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Sensitivity simulation on isotopic fissile measurement using neutron resonances

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ABSTRACT

Uranium and plutonium are required to be accounted in spent fuel head-end and major recovery area in pyro-process for safeguards purpose. The possibility of neutron resonance technique, as a non-destructive analysis, was simulated on isotopic fissile analysis for large scale process. Neutron resonance technique has advantage to distinguish uranium from plutonium directly in mixture. Simulation was performed on U235 and Pu239 assay in spent fuel and for scoping examination of assembly type. The resonance energies were determined for U235 and Pu239. The linearity in the neutron transmission was examined for the selected resonance energies. In addition, the limit for detection was examined by changing sample density, thickness and content for actual application. Several factors were proposed for neutron production and the moderated neutron source was simulated for effective and efficient transmission measurement. From the simulation results, neutron resonance technique is promising to analyze U235 and Pu239 for spent fuel assembly. An accurate fissile assay will contribute to an increased safeguards for the pyro-processing system and international credibility on the reuse of fissile materials in the fuel cycle.

1. Introduction

The pyro-process was under development at KAERI(Korea Atomic Energy Research Institute) to recover and reuse fissile materials in spent fuel and to reduce storage burden of spent fuel [1]. The recycling option of nuclear material in spent fuel is pyro-process technology linked with burning of recovered actinide materials in a sodium fast reactor (SFR) [1,2]. In the process, the nuclear material balance area (MBA) including spent fuel input, main process for recovering nuclear materials and final TRU product was already setup [1]. The nuclear materials produced at each process has a different material composition. Uranium and plutonium co-exists in the head-end process and it has intense radiation background.

The key measurement points (KMPs) in each MBA were determined for the process safeguards purpose [1]. Fissile content must be approved from the spent fuel introduction into the process to the final products to keep the COK(continuous of knowledge) in nuclear material flow. At each KMP, chemical analysis (DA) was a basic technique for the analysis of nuclear material [1]. However, non-destructive assay (NDA) techniques are also considered for engineering scale of pyro-process to get effectiveness and time-saving in analysis of fissile materials.

Several non-destructive technologies were evaluated for uranium and plutonium assay in spent fuel [3–5] and pyro-processed material [4,6], and applicability was simulated as well, based on the direct discrimination of uranium and plutonium [3,4,6–9]. For fissile assay in spent fuel, because of intense neutron and gamma emission, a direct measurement of fissile material is very restricted. In the evaluation, neutron resonance transmission method was one option to assay isotopic uranium and plutonium, among non-destructive techniques [4,9–15]. In spent fuel, many fission products and actinide are produced. However, fission products have very weak resonances under 50eV, for example, Cs, Sm, Xe, Nd, and Tc. Fortunately, oxygen(in UO_2) and zirconium(cladding material) have no resonances in that energy region [15]. Therefore, few interference in resonance transmission occur in measurement [15]. Moreover, there is no interference from background, mainly by neutron emitted from Cm244.

The major advantage of neutron resonance technique is using inherent resonance property of fissile and measured signal can be directly discriminated for uranium and plutonium. However, the

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linearity between measurement and fissile content must be evaluated. Each fissile material, U235, U238, Pu239, and Pu241 in this case, has different and distinguished neutron absorption properties with respect to neutron energy in resonance region. The measurement has a correlation with the content of the fissile materials. In the head-end process, the applicability of neutron resonance technique was simulated for uranium and plutonium assay in spent fuel. The simulation was on the neutron transmission and energy dependent neutron spectrum analysis. The linearity was also evaluated on the increase of fissile material content at resonances energies. Applicable resonance energy finding was key issue to represent linear response when fissile content increased. By adding additional fuel rows in assembly type, scoping test was analyzed to determine measured signal reliability and number of rods to be analyzed in one row. Additionally, low level detection limit (LLDL) was examined on the different sample property and capability on different sample content and composition for actual application in the process, because various outputs involving different nuclear materials are also produced in the process.

NDA technology development for an accurate fissile assay is a challenging area to apply for various fields, including re-use and management of nuclear spent fuel, especially for the pyro-process. Additionally, a technology improvement on fissile content assay will play an important role in international transparency and credibility in the pyro-process.

2. Neutron transmission measurement

2.1. Basic principle

Fissile materials have several reactions with incident neutron depending on its energy; fission, absorption and scattering. Particularly, Uranium and plutonium isotopes have their narrow and big absorption property below keV energy, called neutron resonance. Therefore, their resonance energies can be used for obtaining transmitted neutron signal for uranium and plutonium, because they have their own distinguished energy. The measured signal has the relationship with the content of uranium and plutonium. Generally, the transmitted signal is simply expressed as [4].

$$I = I_o \exp[-\sigma_t\, x] \tag{1}$$

where I_o is an incident neutron flux on sample, I is the transmitted

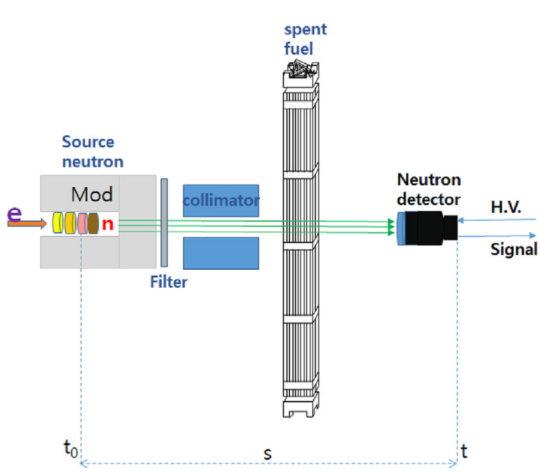


Fig. 1. Conceptual design of neutron transmission measurement (t_o and t are initial and arriving time, s is the distance between source and detector.).

neutron flux, and σ_t is total cross section and x is thickness of sample. The normalized transmitted rate is expressed as

$$I/\,I_o = \exp[-\sigma_t\, x]. \tag{2}$$

Fig. 1 shows the schematic view of neutron source, spent fuel sample and detector geometry for neutron transmission measurement. An intense neutron source is required to transmit spent fuel assembly and low energy (epithermal region) neutron is more favorable to represent dominant resonances for uranium and plutonium isotopes.

Generally, intense neutron is necessary to obtain accurate measurement with less detection error. Electron linear accelerator(LINAC) is recommended with W or Ta target [16,17] to generate neutron. Electron produces bremsstrahlung radiation in target and neutron is produced by $(e,\gamma)\,(\gamma,n)$ reaction [16], having most probable energy around 1 MeV. The energy dependent neutron spectrum is expressed like below

$$\phi(E) \sim \frac{1}{\sqrt{E}} \exp\left(-\frac{E}{T}\right), \tag{3}$$

where E is in MeV and T is target average temperature in MeV. For W and Ta, giant dipole resonance is placed around 15 MeV for gamma production [18]. High current and pulse repetition rate produces intense neutron. The intensity, ~E12 n's/sec, can be obtained with 500 mA and 700 pps(pulse per second) in LINAC and 20–30 MeV electron is available for one section accelerator column. Measurement for several hours is expected in actual transmission experiment.

The energy of source neutron needs to be decreased below keV region by interacting with moderator to get effective reaction with fissile material in spent fuel. The slowed down neutron has advantage in transmission measurement to represent inherent resonance property of fissile. In addition, filter is helpful to cutoff thermal neutron not to occur fissile fission neutron. Moderated neutron is detected at He-3 detector. In the simulation, 1 cm in diameter and 1inch long detector was used. The neutron beam was 1 cm in size and the distance between source and detector was fixed at 2.5 m long. The measurement and spent fuel sample must be placed in hot cell for actual application, therefore, the size of equipment has limitation in hot cell. For spent fuel application, the detected signal must be discriminated from neutron background. The neutron transmission and detection signal was performed using the MCNP code [19]. The detected signal is simply expressed as

$$\varepsilon \int_A\!\!\int_E \varphi\,(r, E, t)\sigma_{nonabs} dEdA, \tag{4}$$

where φ is the source neutron arriving at detector, σ_{nonabs} is the non-absorption cross section, A is the detector area, and ϵ is the detector efficiency.

2.2. Fissile measurement

The possibility of transmission measurement was simulated for isotopic fissile assay of spent fuel [19]. In head-end process, assembly structure is disassembled and fuel rod is extracted from cladding. The content assay of isotopic fissile is required in the process. The simulation was performed on rod and assembly type of spent fuel. In the simulation, the material composition, uranium, TRU, and fission products, was obtained from ORIGEN code [20], with 4.5 % in initial enrichment and 50GWd/MTU burnup and the homogeneous distribution in the fuel rod was assumed. Table 1

Table 1
Major nuclides in spent fuel rod (4.5 % I.E., 50GWd/MTU).

Isotope	content (gram)
U234	2.23E2
U235	8.90E3
U236	6.22E3
U238	9.19E5
Pu238	3.22E2
Pu239	6.27E3
Pu240	2.86E3
Pu241	1.10E3
Pu242	8.37E2
Cm242	3.14E-3
Cm244	6.27E1
Am241	7.34E2
Np237	7.71E2

shows the summary of nuclides used in the simulation. The energy of transmitted neutron was evaluated from eV to 100eV. At low energy region, around tens eV, uranium and plutonium isotopes represent their dominant resonances.

Fig. 2 represents the transmitted neutron signal on U235, U238 and Pu239 as a reference and the spectrum for spent fuel is also shown in the figure. The dominant resonance energies for uranium and plutonium are well shown and the resonances are distinguished each other, based on the rod basis measurement. The differences of measured signal make possible for isotopic fissile assay, for U235, U238 and Pu239. However, the linearity between transmitted signal and content of fissile material must be evaluated for system working.

For application of spent fuel assembly, the sensitivity measurement was simulated [19] and the spectrum analysis was performed at each fissile resonance, before whole assembly was introduced. First of all, by adding up the spent fuel until 5 rod thickness, the detected signal was analyzed for resonance structures and energies. Fig. 3 shows the transmitted detection by rod increase, until 5 cm. The spectrum represents well defined resonance structures at the resonance energies and the total detection decreases in all energies when rod added. The prominent resonances energies were selected for U235 and Pu239 from the spectrum analysis. From the energy dependent measurement, the transmission and absorption rate was obtained at the prominent resonance energies for U235 and Pu239.

Fig. 4 shows the transmitted and absorbed rate at the selected resonance energies for U235 and Pu239. The linearity on transmission and absorption rate was analyzed with respect to the rod increase. As shown in Fig. 4, U235 and Pu239 have various resonance energies at tens eV energy region. The linearity was only satisfied at 26.2eV for Pu239 and at 32eV for U235. At other energies, linearity was not satisfied as rod increase. Therefore, 32eV and 26.2eV for U235 and Pu239 can be used to obtain the content of U235 and Pu239 in spent fuel rods, until 5 cm thickness.

2.3. Scoping test

From the evaluation of sensitivity and linearity simulations on spent fuel rod basis, the possibility of application on assembly type was examined. For the reference assembly, 17by17 geometry was determined for the simulation [19]. The detection geometry was arranged until 17 rod add-up in a row. 0.9 wt% for U235 and 0.6 wt% for Pu239 were used in the simulation with actinides [20], as shown at Table 1. The material distribution in assembly was assumed as a uniform. Fig. 5 shows the transmitted signal when additional rods are introduced above 5 cm rod basis. At each rod increasing case, the resonance structure was also well defined for U235 and Pu239 like previous rod basis case. However, the detected intensity decreased rapidly above 10 cm thickness in all energy range. That's why intense source neutron is required in the measurement.

At the selected resonance energies for U235 and Pu239 (previous rod basis), the linearity was evaluated until 17 cm thickness increase. Fig. 6 shows the transmission and absorption rate at each dominant resonance energies for U235 and Pu239. Pu239 has linear response energy at 26.2eV and at 32eV for U235 in assembly measurement as well. Therefore, the selected and examined energies for U235 and Pu239 can be used for analysis of the content of U235 and Pu239 in spent fuel assembly. However, because of low intensity of neutron at low energy, moderated source is required for assembly type assay to obtain lower statistical error in measurement.

2.4. Interference

The scoping test was examined for assembly type of spent fuel and the applicable number of rods in an assembly was determined. However, on one side, more layers are located around main detection row in an assembly. Therefore, adjacent fuel layers might influence on transmitted detection by scattering or nuclear reaction. In the simulation, 1, 2, 4 layers were added around main row (1 and 2 layers addition at one side of main row) and detection influence was examined in the main row. Fig. 7 shows the simplified geometry of location for source, fuel rod layers and detector. The detector position was fixed along main layer and bare detector was used in the simulation.

The detection increase was obtained in all energies when layers are loaded around the main layer, as shown at Fig. 7. However, for 1 and 2 layer addition, around 5 % detection increase was obtained at the selected resonance energies for U235 and Pu239. For 4 layer addition, around 10 % increase was shown at the selected energies. Except the selected resonance energies, relatively large detection increase was obtained. The scattering might influence on the detection increase at all energies. Therefore, shielding material around bare detector is needed and helpful to reduce the scattered detection by adjacent fuel layers. In addition, for actual application, neutron collimator is suggested around beam path column, after source neutron generation. B_4C can be used as a collimator to protect neutrons of thermal energy region, which have high probability in induced fissile fission for spent fuel. Therefore, collimator will be helpful to reduce unwanted neutron measurement in transmission.

3. LLDL analysis

Neutron transmission technique using dominant and inherent resonance energies has advantage on isotopic fissile assay. In fuel cycle, for example in pyro-process, various nuclear materials are recovered and produced in different composition, density and content at final product, waste and process. For actual application of neutron transmission, capability of the technique in fissile assay, low level for detection, needs to be examined, which still keeps resonance structures in transmission, to obtain credible signal for accurate fissile content. Low level detection limit (LLDL) was simulated by changing the sample thickness(t), density(ρ) and content(m) for U235 and Pu239 material. LLDL provides an information of applicable limit in neutron transmission measurement. In the pyro-process, some sample has highly enriched property,

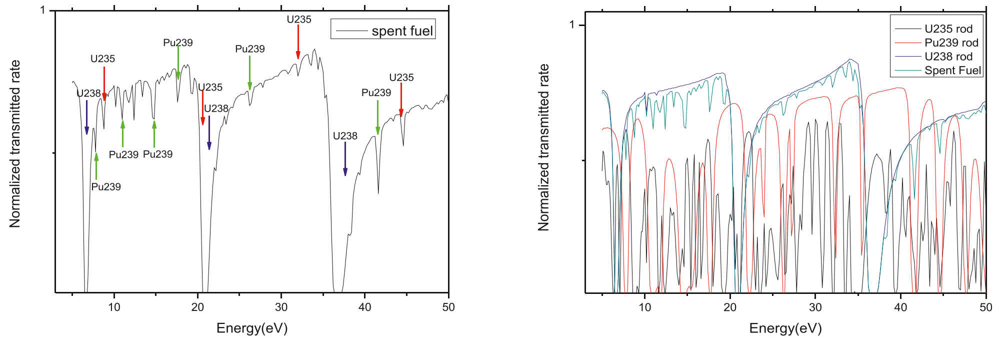


Fig. 2. Characteristics of transmission spectrum for nuclear material (U235, U238, Pu239 and spent fuel).

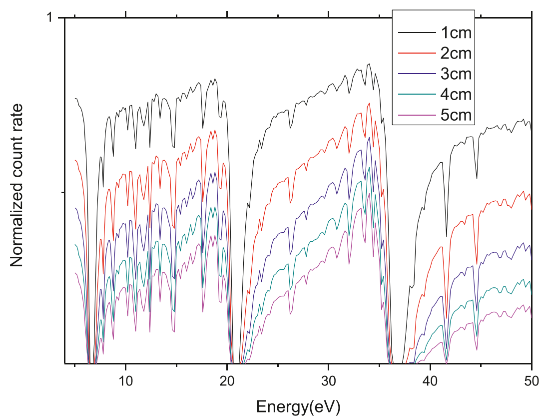


Fig. 3. Neutron transmitted signal by adding up the fuel rods.

however, other samples might have low fissile concentration, like in waste.

The content of U235 and Pu239 was changed from 0.01 % to 0.5 % and the density was changed from 1.0 to 10g/cc. The sample thickness was also changed from 0.1 cm to 1.0 cm. Therefore, the detector response (R_n) in the transmission was defined as a function of density, sample thickness and fissile content.

$$R_n = fn(\rho, t, m) \tag{4.1}$$

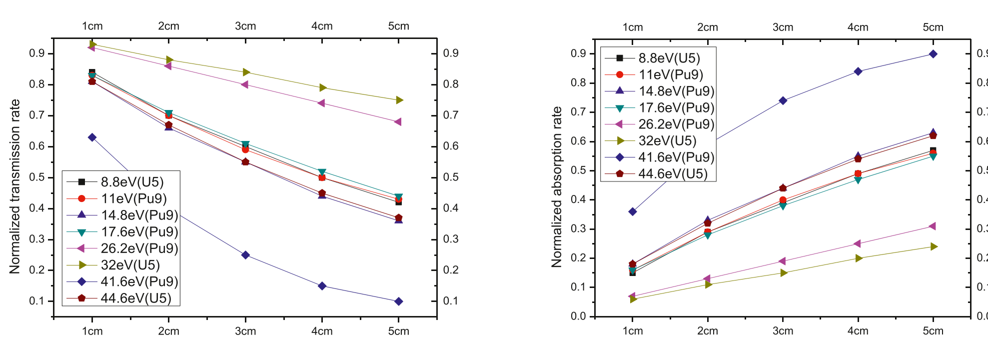
Fig. 8 shows the detection signal by changing sample density, thickness and content. Most of the cases, the results show that the resonance structures are identified as the sample density and fissile content increases. At 1.0g/cc in sample density, when fissile content is less than 0.1 %, resonance depth becomes shallow in all thicknesses. As the sample density decreases, the higher transmission was obtained. However, the resonances were not well distinguished.

At the fixed sample content (0.5 %) of U235 and Pu239, the transmitted signal was evaluated for different sample thickness. Fig. 9 shows the result on the thickness effect by different sample density. From the results, at 0.5 % of U235 and Pu239, the resonance structures were shown for 0.5 cm sample thickness.

4. Neutron moderation

Neutron source is an important factor in fissile transmission measurement. Intense neutron source is required to save detection time and less statistical fluctuation in measurement. Several different types of neutron generation are currently available, for example, D-D, D-T, accelerator driven, nuclear reactor. Neutron production using accelerator is efficient in intensity, however, it has relatively high cost and large scale.

Neutron energy can be detected by time-of-flight(TOF) or average energy in spectrum. The energy is described by measured time in TOF system. Most useable resonance energies in transmission are normally under keV. For spent fuel or pyro-produced material, in which fissile and fertile materials are mixed with fission products, various resonance energies are presented and complicated resonances make difficulty in identification of isotopic fissile materials. In particular, fissile (uranium and plutonium) has identified resonance energies at eV and tens eV region, which are



a) transmission b) absorption

Fig. 4. Transmission and absorption rate at dominant resonance energies by adding up the fuel rods (until 5 rods).

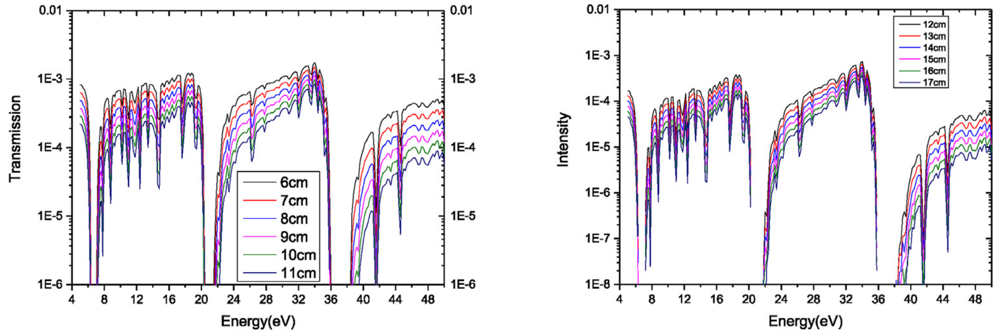


Fig. 5. Energy dependent transmitted detection by rod add (until 17 cm).

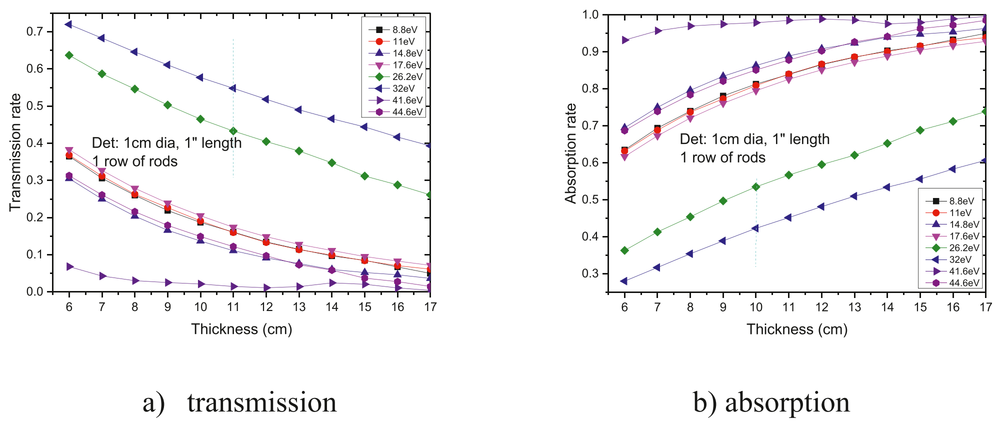


Fig. 6. Transmission and absorption by rod increase in fuel assembly.

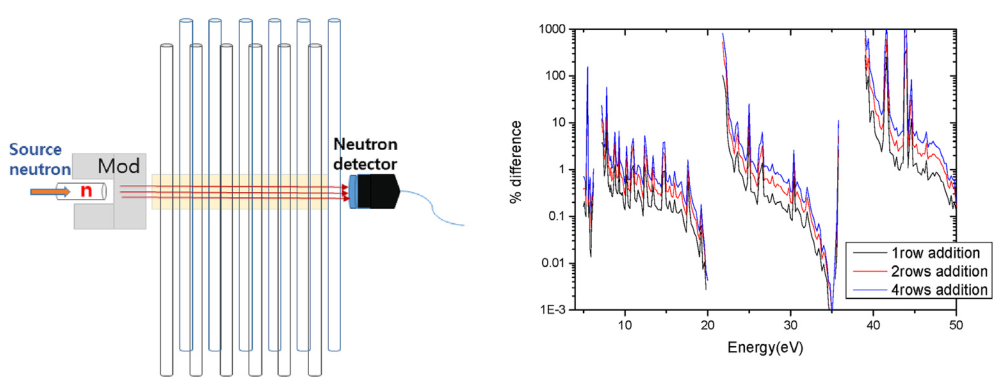


Fig. 7. Geometry for layer detection and detection influence by adjacent fuel layers.

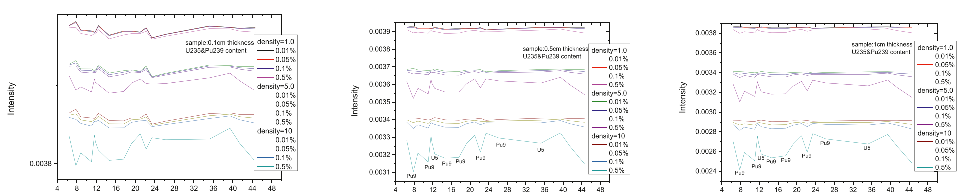


Fig. 8. Detection for U235 and Pu239 with respect to sample thickness (density:1.0–10g/cc, content of U235 and Pu239: 0.01–0.5 %).

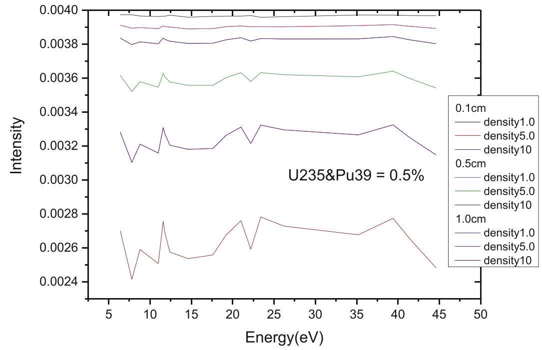


Fig. 9. Detection signal for U235 and Pu239 at 0.5 % (density:1.0–10g/cc, thickness: 0.1–1.0 cm).

very distinguished from those of fission products [4,15].

In spectrum of neutron energy, the region of interest for resonance transmission is placed at tail part of spectrum, longer detection time is generally required to get accurate measured data for low energy region. Therefore, neutron moderation was simulated using several medium materials, as show in the geometry of Fig. 1. The neutron detection was obtained after moderator, passing through the moderator. If properly moderated neutron can be obtained, transmission measurement will be very effective and efficient. In the paper, the moderation was examined at CH2 and graphite medium. Fig. 10 shows energy dependent moderation property with respect to the change of material thickness. In the simulation, the energy range, from 10eV to 100eV, was examined. From the results, at 30 cm thickness of graphite, the highest intensity was obtained in all energy range. For CH2 medium, the highest intensity was obtained at 8 cm thickness. For the selected energy range, the effective moderation was obtained at CH2 with less thickness. Therefore, it has relatively higher moderation power for source neutron. The moderation behavior by flying time was examined as well for the moderators. Fig. 11 shows energy dependent neutron intensity at 5 and 2 μsec moderation time after graphite and CH2 medium, in energy range from 10eV to 100eV. Energy broadening was shown at CH2 moderation. However, the intensity was higher at CH2 medium than that of graphite in all energies.

Gd filter was placed after moderator to cutoff thermal neutrons, below eV region which has highly sensitivity on fissile fission. 3 mm thickness of Gd filter was simulated after CH2 and graphite. Fig. 12 shows neutron cutoff in energy spectrum after 2, 4, 6 cm of CH2 and 10, 20, 30 cm thickness of graphite. For all cases, the results show that the neutron intensity drastically decreases below eV energy by neutron absorption when Gd is placed. Therefore, Gd filter will be helpful to remove induced fission by source neutron in spent fuel.

5. Results and conclusion

Several sensitivity simulations were performed on neutron transmission technique for spent fuel. For large scale of pyro-process, the resonance technique is suggested for the analysis of fissile material content, as an additional way to help chemical analysis. From the simulation results, the transmitted signal could be identified for uranium and plutonium in the mixture and the prominent resonance energies were determined for U235 and Pu239 in spent fuel. The possibility of application was shown for fissile materials in assembly type. From the LLDL simulation, the relation between sample thickness and density was determined and the reasonable fissile content was proposed for better application. However, intense neutron source is required to analyze assembly type of spent fuel with good statistics. In addition, for assembly type application, shielded detector is required to eliminate scattered neutron from the adjacent fuels. Moreover, linearity between measured signal and fissile content change is an important factor for system operation because non-linearity in transmission could be obtained in different sample density, thickness and content for different resonance energies.

The source beam size influences on spatial resolution, rod size, detector and flight tube size. CH2 has good moderation power for interesting assay energy region, around tens eV, and it is effective in neutron slowing down within less thickness. By using Gd filter, the reduction of neutron intensity in thermal energy region was obtained not to interfere the neutron measurement. An induced fissile fission neutron by low energy of source neutron (at thermal and just below epithermal region) could be eliminated. Additionally, B4C collimator is helpful to reduce the fissile fission by adjacent rod layers and to measure transmitted neutrons for real application. The major advantage of neutron transmission measurement is that transmitted signal is very direct on isotopic fissile material. An accurate measurement of plutonium content in the pyro-process will contribute to international nuclear safeguards of the pyro-facilities.

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.

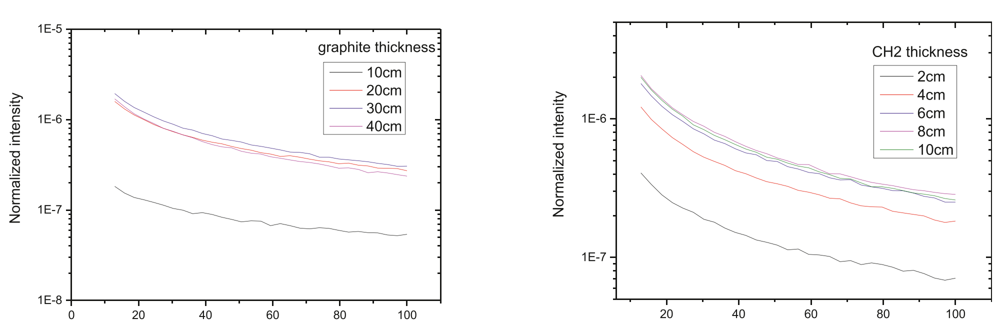


Fig. 10. Neutron intensity by medium thickness (graphite, CH2).

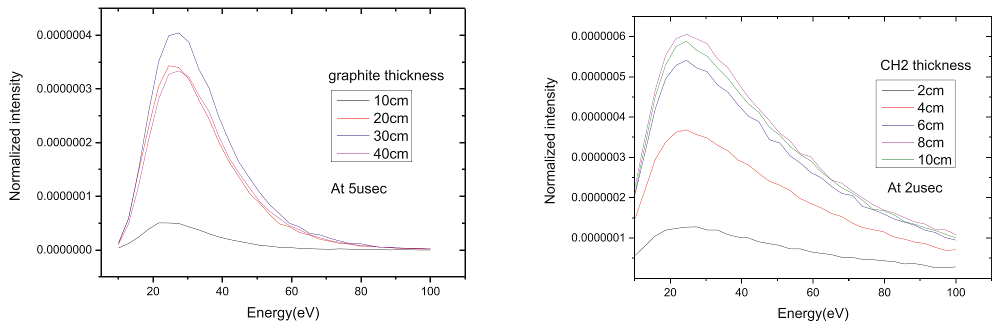
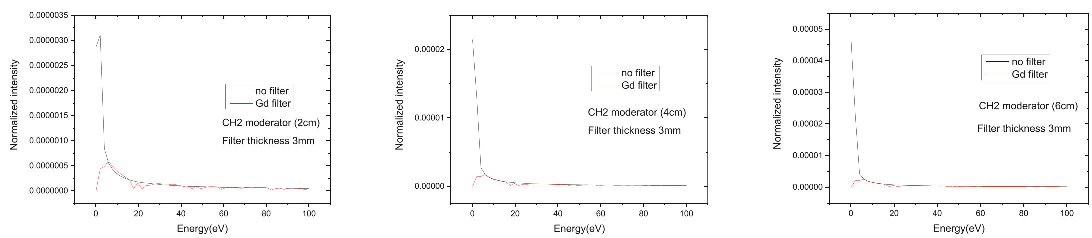
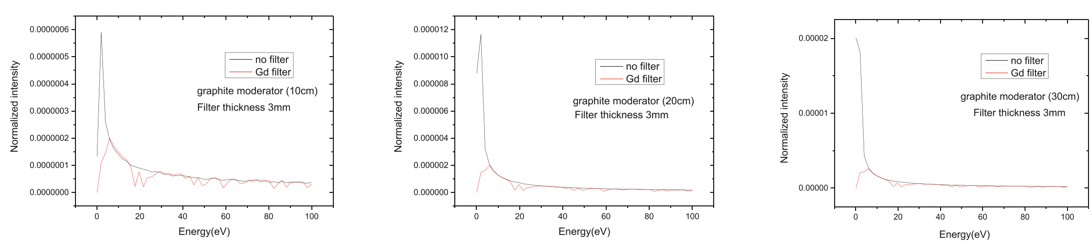


Fig. 11. Neutron energy spectrum by moderation (at graphite, CH2).



a) CH2



b) Graphite

Fig. 12. Neutron cutoff by Gd filter (at CH2, graphite).

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