Development of Gamma Imager and Its Application to Identification of Nuclear Materials

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INTRODUCTION: Identification of a hidden or shielded gamma radioactive sources and nuclear materials is required in anti-terrorism measures. We are developing passive and active gamma spectroscopy combined with gamma-ray imaging for these applications. Compton gamma imager can measure energy of incident gamma-ray and its direction to the detector. We applied a portable and 4π direction sensitive Compton imager [1-2] to identify ¹⁰B sample by prompt gamma-ray with neutron irradiation at the pulsed neutron source at Kyoto University Institute for Integrated Radiation and Nuclear Science Linear Accelerator (KURNS-LINAC).

EXPERIMENTS: A 3D pixel array CdTe detector with 1440 ch. was used as a prototype of the 4π sensitive gamma-ray imager. Three kinds of samples: ¹⁰B, natural uranium (NU) and highly enriched uranium (HEU) were irradiated by pulsed neutrons at a distance of around 12 m from the neutron source. In the case of the ¹⁰B sample, prompt gamma-ray spectrum was measured by the prototype imager with LiF shielding during neutron irradiation. For spectroscopy of delayed fission gamma-rays, gamma-ray spectra from NU and HEU samples were measured from 0 to 6 hours after neutron irradiation.

RESULTS: In reconstructed image of ¹⁰B prompt gamma-rays (478 keV) on a projection sphere around the imager clear peak was found at direction of the sample. LiF shielding had no effect on additional back ground or signal to background ratio. Figure 1 shows a dependence of peak intensity on a distance between the imager and the ¹⁰B sample. Even in rather high event rates caused by gamma flash from the neutron source and neutron induced gamma-rays, peak intensity decreased with the distance as expected.

Figure 2 shows typical background subtracted energy spectra of delayed gamma-ray from the HEU sample after 1-2, 2-3, 5-6 hours of neutron irradiation. According to ref. [3], intensity ratios of delayed gamma-rays from 0.8 to 1.5 MeV can be used to identify the fissile materials: ²³⁵U and ²³⁹Pu. Five peaks of the delayed gam-

ma-rays in the energy region were obviously resolved on the measured spectra as indicated in Fig.2. The ratio of following intensities would be useful for identification:

1031.9 keV (⁸⁹Rb) / 1000.9 keV (¹⁴²Ba)

1383.9 keV (⁹²Sr) / 1532.5 keV (¹⁰¹Mo)

In addition, imaging of delayed fission gamma-rays above 1.5 MeV is also useful to localize HEU material because gamma-rays from NU sample had less contributions on the spectra than those from HEU sample in this energy region.

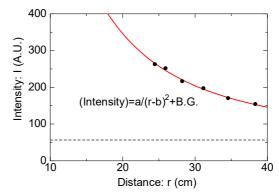


Fig. 1. Dependence of peak intensity of ¹⁰B prompt gamma-rays on a distance between the prototype detector and the ¹⁰B sample.

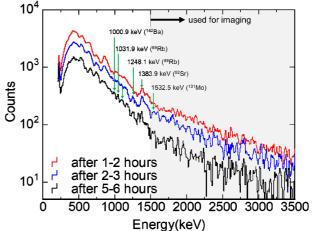


Fig. 2. Typical background subtracted energy spectra of delayed gamma-ray from the HEU sample after 1-2, 2-3, 5-6 hours of neutron irradiation.

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CO2-2 Measurement of Doppler Effect by Small Accelerator Neutron Source (III)

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INTRODUCTION:

We have measured Doppler effect for metal samples which were Nb and Mo by TOF method at KURNS-LINAC since 2016 [1-2]. Doppler effect is depend on the sample and moderator temperature. In numerical calculation of Doppler effect for nuclear systems, the accuracy of neutron spectrum at the moderator is very important. When the moderator in a nuclear reactor is common material such as light-water, researchers and engineers could calculate the accurate neutron spectrum in the moderator by using evaluate nuclear data libraries such as the JENDL-4.0 [3]. On the other hand, if evaluate nuclear data libraries has poor data for moderator materials such as CaH₂, the calculations of accurate neutron spectrum in the moderator will be difficult. CaH2 is compelling solid moderator for new small nuclear reactor [4]. Therefore, in order to perform a nuclear design for the reactor with CaH₂, it is necessary to experimentally investigate the temperature-dependent neutron spectrum in the moderator.

EXPERIMENTS:

To measure the temperature-dependent thermal neutron spectrum in CaH₂ sample, a preliminary experiment was conducted method in KURNS-LINAC. The CaH₂ sample was set in a heater to be increased the sample temperature from about 300K to 600K. Figure 1 shows the heater. The heater was installed at a positon of 12.6 m from the pulsed neutron source at KURNS-LINAC. In this experiment, a GEM type neutron detector has low sensitivity for gamma-ray from the neutron source was employed. Table 1 shows the experimental conditions and Table 2 is the sample information. The transmitted neutron flux from the sample was observed by the TOF method in the present study.

RESULTS:

Table 3 shows the observed and calculated mean energy of Maxwell peak in each temperature. Here, the numerical calculations were performed with MCNP [5] and JEFF-3.2. The change of the peak energies with increasing the sample temperature were observed, respectively. At the low temperature (21 and 119°C), the experimental data was agreed with the numerical calculations. On the other hand, there were about 13 eV difference of the peak energy between the numerical and the experimental result at 292 °C. Figure 1 shows the measured and the calculated neutron spectrums.

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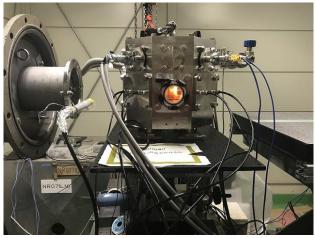


Fig.1 Heater.

Table 1 Experimental condition

Sample	LINAC operational con- dition (Frequency, pulse width, current)	Sample temperature (degree)	Measuring time (h)
CaH ₂	40Hz, 3 µ s, 56 µ A	21	10
CaH ₂	40Hz, 3 μ s, 56 μ A	119	11
CaH ₂	35Hz, 3 μ s, 48 μ A	292	12
Blank	40Hz, 3 μ s, 56 μ A	21	0.5

Table 2 Sample information

Chemical formula	CaH ₂	
Density	1.33 g/cm3	
Geometry	$70 \text{ mm} \times 70 \text{ mm} \times 60 \text{ mm}$	

Table 3 Observed mean energy of Maxwell peak in each temperature

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Temperature	Experiment	MCNP	
(degree)	(eV)	(eV)	
21	0.055 ± 0.005	0.051 ± 0.001	
119	0.068 ± 0.005	0.070 ± 0.001	
292	0.085 ± 0.005	0.098 ± 0.001	

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INTRODUCTION: Evaluation of neutron fluence and neutron dose equivalent for the epi-thermal neutron region is very important in work places with neutron sources or nuclear fuels as well as irradiation fields in a boron neutron capture therapy. In the present study, we have developed a precise epi-thermal measurement method that is not affected by nuclear reaction cross sections such as the ¹⁰B(n, $\alpha\gamma$), ³He(n,p) and ⁶Li(n, α) reactions. The epi-thermal neutron detection system for absolute measurement is composed of a 50 mm diameter and 5 mm thick LGB scintillation detector and two 50.8 mm diameter and 50.8 mm thick BGO scintillation detectors. In the present study, we tried to demonstrate the epi-thermal neutron measurements using a white neutron source including epithermal region. In addition, we have developed a current-mode neutron detector that can be used in intense neutron flux field such as BNCT fields. The neutron detector is composed of a ⁶Li-glass and 7Li-glass scintillators.

EXPERIMENTS: A collimated neutron beam was obtained by the photo neutron reaction using a water-cooled tantalum target at the KURRI Linac [1]. The LGB scintillation detector was set at the center of beam line. The BGO scintillation detectors were placed on both side of the LGB scintillation For low energy neutrons, monoenerdetector. getic gamma rays with energy of 478 keV and alpha particles are produced in the LGB scintillation detector by the ${}^{10}B(n,\alpha\gamma)$ reaction. The alpha particles and a part of gamma rays are observed with the LGB scintillator. The BGO scintillation detectors detects the rest of gamma rays. Characteristics of the neutron detection system were experimentally evaluated by means of the time-offlight (TOF) method.

The current-mode neutron detector was demonstrated at the E-3 beam port of KUR. The linearity of the neutron detector output to neutron flux was measured.

RESULTS: Fig.1 shows the detection efficiency for 478 keV of the BGO scintillation detectors. Constant detection efficiency of BGO detectors for The response for $4 \ge 10^5$ cm⁻²s⁻¹ neutron flux was obtained for the current-mode neutron detector using the E-3 beam port. The spatial distribution of relative neutron flux in the beam area of the beam port was evaluated using a small-sized ⁶Li-glass scintillation detector with a plastic fiber. The linearity for the neutron flux region up to $4 \ge 10^5$ cm⁻²s⁻¹ is confirmed as shown in Fig 2.

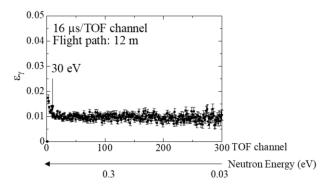


Fig. 1. Detection efficiency for 478 keV of the BGO scintillation detectors in the epi-thermal neutron detector

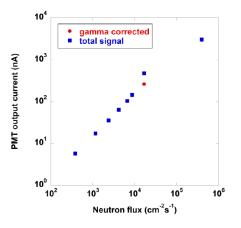


Fig. 2. Relation between the current output of the detector and neutron flux $(cm^{-2}s^{-1})$

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Study on the non-destructive nuclide assay for nuclear materials with a self-indication method

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INTRODUCTION: For decommissioning of Fukushima Daiichi Nuclear Power Plants, the identification and quantification of the nuclear material in fuel debris are important issues for appropriate nuclear material management. However, it is considered that fuel debris contains unknown amounts of minor actinides, fission products and neutron absorbers except for nuclear materials. Therefore, it is difficult to apply conventional non-destructive assay to fuel debris. Neutron Resonance Densitometry (NRD) is one of the candidate techniques of non-destructive nuclide assay applicable to quantity nuclear materials in fuel debris. Especially, a self-indication method [1] is considered as a suitable technique for the identification and quantification of nuclides in fuel debris.

In the self-indication method, an indicator consisting of the target nuclide is placed at the neutron beam downstream from a sample. The transmitted neutron thorough the sample can be measured indirectly by detecting the reaction products from the indicator with the neutron time-of-flight (TOF) method. The self-indicator is a transmission neuron detector that has high efficiency around the objective neutron resonance energies of the target nuclide, enabling us to quantify effectively the amount of resonance absorption of the target nuclide. Moreover, it is not easily affected by the decayed gamma-rays from the fuel. We carried out the examination of self-indication method for a nuclear material with highly enriched uranium-aluminum alloys.

EXPERIMENTS: The experiment was performed at the 46-MeV electron linear accelerator in Institute for Integral Radiation and Nuclear Science, Kyoto University. The linac was operated with a repetition rate of 50 Hz, a pulse width of 4 μ s, a peak current of about 0.5 A, and an electron energy of about 30 MeV. We used a flight path in the direction of 135 degree with respect to the linac electron beam.

An ionization chamber was employed for the measurement of fission fragments from a self-indicator. The chamber is made of aluminum and is 40 mm in diameter and 39 mm in length, and the wall thickness of the chamber is about 2 mm. We used the highly enriched uranium oxide (99.91% of 235 U) which was electrodeposited on a stainless steel disk as a self-indicator. The distance between the electrode and the 235 U deposited layer in the chamber is 8 mm. The chamber is filled with a mixed gas of 97 % Ar and 3 % N₂ at a pressure of 1 atm. The signals from the chamber were stored as a two dimensional data of TOF and Pulse-Height (PH). The fission events were selected by PH discrimination between pulses of α ray and fission fragment. The measurements with and without sheets of highly enriched uranium-aluminum alloys (93% of ²³⁵U, HU filter) were performed, respectively. Each measurement time was about 40 hours.

RESULTS: The TOF spectra with and without HU filter are shown in Fig. 1. The large resonance peaks of ²³⁵U were clearly observed at 1.13, 3.14, 6.99 and 9.70 eV. It can be seen that those resonance peaks were reduced by inserting the HU filter. The counting rates were estimated in thermal energy region from 0.002 to 1 eV and resonance region from 1 to 100 eV. The reduction ratios to the counting rate without the HU filter were obtained as 0.91 ± 0.01 and 0.85 ± 0.05 for thermal and resonance regions, respectively. The reduction in thermal region indicates the neutron absorption by all nuclei in the filter. On the other hands, the reduction in resonance region almost indicates the neutron resonance absorption by ²³⁵U in the sample. In the present study, those ratios were good agreement since we used uranium-aluminum alloy with high purity as a filter. When we used a mixture filter, it is expected that the difference between the reduction ratios for thermal and resonance regions is observed. In next stage, we try to measure with a mixture filter and quantify the areal density of ²³⁵U in the filter.

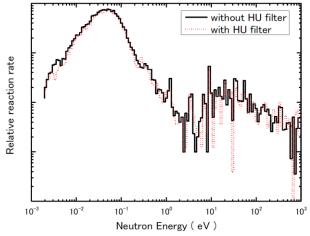


Fig.1 Comparison of TOF spectra for fission events of the ²³⁵U indicator with and without HU filter

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