation. The fluctuation coms from the interaction with nuclear materials existing in the fuel assay area and the absorption of neutrons at the resonance energy, mainly by the plutonium and uranium.

Regarding other elements in the medium affecting the neutron spectrum, light elements, such as carbon, hydrogen, and copper distort the spectrum. In the simulation, hydrogen(1~10ppm), carbon(10ppm), and silver(10ppm) were selected. Fig. 5 shows the energy resolution effect due to the different impurity material and content. In the figure, carbon and silver do not affect the resolution much, but, for the hydrogen case, the resolution is highly influenced by the content. Therefore, for hydrogen, the content must be limited to below 5ppm to obtain a better resolution.

Some fission products in spent fuel or pyro processed material have a large neutron absorption property. Such a large absorption has an effect on the spectrum distortion and resolution. Therefore, the selected fission products are inserted into the fuel area. Table 1 shows the concentration of fission products in grams per ton for the simulation of the pyro material. Fig. 6 shows the resolution result for fuel alone and fuel with fission products. The resolution is almost the same as that of the fuel alone in the whole energy range. Therefore, the effect of the fission products is small enough to be neglected.

2.3 Fast Fission Neutron Property

The fission property of isotopic fissile material was investigated with respect to the slowing down source neutron energy [7]. The calculation was based on 1wt% of U235, Pu239, and Pu241. The fission event is directly detected at the surrounding threshold detectors. The detected signal has a direct relationship to the content of isotopic fissile material. Fig. 7 shows the fission probability of U235, Pu239, and Pu241 per incident source
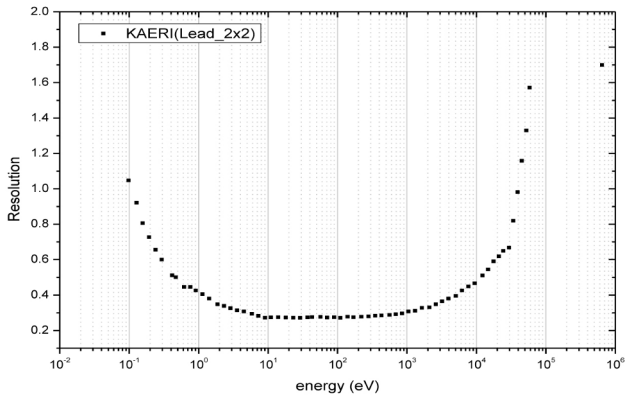

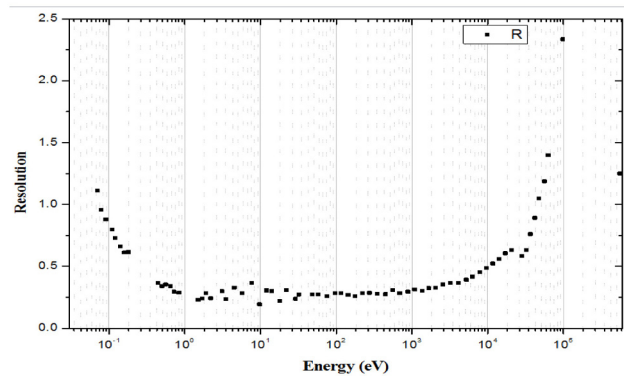Fig. 3. Resolution at Slowing Down Neutron Energy in the Lead.

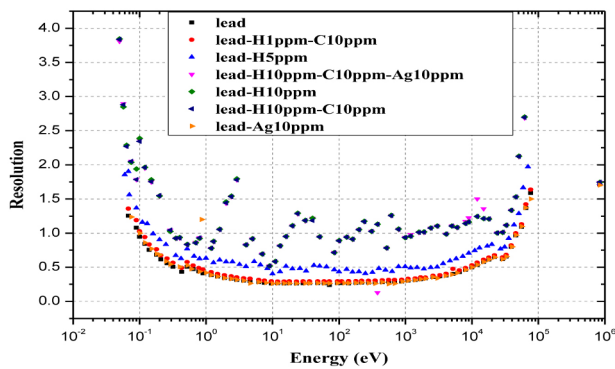Fig. 4. Energy Resolution in the Fuel Area.
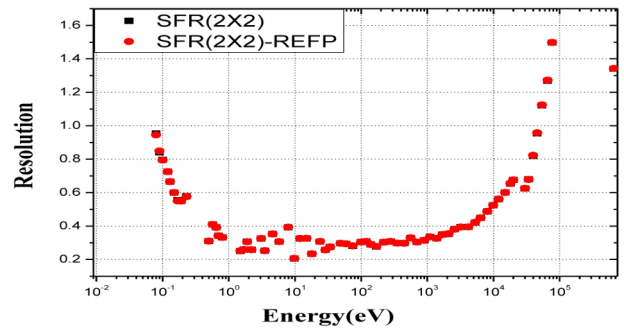

Fig. 5. Energy Resolution Effect by Light Material.

Fig. 6. Energy Resolution Effect with and without Fission Products.

Table 1. Fission Products in Pyro Processed Material

Nuclei	Content(gram) per 1ton
Pr141	1.40E3
Nd143	8.88E2
Nd145	7.97E2
Sm147	3.23E2
Sm149	4.43E0
Sm150	4.94E2
Sm151	2.04E1
Sm152	1.90E2
Eu153	2.03E0
Gd155	5.99E0
Gd157	1.70E-1
Dy160	4.78E-1
Dy161	4.82E-1
Dy162	3.59E-1
Dy163	3.11E-1
Dy164	7.00E-2
Er166	3.87E-2
Er167	1.32E-3

neutron. The fissile isotope fission inside the fuel area is calculated as

$$\int_{\Delta t} \int_0^{\sim} \int_{Volume} \sigma_{fi}(E)\emptyset \quad (r, E, t)dr, dE, dt, \qquad (2)$$

where $\sigma_{fi}(E)$ is the fission cross section for isotope i and $\emptyset$ is the neutron flux at the volume. The integration is taken over Δt, time bin. Above 1keV, there is no special fission structure to distinguish fissile materials. The individual fission characteristic appears below 1keV. Especially, the figure shows the dominant fission property at 0.3eV by Pu239. However, below 0.1eV, the dominant fission structure is not obtained. Therefore, the resonance energy region is proper to get the isotopic fission characteristics.

A linear detection model was set up over the slowing down neutron energy to get each fissile material content as below:

$$\mathbf{Y_{det} = a*X_{U235} + b*X_{Pu239} + c*X_{Pu241}}. \qquad (3)$$

Where $\mathbf{Y_{det}}$ is a detector response function. $\mathbf{X}$ represents

the content of U235, Pu239, and Pu241, and **a**, **b**, and **c** are the reference contribution parameters of the detectors. This model assumes that no significant self shielding is present and that the interrogation neutron flux does not vary much over the assay region. Therefore, the source neutron intensity and detector signal by considering their characteristic resonance fission energy is very important in the final fissile assay.

2.4 Detection Sensitivity

In order to get more fissile fission neutrons effectively, the detection sensitivity was tested for 3 layers. The first layer is located 1cm above the fuel assay area. The second and third layer is placed 1cm above the first and second layer. Fig. 8 represents the configuration of the detector layers. A U238 fission chamber was used in the calculation [7]. The detection is expressed as

$$\int_{S_{det}} \int_{E_1}^{E_2} \sigma_f \emptyset_f (r, E, t)dEdA, \qquad (4)$$

where σ_f is the fission rate at the detector, φ_f is the fission neutron arriving at the detector, and S_{det} is the detector area. The detection efficiency is summarized in Table 2. The table shows a ~50% decrease in the second layer compared to the first layer detection. In addition, a ~45% decrease occurs in the third layer compared to the second layer. The sum of first and second layers has a 85% detection probability in total accumulation. Therefore, the first and second layer detectors are good at obtaining the fission signals.

Generally, the U238 fission chamber involves U235 impurity. The impurity makes an unwanted fission signal by interaction with the source neutron. The unwanted signal has an influence on the accuracy decrease of the
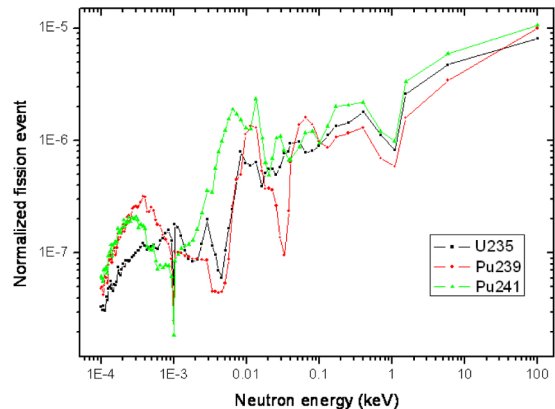
Fig. 7. Isotopic Fissile Fission Property with Respect to Neutron Slowing Down Energy.

fissile assay. Therefore, to avoid such an unwanted signal, a Thorium chamber is considered as a replacement. Thorium also has a threshold property to detect the fast fission neutrons from fissile fission. Table 2 shows the normalized detection probability on U238 and Th232 at the six surrounding detectors as shown in Fig. 1 for an actual detector system. In the table, the detection sensitivity of Th232 is about 76% that of U238. Therefore, if Th232

Table 2. Normalized Detection Probability at Each Layer

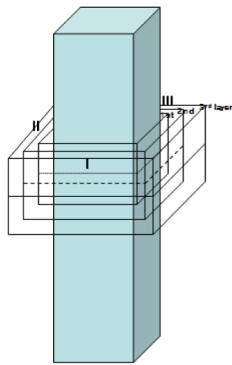| Position	Detection	Detection probability	Accumulation
1st layer	2.78741E-4	58.94%	58.94%
2nd layer	1.24257E-4	26.27%	85.21%
3rd layer	6.98457E-5	14.77%	100%

Fig. 8. Detector Configuration for 3 Layers.

is selected, a large number of detectors or a longer detention time is required to get sufficient detection statistics.

2.5 Photo Fission Effect

For source neutron generation, an intense and high energy gamma ray is produced in the target by (e, γ) reaction. The photon is slowed down and shielded in the lead medium. However, some portion of the photons arrives at the fuel area. When the photon energy arriving at the fuel is greater than the photo fission threshold, photon fission occurs and produces neutrons. These neutrons are detected by the surrounding detectors as an unwanted fission signal. Therefore, a photo fission effect was calculated by varying the distance from the target: 10, 20, 30, and 40cm. Fig. 9 shows the normalized photo fission rate with the different distances. When the results are compared with the fissile fission event by the source neutron, the photo fission effect is small enough to be neglected, as shown in Fig. 7.

2.6 Target Neutron Yield

An electron linear accelerator is under development to produce high neutron yield effectively. A compact, easy maintenance and low cost system was considered. To produce neutrons, various electron energies, currents and pulse widths were examined. For efficiency, electron energies from 15MeV to 40MeV were considered_[12]. The total number of electrons is calculated based on the beam current(C), pulse width(W) and repetition rate(R).

$$N_{electron} = C(Amp) \times W(sec) \times R(Hz) \times 6.242E18 \quad (5)$$

From the simulation[7], ~E12 n's/sec was obtained from 25MeV electron energy. Table 4 shows a summary of the neutron production rate by different incident electron en-

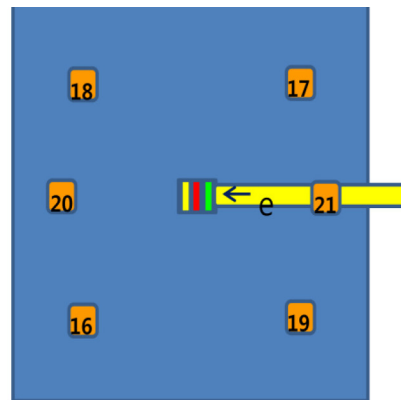| Cell No.	10cm	20cm	30cm	40cm
16	5.32E-09	2.27E-11	3.06E-11	4.11E-12
17	7.07E-11	5.80E-12	6.35E-12	1.96E-11
18	5.29E-09	2.82E-11	6.44E-12	2.52E-12
19	7.20E-11	1.11E-11	2.54E-11	4.25E-12
20	7.28E-08	1.76E-10	4.86E-12	6.38E-12
21	8.35E-11	1.72E-11	1.20E-11	9.81E-12

Fig. 9. Photo Fission Event at Fuel Area with Different Distances From Target.

Table 3. Normalized Detection Probability for U238 and Th232

Detector	E(MeV)	Normalized neutron flux	U238 detection	Th232 detection
D2	1.00E-01	5.26E-04	6.68E-08	0.00E+00
	1.00E+00	1.49E-03	2.48E-06	9.93E-08
	5.00E+00	6.37E-04	2.22E-04	5.23E-05
	1.00E+01	3.07E-05	2.17E-05	6.78E-06
D3	1.00E-01	5.16E-04	5.43E-08	0.00E+00
	1.00E+00	1.50E-03	2.43E-06	9.74E-08
	5.00E+00	6.35E-04	2.22E-04	5.22E-05
	1.00E+01	3.04E-05	2.14E-05	6.40E-06
D5	1.00E-01	5.21E-04	5.47E-08	0.00E+00
	1.00E+00	1.49E-03	2.43E-06	9.97E-08
	5.00E+00	6.25E-04	2.18E-04	5.09E-05
	1.00E+01	2.97E-05	2.12E-05	6.51E-06
D6	1.00E-01	5.17E-04	7.37E-08	0.00E+00
	1.00E+00	1.50E-03	2.46E-06	9.65E-08
	5.00E+00	6.29E-04	2.19E-04	5.08E-05
	1.00E+01	2.93E-05	2.05E-05	6.24E-06
D8	1.00E-01	5.23E-04	6.94E-08	0.00E+00
	1.00E+00	1.47E-03	2.39E-06	9.48E-08
	5.00E+00	5.80E-04	1.99E-04	4.64E-05
	1.00E+01	2.73E-05	1.90E-05	5.67E-06
D9	1.00E-01	5.25E-04	6.78E-08	0.00E+00
	1.00E+00	1.52E-03	2.51E-06	1.01E-07
	5.00E+00	6.59E-04	2.31E-04	5.46E-05
	1.00E+01	3.18E-05	2.26E-05	6.99E-06

Table 4. Neutron Yield Rate by Incident Electron Energy

Energy (MeV)	Neutron production per one electron (n's/cm^{2}sec)	Converted Neutron intensity (n's/sec)
15	1.90E-07	2.68E+11
20	6.42E-07	9.07E+11
25	1.18E-06	1.67E+12
30	1.70E-06	2.40E+12
35	2.14E-06	3.02E+12
40	2.77E-06	3.91E+12

ergies. Therefore, with a one section linac, ~E12 n's/sec is available.

2.7 Target Activation Analysis

In the intense radiation field, for example, the induced fissile fission, source neutron generation, and spontaneous emission from spent fuel, target, structure material and lead are activated for long term usage. The simulation was conducted on the Ta, W, and Pb major materials [13]. The neutron intensity for irradiation was 2.0E13n's/sec for a 10hr exposure. Table 5 shows the isotopic activation with respect to irradiation time. In Table 5, after 10hrs irradiation, 1.1E4Ci was obtained for all Ta isotopes. For the W material, it was 2.15E3Ci. Lead is the major medium material for spectrometer. However, the lead activation was much less than that of Ta and W. Fig. 10 shows the decay heat change for Ta, W, and Pb by the irradiation time.

fissile has its fission characteristics in the neutron energy range of 0.2eV to 1keV. In that energy range, the signature is very proportional to each fissile content. The Th232 detector is a possible replacement for U238 because of the threshold property. However, an intense neutron source is required to get enough detection statistics. A linear electron accelerator system is a good choice to get intense source neutrons. A 25MeV one section electron accelerator was able to generate ~E12 n's/sec in the simulation.

Finally, the LSDS system will play an important role in pyro process fuel cycle development for the approval of fissile material content, fuel safety and economics. Moreover, by approving the isotopic fissile content technology, it will increase the proliferation resistance of the fuel cycle and provide transparency and credibility of reutilization of the nuclear materials.

3. RESULTS AND CONCLUSION

A lead slowing down spectrometer is under development at KAERI to directly analyze the isotopic fissile content in recycled fuel. The threshold fission chamber makes it possible to select fissile fission neutrons from the spent fuel background. Several calculations were performed to setup the optimized LSDS system. The shape of the source neutron spectrum was consistent in cases with the detector present and without it. However, the peak amplitude decreases when the detector is introduced.

The energy resolution was around 0.5 at 0.3eV, at which Pu239 has a big fission resonance. Below 0.3eV, the resolution increases drastically. A linear assay model was set up for U235, Pu239 and Pu241. No significant self shielding was assumed. From the simulation, each
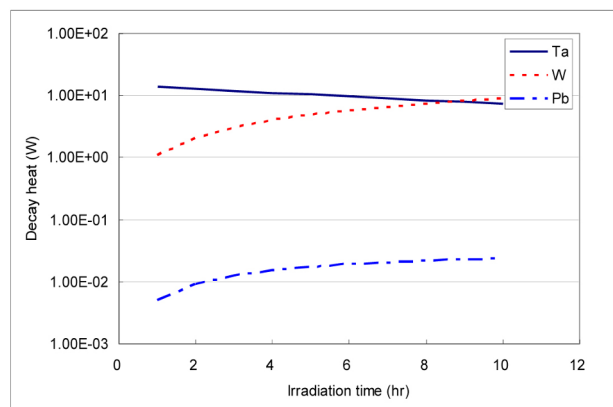

Fig. 10. Decay Heat by Exposure Time.

Table 5. Activation Analysis on Major Material in Curies (Ta, W, Pb)

Mat	Time (hr)	1.00E+00	2.00E+00	5.00E+00	7.00E+00	1.00E+01
Ta	Ta180	2.34E+04	2.15E+04	1.67E+04	1.41E+04	1.09E+04
	Ta182	9.06E+00	1.81E+01	4.53E+01	6.33E+01	9.04E+01
	Ta182m	1.64E+01	1.76E+01	1.77E+01	1.77E+01	1.77E+01
	W183m	3.72E-02	1.48E-01	9.22E-01	1.80E+00	3.65E+00
	Ta183	3.72E-02	1.48E-01	9.22E-01	1.80E+00	3.65E+00
	Total	2.34E+04	2.15E+04	1.67E+04	1.41E+04	1.10E+04
W	W187	2.38E+02	4.70E+02	1.13E+03	1.53E+03	2.10E+03
	W183m	5.26E+01	5.26E+01	5.26E+01	5.26E+01	5.26E+01
	W185m	1.69E+00	1.69E+00	1.69E+00	1.69E+00	1.69E+00
	W185	1.42E-01	2.84E-01	7.10E-01	9.94E-01	1.42E+00
	Total	2.93E+02	5.24E+02	1.18E+03	1.59E+03	2.15E+03
Pb	Pb209	4.33E+00	7.83E+00	1.48E+01	1.75E+01	1.99E+01
	Bi210	1.89E-11	1.43E-10	1.93E-09	4.84E-09	1.24E-08
	Pb205	1.30E-11	2.60E-11	6.50E-11	9.11E-11	1.30E-10
	Total	4.33E+00	7.83E+00	1.48E+01	1.75E+01	1.99E+01

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