New Approach in Fissile Assay Using LSDS – 17076

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ABSTRACT
A quantitative assay of isotopic fissile materials (U235, Pu239, Pu241) was conducted at the Korea Atomic Energy Research Institute (KAERI), using a lead slowing down spectrometer (LSDS). Using a uranium rod and plutonium, the actual measurement of induced fission neutrons was performed in the RPI (Rensselaer Polytechnic Institute) LSDS facility. In the simulation, a content of U235, Pu239, and Pu241 was changed. The detected signal has a direct relationship with the mass of the existing fissile isotopes. The mathematical model for fissile assay was applied to the measured and simulated detection data. The assay results show that the isotopic content was consistent with 2 to 3% uncertainty for Pu239. The simulation indicated that the assay result was affected by the self-shielding correction. Usually, in highly enriched plutonium, the correction plays an important role and good understanding of it is required in order to increase assay accuracy. The combination of LSDS technique measurement and simulation is a very powerful and direct way to analyze the isotopic fissile content. The information provided by such analysis is applicable to nuclear fuel cycle development and spent fuel management for safety and economics.

INTRODUCTION
An isotopic fissile content assay is very important for reuse of fissile materials through the fuel cycle and management of spent fuel. The accumulation of spent fuel is currently a significant issue in Korea. The storage capacity for spent fuel will reach to the maximum soon. For the optimum design of spent fuel storage site, a fissile content is one of the important factors to be given to obtain the maximum burn credit. In addition, accurate data for fissile content is basic information to be provided for increased safety and economics in the fissile reuse. The pyro processing technology is under development at KAERI, as one option of the nuclear fuel cycles, which increases the proliferation resistance. The pyro process plays a role in the volume reduction of storage for spent fuel and produces the source material including trans-uranium to fabricate a fuel rod for a sodium fast reactor (SFR). An energy production system through a nuclear power plant contributes to a carbon dioxide reduction. Therefore, a reutilization of fissile material by the fuel cycle contributes to not only the energy production but also the environment. Many nondestructive methodologies have a limitation in the direct assay of isotopic fissile content for spent fuel. The LSDS is a feasible technology to analyze the content of isotopic fissile material in spent fuel or pyro produced material. Based on the designed LSDS device [1], the neutron induced fissile fission was measured at the surrounding detectors. The detector signal was used to analyze the content of isotopic fissile materials. However, to obtain a high level of assay accuracy, the

self-shielding must be examined in the nuclear fuel area [2]. In the simulation, the scope of the plutonium content assay was determined without a self-shielding correction. Moreover, an actual induced fission measurement was conducted in the RPI LSDS facility using uranium and plutonium sample.

FISSILE CONTENT ASSAY

The isotopic fissile content assay was performed based on the experimental measurement and simulation data. In the experiment, RPI LSDS system was used and the uranium fuel rod (U235 4.8wt%) and plutonium (PuBe 47gram and 91gram) sample were inserted into the assay area and the induced fission neutron was measured. In the simulation, the contents of uranium and plutonium were changed and the sensitivity on the fissile content assay was examined. A linear detection model was setup to obtain the content of the isotopic fissile material [3,4]. The detector signal involves information of the fission of U235, Pu239, and Pu241 by the interrogation source neutron. For neglecting self-shielding, the assay model has a linear relation between the detector signal and fission rate. The model is expressed as below,

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

where y_i is the detector signal at slowing down time channel *i*, k is a normalization constant, and ϵ is the detector efficiency. The numbers 1, 2, and 3 represent the fissile materials, U235, Pu239, and Pu241 respectively and N is the fissile material mass. In addition, ν is the average prompt fission neutron yield of each fissile material, σ_f is the fission cross section, and ϕ is the neutron intensity entering into the fuel.

Measurement

The source neutron slows down in the lead medium and enters the fuel area to induce fission with respect to the neutron energy. The fast fission neutrons are measured using surrounding threshold fission chambers that discriminate the fast neutrons from the complex radiation field (for example, intense gammas, source neutron, spontaneous fission neutron, and (α, n) reaction).

Fig. 1. LSDS Equipment for Fissile Fission Measurement in RPI.

Figure 1 shows the LSDS system involving a lead spectrometer, electron beam-line, and the fuel and detector assembly. Figure 2 illustrates the location of the uranium rod and plutonium in the assembly. Two different plutonium (Pu239) contents were used in the measurements : 47g and 91g. The uranium fission chamber is located above the fuel assembly, and the thorium fission chamber was also located as shown in the figure.

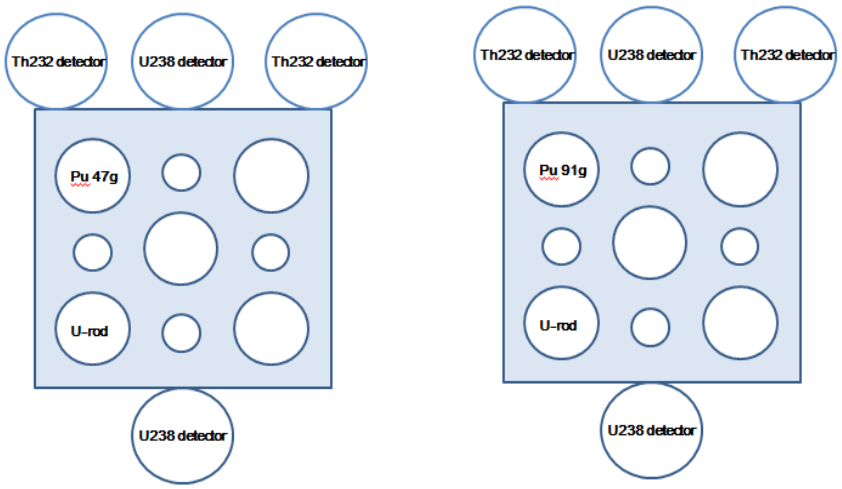

Fig. 2. Assembly Geometry for The Fuel Rod and Plutonium Position.

Figure 3 shows the detected signal at the uranium fission chamber with respect to the neutron slowing down time. In the graph, the dominant fission property was shown well around 850μsec by Pu239 fission. Tables I and II show the assay result on uranium and plutonium from the measurements. For uranium, the assay result shows a good agreement with the actual content with less than a 2% error. The plutonium content was analyzed to have about a 1% error rate compared with the actual amount. Therefore, from the actual fission measurement, the uranium and plutonium could be directly assayed with 1 to 2% uncertainty in the LSDS system.

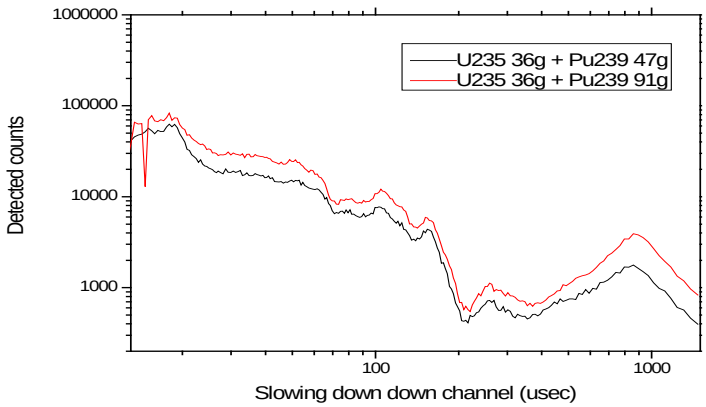

Fig. 3. Induced Fissile Fission Measurement at The Uranium Fission Chamber.

TABLE I. Uranium and Plutonium Assay (U235: 36g, Pu239: 47g).

U235(g)	Fitting error	Pu239(g)	Fitting error	Pu241(g)	Fitting error
36.82	1.77E-5	46.26	7.45E-6	-1.20	3.02E-6

TABLE II. Uranium and Plutonium Assay (U235: 36g, Pu239: 91g).

U235(g)	Fitting error	Pu239(g)	Fitting error	Pu241(g)	Fitting error
36.13	2.23E-5	90.40	8.62E-6	-2.89	3.91E-6

Simulation

In the designed LSDS system, the fission measurement was simulated by changing the content of uranium and plutonium, as shown at figure 4. In the simulation, Pu239 was mainly changed from 0.5% to 5%. However, for U235 and Pu241 content, 0.5% and 1% were used. From the simulated fission measurement, an isotopic fissile assay was performed in the fuel rod for U235, Pu239 and Pu241. The detection signal in each channel is the sum of the fission by U235, Pu239 and Pu241. Table III shows a summary of the content assay result. For 0.5% of uranium and plutonium, a good agreement was obtained.

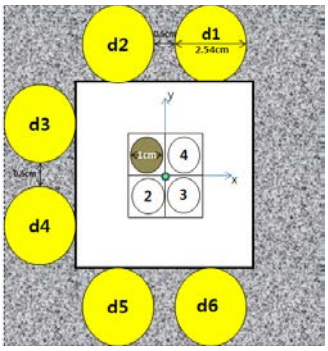

Fig. 4. Induced Assembly Geometry for Fuel Rod Fission Simulation.

For a change of 1 to 3% of Pu239, the assay was obtained with 1% uncertainty comparing to the actual content. However, for Pu241, a relatively increased error was obtained, but the assay content was very close to the actual amount. For the 5% enriched Pu239 case using uncorrected detected data for self-shielding, the assay results show that a relatively large uncertainty was obtained. However, when the correction is applied, the result shows a good agreement with the actual

content. Therefore, in the induced fission measurement, the correction must be applied for the highly enriched fissile content.

TABLE III. Assay Sensitivity on Different Uranium and Plutonium Content.

1) U235(0.5%), Pu239(0.5%), Pu241(0.5%)

U235(%)	Fitting error	Pu239(%)	Fitting error	Pu241(%)	Fitting error
0.51	0.1	0.53	0.05	0.47	0.04

2) U235(1%), Pu239(1%), Pu241(1%)

U235(%)	Fitting error	Pu239(%)	Fitting error	Pu241(%)	Fitting error
0.99	0.005	0.99	0.003	1.05	0.008

3) U235(1%), Pu239(3%), Pu241(1%)

U235(%)	Fitting error	Pu239(%)	Fitting error	Pu241(%)	Fitting error
0.99	0.12	2.97	0.08	0.94	0.07

4) U235 (1%), Pu239(5%), Pu241(1%) (self-shielding uncorrected)

U235(%)	Fitting error	Pu239(%)	Fitting error	Pu241(%)	Fitting error
0.84	0.09	4.7	0.07	0.81	0.04

5) U235 (1%), Pu239(5%), Pu241(1%) (self-shielding corrected)

U235(%)	Fitting error	Pu239(%)	Fitting error	Pu241(%)	Fitting error
0.97	0.13	4.98	0.1	1.05	0.07

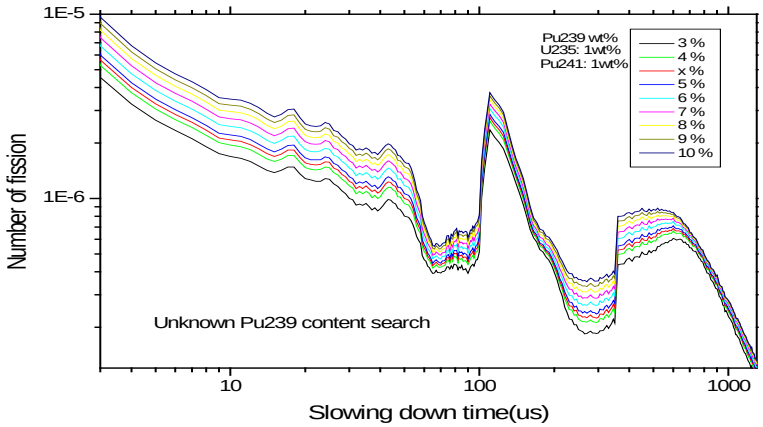

Fig. 5. Fission Location of Unknown Pu239.

Based on the assay model [4], using the assay results by changing the content of Pu239, the fission measurement for an unknown Pu239 content (actually 4.5%) was also examined. The fission event of the unknown sample was located somewhere between 1% and 10% of Pu enrichment. Figure 5 shows the fission signal with respect to the slowing down time from 1% to 10% of Pu239 enrichment, including the enrichment of the unknown Pu239 sample (4.5%). As shown in the figure, the fission signal by the unknown sample was located between 4% and 5% of Pu239. Table IV shows the assay result on the unknown Pu239 sample. The result shows very good agreement with the actual content. From the assay result, the isotopic fissile content can be obtained within 2% error compared to the actual content of Pu239.

TABLE IV. Content Assay of The Unknown Pu239 (Actual: 4.5%, The content of U235 and Pu241 fixed to 1%)

U235 (%)	Fitting error	Pu239 (%)	Fitting error	Pu241 (%)	Fitting error
1.02	0.10	4.58	0.04	0.99	0.02

RESULTS AND CONCLUSION

In the LSDS system, the induced fissile fission measurement was performed using uranium and plutonium materials. A mathematical assay model was applied to obtain the uranium and plutonium content. In the isotopic fissile content analysis, the correction for self-shielding shows a very important factor to represent the linear response with respect to the content variation. The applied correction improves the assay accuracy on highly enriched fissile material.

The isotopic fissile assay results show very good consistency with the actual mass for uranium and plutonium. In particular, the content for Pu239 matched well with the actual content, even for the unknown content. The assay shows ~1%

uncertainty for the Pu239 assay in the actual measurement. The assay was in good agreement with the U235 content, with a ~2% error. From the assay sensitivity result, the effect of the self-shielding correction was negligible until 3% of Pu239. However, for spent fuel assembly assay, the correction must be applied even though the content of plutonium is small.
From the actual measurement and simulation, the LSDS technique is very promising for an analysis of the isotopic fissile content in spent fuel. In the LSDS, direct fissile fission is obtained in the surrounding detectors. Therefore, the LSDS is very powerful and direct in isotopic fissile assay. The LSDS is applicable for the nuclear fuel cycle and spent fuel management for safety and economics.

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