CO2-1 Development of Measurement Method for Epi-Thermal Neutrons Using the $^{10}B(n,\alpha\gamma)$ Reaction

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INTRODUCTION: Evaluation of 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. A new calibration method for the response of neutron dosimeters has been developed using a pulse white neutron beam[1]. On the other hand, it is not easy to measure precisely the neutron fluence for epi-thermal neutrons in an irradiation field, although a gold activation method is usually used in a thermal neutron region, and an elastic neutron scattering reaction with hydrogen atoms is adopted in a fast neutron region to measure precise neutron fluence. In the present study, we have developed a measurement method for epi-thermal neutrons using a $^{6}Li_{6}{}^{nat}Gd^{10}B_3O_9$:Ce+ (LGB) and an NaI(Tl) scintillators. We also developed an epi-thermal neutron camera consisting of GEMs and resonance filters for neutrons up to 10 keV.

EXPERIMENTS: A collimated neutron beam through collimators was obtained by the photo neutron reaction using a water-cooled tantalum target at the KURRI Linac [2]. The 50 mm-diameter and 5-mm thick LGB scintillator was set at the center of the neutron beam. The 76.2 mm-diameter and 76.2 mm thick NaI(Tl) was located out of neutron beam at an angle of 135 degrees with respect to the neutron beam direction. When the LGB scintillator detects neutrons by the $^{10}B(n,\alpha\gamma)$ reaction, 478 keV monoenergetic gamma rays are produced and subsequently detected with the NaI(Tl) scintillator. In the coincidence measurements, the neutron capture reaction rate in the LGB scintillator is obtained without detection efficiencies of the LGB and the NaI(Tl) scintillators. Moreover, the absolute neutron fluence is determined by measuring gamma rays from the $^{10}B(n,\alpha\gamma)$ reaction with the NaI(Tl) scintillator in setting a 5-cm thick ^{nat}B total absorption sample in front of the LGB scintillator.

The detection system were experimentally tested using the neutron beam from a neutron source with the photo neutron reaction by means of the TOF method. Fig.1 shows the pulse height spectra of the NaI(Tl) scintillator with and without the coincidence measurements. In the NaI(Tl) scintillator, 478 keV gamma rays due to the $^{10}B(n,\alpha\gamma)$ reaction is detected around 600 channel. The counts due to other gamma rays by the coincidence measurements. In the experimental room, many boron is used in shielding materials and collimators. Background measurements with only NaI(Tl) detector were also performed.

Characteristics of a prototype of epithermal neutron imaging camera were also measured using the TOF method. The prototype detector consists of a silver plate as a resonance filter, a B_4C thermal neutron absorber, and a GEM with a neutron converter of ^{10}B.

RESULTS: From the coincidence measurement, count rates for the LGB detector (N_α)) and the NaI(Tl) detector (N_γ) and coincidence count rate ($N_{\alpha\gamma}$) are derived. Neutron capture reaction rate due to the $^{10}B(n,\alpha\gamma)$ reaction is derived from $N_\alpha N_\gamma / N_{\alpha\gamma}$. Neutron fluence was finally determined using ratio of count rates in measurements with and without the thick ^{nat}B total absorption sample. The experimental results are checked by Monte Carlo simulations in future.

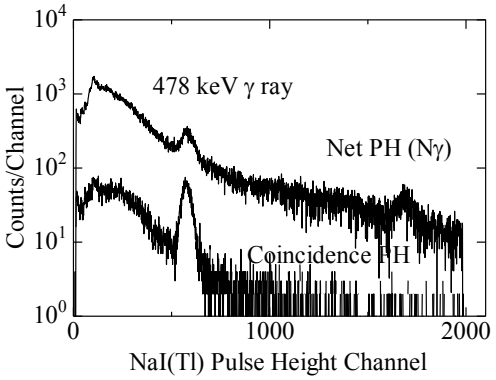

Fig.1. Pulse height spectra measured with the NaI(Tl) detector.

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採択課題番号26055 熱中性子フルエンス率の測定の高度化とその国際標準化に関する研究 通常採択
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CO2-2 Measurement of Gamma Ray and Neutron Spectrum of Curium Isotope (3)

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INTRODUCTION: Yields and spectra data of neutrons and higher energy (>3MeV) γ rays from the spontaneous fission of ^{244}Cm has been focused on since they are major source in spent nuclear fuel. The radiations would be significant indicators of existence of nuclear materials. However those measured data are scarce. KURRI have possessed Cm samples (**Table 1** [1]) in which Cm_2O_3 are diluted in aluminum (Al) powder encapsulated in Al cladding. In this work, preliminary characterizations of CM244 and CM246 are done.

GAMMA RAY MEASUREMENTS: γ rays from samples were measured with a Ge detector of 20% effeiciency. γ rays radiated subsequent to α decays were measured. For CM244 and CM246 samples, multiple γ rays of different energies from $^{243, 244, 245}$Cm were well measured. The activity ratio of CM244/CM246 are 0.935± 0.0191 and 0.714±0.004 for ^{244,245}Cm, respectively. The ratios agree with the data in Table 1.

Then, higher energy γ rays from 2 to 5MeV are focused on to clarify whether noise spectra exist or not for measurements of 2.223MeV and fission γ rays. As shown in **Fig. 1** and broader peaks from 2.5 to 5MeV are found for CM244. The bump spectra exist only slightly for CM246 so it is considered the fission γ ray spectrum dominates the energy region above 2.3MeV in CM246. The resolutions of the peaks are poorer than 2.6145MeV γ ray from ^{208}Tl. That indicates the γ rays are radiated from compound nucleus moving with a certain kinetic energy more than keV. Based on literatures, the 2.235MeV γ rays might be attributed to ^{27}Al(α,γ) reactions [2]. The origin of the other higher energy γ rays are unknown although neutron direct interactions with Ge detertor materials are not the candidates since such bump spectra were not found for a case when ^{252}Cf was put alternatively to CM samples.

The higher energy bumps should be eliminated to quantify fission γ ray spectrum. For the purpose with the samples, coincidence measurements with multiple detectors would be required as well as studies on nuclear physics to explore the reactions radiating such γ rays.

NEUTRON MEASUREMENTS: By putting CM samples and Californium (^{252}Cf) neutron source behind polyethylene block of 10cm thickness, the H(n,γ) 2.223MeV γ rays were measured with the Ge detector. As shown in **Fig. 2**, 2.235MeV γ ray spectrum overlaps the 2.223MeV one for CM samples. The former was subtracted from the latter by normalizing the count rate of the former over a region from 2.235 to 2.250MeV to that of latter. Then the count rate of the 2.223MeV γ rays was estimated. On the other hand, fast neutrons from the CM samples were measured with a NE213 scintillator. The measured count rates are compared in **Table 2**. Considering the difference of neutron spectra shown in **Fig 3**, the count rates in the two detectors agree fairly well (18% difference in CM244). Accordingly, absolute measurement of neutrons from the sample would be capable for CM samples taking the neutron spectrum into account.

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Table 1: Isotopic composition of Cm samples [1]

Sample	^{244}Cm	^{245}Cm	^{246}Cm	^{247}Cm	^{248}Cm
CM244	88.53	3.13	8.34		
CM246	24.18	1.11	62.18	3.00	9.53

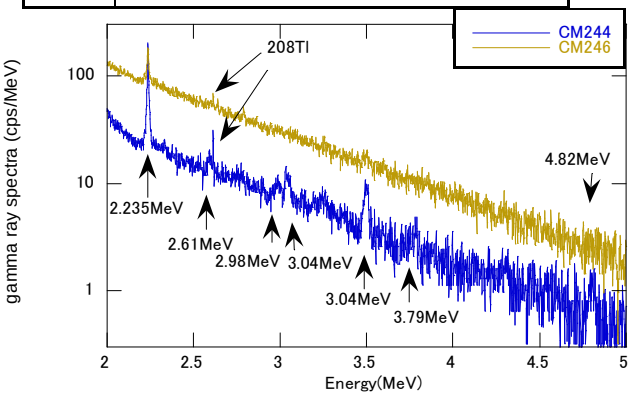

Fig. 1 γ ray spectrum measured for CM244 and CM246 .

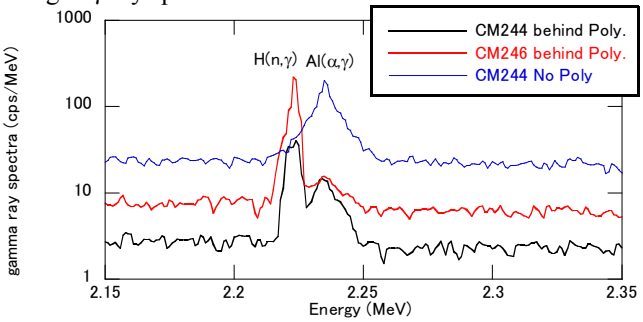

Fig. 2 H(n,γ) spectrum overlapped by Al(α,γ) spectrum

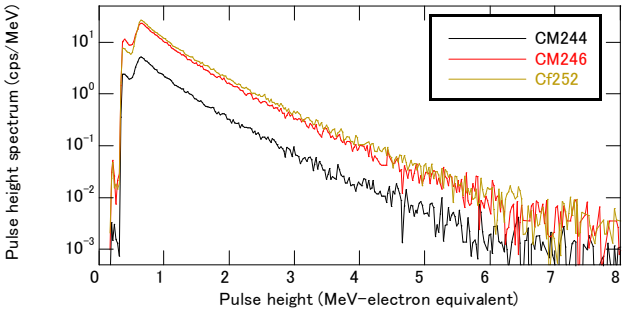

Fig. 3 Neutron spectrum measured with NE213.

Table 2: Count rate ratio of neutrons for CM / ^{252}Cf

Method	CM244/^{252}Cf	rel.err	CM246/^{252}Cf	rel.err
H(n,γ)	0.212	0.030	0.860	0.019
NE213	0.180	0.063	0.843	0.012

採択課題番号 26057 キュリウム核種のガンマ線と中性子発生スペクトルの測定(3) 通常採択
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CO2-3 Development of Hybrid Ce:LiCAF Scintillator for Nuclear Data Measurement

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INTRODUCTION: Precise nuclear data measurement is needed for new generation reactor design and development. In this field, neutron Time-of-Flight (TOF) measurement with pulsed neutron source is playing important role.

In this work, we report on the first nuclear data measurement at KURRI-Linac neutron source with hybrid Ce:LiCAF scintillator which has high neutron detection efficiency and better gamma-ray discrimination[1][2]. Ce:LiCAF covered with plastic scintillator[3] showed excellent characteristics in the detection efficiency and gamma-ray discrimination compared with other conventional scintillators, such as Li-Glass and Li:ZnS scintillators. Especially, excellent pulse height discrimination of neutrons from gamma rays and very short decay time are very promising for a new field of fast neutron detection and great recovery from the gamma-flash.

EXPERIMENTS: Neutron transmission experiments (NRTA: Neutron Resonance Transmission) were carried out at 8m TOF line at KURRI-Linac. ^{237}Np sample was placed at the neutron beam line, and the detector were place in 50 cm behind the sample.

Accelerator was operated in 50Hz and 100ns pulse width. We have fabricated hybrid scintillator based on 10 ×10 × 2mm size Ce:LiCAF scintillator crystal. The crystal were covered with plastic scintillator (PPO and POPOP) and coupled to photo multiplier tube (Hamamtsu R329-05). 5mm thick Pb filter was placed to reduce gamma-rays. *Fast Comtec MPA3* were used for DAQ system.

RESULTS: Fig. 1 shows pulse height spectrum of neutron detector. The line in green shows energy spectrum of neutron and gamma-ray mixed field, and the ling in pink represents a energy spectrum when neutron were shielded with Boron. As shown in Fig. 1, clear neutron peak and very low sensitivity to gamma-ray were observed, which stands for great gamma-ray discrimination. This is due to coupling Ce:LiCAF scintillator with plastic scintillator which enables to improve alpha/beta ratio. With our detector, we successfully measured high energy neutrons up to 2.4 keV. Fig. 2 shows the absorption neutron cross-section of ^{237}Np.

We conclude that TOF spectrum of Ce:LiCAF showed excellent performance for high energy neutron measurement. 8 m TOF line at KURRI.

In addition, low gamma-ray sensitivity great gamma/neutron discrimination with pulse height and showed very quick recovery from gamma-burst which enables high energy neutron measurement.

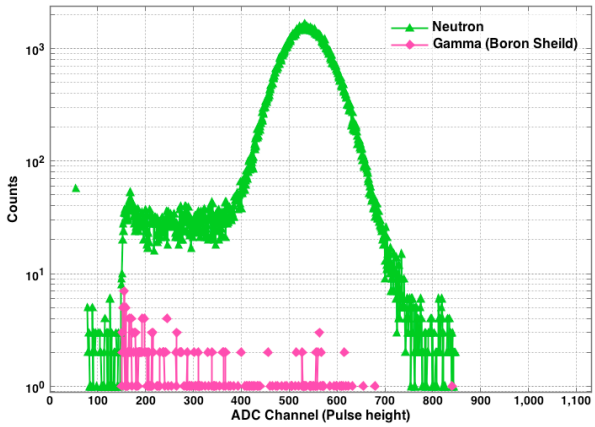

Fig. 1. Pulsed height spectrum of Ce:LiCAF based hybrid scintillation neutron detector. Showed clear neutron peak and low sensitivity to gamma-ray.

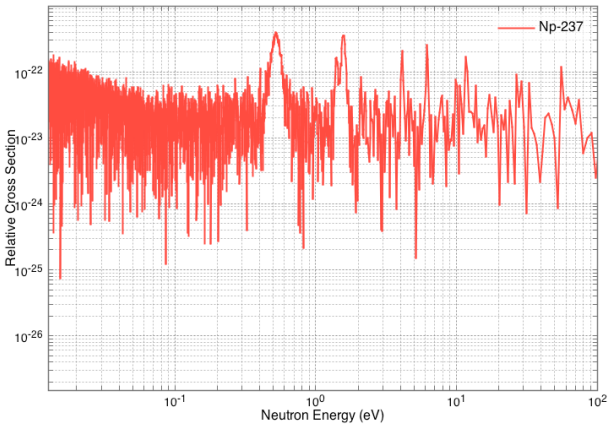

Fig. 2. First result of neutron transmission experiment (NRTA: Neutron Resonance Transmission) of ^{237}Np. The measurement were carried out at 8m TOF line at KURRI-Linac

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採択課題番号 26058　高速シンチレータアレイを用いた二次元核データイメージング装置の開発　通常採択
（東大・工、産総研）藤原　健　（東大・工）松山　大樹、田儀　和浩、Yudhitya Kusumawati、上坂　充　（京大・京大炉）堀　順一、佐野　忠弘

CO2-4 Investigation of Variable Neutron Field for Accurate Determination of Thermal Neutron Capture Cross Section at KURRI-LINAC

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INTRODUCTION: In order to reduce radioactive toxicity, feasibility for nuclear transmutation of minor actinides (MAs) and long-lived fission products (LLFPs) by utilizing innovative nuclear system has been investigated. The report on OECD/NEA No.6410 [1] indicates that there are large gaps on capture cross sections of minor actinides (MAs) between current uncertainty and required accuracy.

Since high-intensity-pulsed neutron sources with spallation reaction such as CERN, LANL and J-PARC become available, statistical error of relative neutron capture cross sections of MAs with time-of-flight (TOF) method was decreasing drastically. However, accurate normalization of TOF data is still an important issue, and unrecognized bias effect needs to be eliminated as much as possible. In this study, we proposed the thermal capture cross section measurement methods using variable neutron fields and developed variable neutron fields in the KURRI-LINAC.

EXPERIMENTS: In order to cross checked the neutron capture cross sections by integral measurements using variable neutron flux field, KURRI-LINAC based neuron source was utilized. Neutrons produced by photonuclear reactions were moderated by light water. Four kinds of variable neutron flux fields were constructed by merging boric-acid solution into the light water moderator, as listed in Table. 1. Neutron spectra of these fields were obtained by TOF measurement of the capture gamma-rays from the $^{10}B(n,\alpha\gamma)$ reaction with twelve $Bi_4Ge_3O_{12}$ (BGO) scintillators, which located at 12.7±0.02 m from the photoneutron source. The linac was operated with a repetition rate of 50 Hz, a pulse width of 100 ns, an averaged current was 18 μA and an electron energy of about 30 MeV. Gold foils with and without Cd cover were also irradiated for each moderator. Reaction rates were obtained with an activation foil method.

RESULTS: Figure 1 shows the neutron spectra for moderators with different boric-acid concentration measured by the TOF method. The thermal bumps were fitted with the Maxwell distribution and the neutron temperatures were determined from 319.5 to 430.8 K. On the other hands, the reaction rates of $^{197}Au(n,\gamma)^{198}Au$ derived from the activation method experimentally were compared with the calculated reaction rates using the measured TOF spectra and the evaluated neutron capture cross sections of JENDL-4.0 [2] as listed in Table 2. The calculated reaction rates were in agreement with the experimental ones within 2 % accuracy. As a result, it was confirmed that the neutron spectrum information obtained by the TOF method can be used for activation analysis.

In nearly future, we will apply the spectrum determination method to the activation experiments for MA.

Table 1 Boric-acid concentration of the light water moderator

Case	Concentration [ppm]
Case 1	0
Case 2	318
Case 3	1404
Case 4	2946

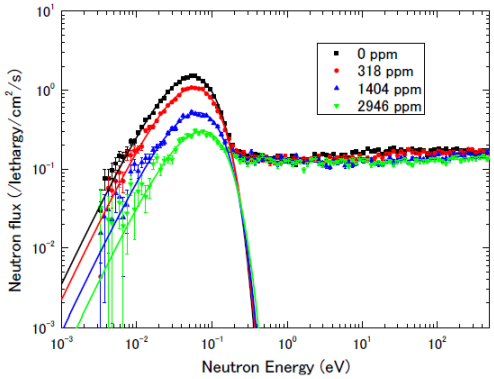

Fig. 1 Neutron spectra for moderators with different boric-acid concentration measured by TOF method

Table 2 Comparison of normalized reaction rates of $^{197}Au(n,\gamma)^{198}Au$

Case	Experiment	Calculation
Case 1	1.858 ± 0.040	1.874
Case 2	1.535 ± 0.032	1.577
Case 3	1.152 ± 0.022	1.141
Case 4(normalized)	1.000	1.000

Present study includes the results of “Research and Development for accuracy improvement of neutron nuclear data on minor actinides” entrusted to the Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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採択課題番号 26069　長寿命放射性核種の中性子核データ精度向上に係る研究開発　通常採択
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CO2-5 Experimental Study on Non-destructive Assay with a Pulsed Neutron Source

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INTRODUCTION: Non-destructive assay of nuclear fuel materials in spent nuclear fuel is a key technology for nuclear material accountancy and critical safety. Neutron Resonance Densitometry (NRD) with a pulsed neutron source has been developed for a non-destructive assay of material [1]. In this method, we can identify and quantify the target nuclide by measuring the neutron resonance absorption with the Time-of-Flight (TOF) technique. There are two ways to measure the neutron resonance absorption. One is Neutron Resonance Transmission Analysis (NRTA) [2] and another is Neutron Resonance Capture Analysis (NRCA) [3, 4].

In this work, we have proposed a new concept of "self-indication method" combined with NRTA and NRCA. In the self-indication method, we set an indicator consisted of target nuclide with a high purity at the beam-downstream from a sample. By detecting the reaction products such as neutron capture γ rays or fission products from the indicator with the TOF method, the transmission neutron can be measured indirectly. The self-indicator is a transmission neutron detector which has a high efficiency around the objective neutron resonance energies of target nuclide, so that it enables us to quantify effectively the amount of resonance absorption of the target nuclide. Moreover, it is hard to be affected by the decayed γ-rays from the fuel.

EXPERIMENTS: We demonstrated a nondestructive assay for nuclear material using a 46-MeV electron linear accelerator at the Kyoto University Research Reactor Institute. A mixture sample of a natural uranium plate and sealed minor actinides such as ^{237}Np and ^{243}Am was used as a sample. The natural uranium plate was 10×20 mm^2 and weighted 5.8 g. The samples of ^{237}Np and ^{243}Am were oxide powder, which was pressed into a pellet 20 mm in diameter and encapsulated in an aluminum disk-shaped container 30 mm in diameter with 0.5-mm-thick walls. The activities of ^{237}Np and ^{243}Am were 26 and 868 MBq, respectively. The ^{10}B plug 8 mm thick or a natural uranium plate of 10×20 mm^2 and weighted 5.8 g was used as an indicator. Prompt-capture γ-rays from the indicator were measured with a pair of $Bi_4Ge_3O_{12}$(BGO) detectors in the TOF experiment. Since the ^{10}B indicator can absorb most neutrons with energies below the epithermal-region, it was equivalent to the conventional NRTA. In the latter case, it was the self-indication measurement.

RESULTS: The TOF spectra for the mixture composed of ^{nat}U, ^{237}Np and ^{243}Am are shown in Figs. 1 and 2. Although many resonance dips from the impurities of ^{237}Np and ^{243}Am were observed (Fig. 1), there are no differences between TOF spectra with only ^{nat}U (blue line) and the mixture (red line) (Fig. 2). This result indicates that the self-indication TOF spectrum was not greatly influenced by nuclide impurity. It was experimentally shown that the contribution from the other nuclide can be remarkably suppressed by applying the self-indication method.

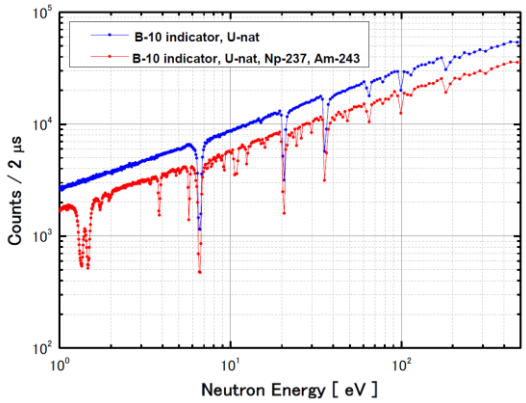

Fig. 1 TOF spectra obtained with NRTA for ^{nat}U and mixture of ^{nat}U, ^{237}Np and ^{243}Am

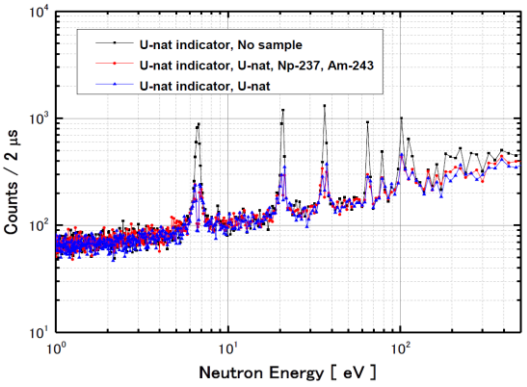

Fig. 2 TOF spectra obtained with the self-indication method for ^{nat}U and mixture of ^{nat}U, ^{237}Np and ^{243}Am

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採択課題番号 26070　　パルス中性子源を用いた核燃料物質の非破壊定量及び可視化技術の開発　　通常採択

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