CO2-1 Research and Development for Accracy Improvement of Neutron Nuclear Data on Long-lived Radioactive Nuclei at KURRI-Linac

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INTRODUCTION: There are strong requests for reducing the uncertainty of neutron capture cross section data of minor actinides (MAs) to estimate the transmutation rate of those long-lived radioactive nuclei in the innovative reactor system. In recent years, intense pulsed spallation neutron sources such as J-PARC[1], n-TOF[2] and LANSCE[3] facilities became available to remarkably improve the precision of neutron TOF data. However, there are discrepancies out of a range of tolerance between current experimental results. It is understood that the unrecognized systematic errors make a difference. In order to recognize and reduce the systematic errors, the project entitled as "Research and development for Accuracy Improvement of neutron nuclear data on Minor ACtinides (AIMAC)" has been started. In this project, we aim at obtaining the resonance parameters precisely of MAs by combining the neutron capture γ-ray measurement to transmission neutron measurement at KURRI-Linac. In this year, new detection system of neutron capture γ rays and transmission neutron was constructed and its performance was confirmed.

EXPERIMENTS: We constructed 4π bismuth germinate (BGO) scintillation detectors composed of 12 BGO cylindrical crystals having 2 inch. in diameter and 2 inch. in length for measurement of neutron capture γ rays. On the other hands, a 6 mm thick GS20 ^{6}Li-glass scintillator was used as a transmission neutron detector.

In the performance test, we used a sealed MA sample of ^{237}Np. Neptunium oxide powder of 1.13 g packed in an aluminum disk container of 30 mm in diameter and 0.4 mm thick wall. The sample was placed in the geometrical center of the BGO detectors using an aluminum sample folder. To decrease the constant background due to decay γ rays from radioactivity (26 MBq) of ^{237}Np, a 3 mm thick lead shield was inserted in the sample folder and BGO detectors. A distance between the sample and the KURRI-LINAC neutron source was 10 m. To obtain the incident neutron spectrum, a ^{10}B sample was also used. Transmission neutron spectra with and without the ^{237}Np sample were measured by the ^{6}Li-glass detector.

RESULTS: Preliminary result of neutron capture cross section of ^{237}Np is shown in Fig. 1. The relative cross sections were normalized to the evaluated value of JENDL-4.0[4] at 0.0253 eV. A correction of neutron multiple-scattering and self-shielding in the sample has not be made yet. Although the amount of measuring time is about one hour, enough net counts were obtained in the resonance region. Comparison of transmission neutron spectra with and without ^{237}Np sample by the ^{6}Li-glass detector is shown in Fig. 2. We could observe the significant differences between those spectra with a high signal to noise ratio..

The results show that the developed capture γ-ray and transmission neutron detection system has a high performance to determine the resonance parameters.

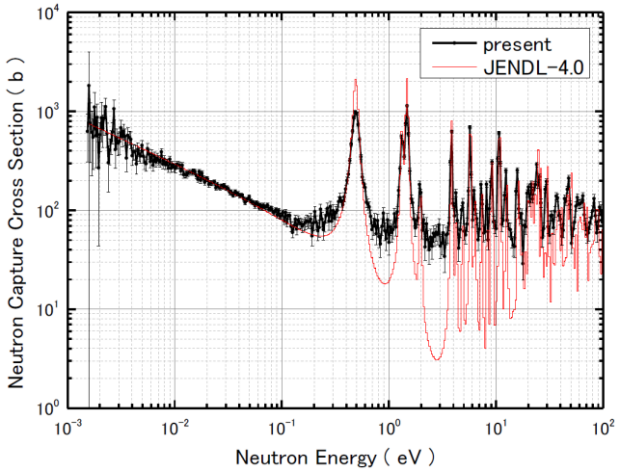

Fig.1 Preliminary results of neutron capture cross section of ^{237}Np

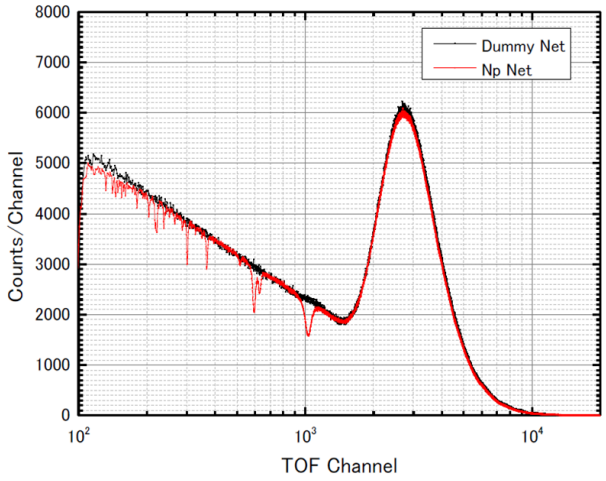

Fig.2 Comparison of transmission neutron spectra with and without ^{237}Np sample by the ^{6}Li-glass detector

Present study includes the result 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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CO2-2 Activation Measurements of Neptunium-237 at KURRI-Linac

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INTRODUCTION: A series of experiments with an activation method has been performed to measure thermal-neutron capture cross-sections of Minor Actinides (MAs) under the project entitled by "Research and development for Accuracy Improvement of neutron nuclear data on Minor ACtinides (AIMAC)". Neptunium-237 is one of important MAs because it contributes to the long-term radiotoxicity of nuclear wastes. Then, the present work performed cross-section measurements of ^{237}Np by an activation method with a neutron source generated by the KURRI-Linac.

EXPERIMENTS: A standardized solution of ^{237}Np (4 kBq) was pipetted on a quartz plate (10mm×15mm×2mm^t), and dried by an infrared lamp. Then, the Np sample was put into an aluminum target holder together with Co and Au foils as neutron monitors. The target holder was set near the moderator tank at the target room of KURRI-Linac. The irradiation was performed for 14 hours. The KURRI-Linac was operated under the condition: repetition rate 50Hz, pulse width 4μs, 108μA beam current and 3-kW power.

After the irradiation, induced activities of the samples were measured with a high purity Ge detector. **Figure 1** shows an example of γ-ray spectrum of the irradiated ^{237}Np sample.

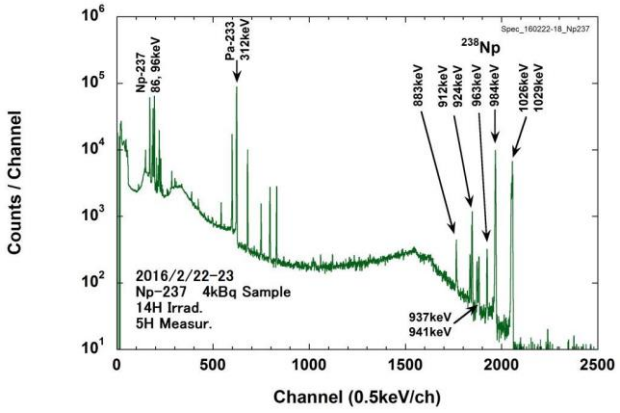

Fig.1 γ-ray spectrum of the irradiated ^{237}Np sample

RESULTS and DISCUSSION: Neutron flux components were derived from the induced activities of the monitors on the basis of the Westcott's convention**[1]**. The thermal-neutron flux was 2.97×10^{8}(n/cm^{2}s) at the irradiation position of 3-kW operation. The epi-thermal index was 0.053. The reaction rate of the ^{237}Np sample was calculated with 312-keV and 984-keV γ-ray yields, detection efficiencies, decay data and γ-ray emission probabilities. The reaction rate was $(7.12\pm0.25)\times10^{-14}$ (1/s). The experimental reaction rate was compared with the calculated one with evaluated nuclear data of ^{237}Np. The neutron flux distribution at the irradiation position was calculated by MVP2.0**[2]** with JENDL-4.0**[3]**, and plotted in **Fig.2**. The calculated flux was normalized with the reaction rate of Au monitor. With the normalized neutron flux and the cross-section data ^{237}Np from JENDL-4.0**[3]**, the calculated reaction rate resulted in $(7.41\pm0.14)\times10^{-14}$ (1/s). The calculated reaction rate was good agreement with the experimental one within the limits of error.

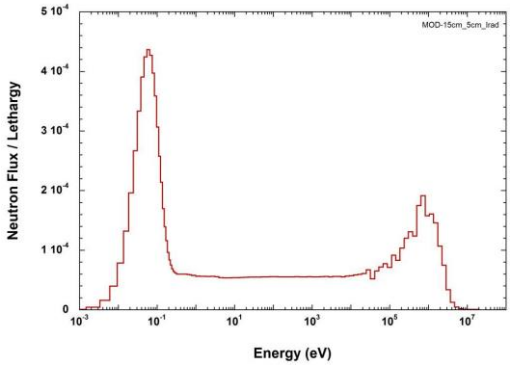

Fig.2 Neutron flux distribution calculated by MVP Codes

CONCONCLUSION: Since the thermal-neutron flux was found to be 10^{8} (n/cm^{2}s) order, activation measurements would be possible under adequate experimental conditions: irradiation time, repetition rate, beam current and so on. The activation measurement of ^{237}Np was performed. It was found that this measurement supported the evaluated data of JENDL-4.0.

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Present study includes the result 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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CO2-3 Development of the Epi-Thermal Neutron Measurement Method Using a Thick Boron Sample

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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. The epi-thermal neutrons are usually measured using a gold activation detector, a gas proportional counter or a scintillator. However, it is not easy to determine precisely the neutron fluence for the epi-thermal neutrons in an irradiation field because of large uncertainty of reaction cross sections in the epi-thermal region. In the present study, we have developed an epi-thermal measurement method that is not affected by nuclear reaction cross sections. 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 was obtained by the photo neutron reaction using a water-cooled tantalum target at the KURRI Linac [1]. Characteristics of a neutron detection system composed of a $^{6}Li_{6}{}^{nat}Gd^{10}B_{3}O_{9}$:Ce+ (LGB) scintillator and an NaI(Tl) scintillator were experimentally evaluated by means of the time-of-flight (TOF) method. The 50 mm-diameter and 5-mm thick LGB scintillator was set at the center of the beam line. The 76.2 mm-diameter and 76.2 mm thick NaI(Tl) was located out of neutron beam at an angle of 135 or 90 degrees with respect to the neutron beam direction. When the LGB scintillator detects neutrons by the $^{10}B(n,\gamma\alpha)$ reaction, 478 keV monoenergetic gamma rays are produced and subsequently detected with the NaI(Tl) scintillator. Moreover, the absolute neutron fluence is determined by measuring gamma rays from the $^{10}B(n,\gamma\alpha)$ reaction with the NaI(Tl) scintillator in setting a 5-cm thick ^{nat}B total absorption sample in front of the LGB scintillator. Number of alpha particles and gamma rays produced by the $^{10}B(n,\gamma\alpha)$ reaction in the LGB scintillator are absolutely determined by the coincidence measurements. Uncertainty caused by the cross sections of the $^{10}B(n,\alpha\gamma)$ reaction is reduced by using the boron total absorption sample.

RESULTS: Fig.1(a) and (b) show ratio of the pulse height spectra and the TOF spectra of the NaI(Tl) detector with and without the thick ^{nat}B total absorption sample. In Fig.1(a), 478 keV gamma rays due to the $^{10}B(n,\alpha\gamma)$ reaction is detected around 600 channel. These figures indicate the relative ratio of count rate of the NaI(Tl) scintillator with and without the boron total absorption sample. In the measurements, it was confirmed that the present detection system derived the neutron fluence in the thermal and epi-thermal region.The experimental results will be basic data in order to develop a novel portable epi-thermal neutron detector.

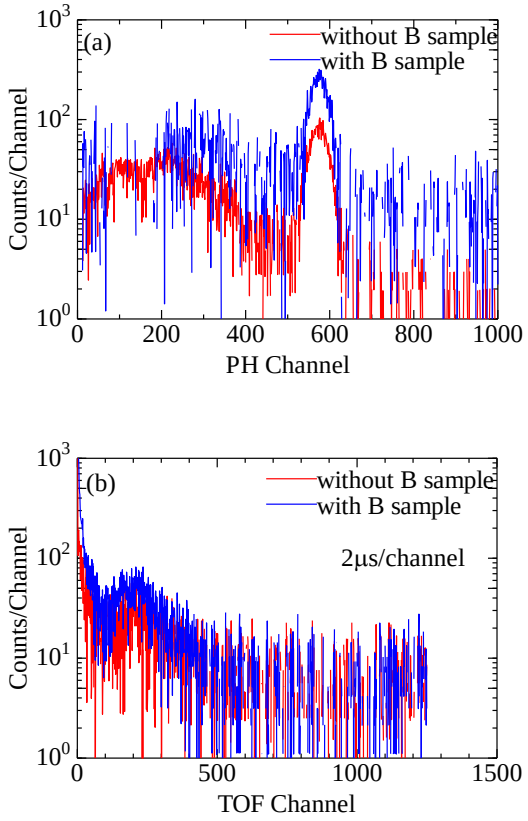

Fig.1. Ratio of (a) the pulse height spectra and (b) the TOF spectra of the NaI(Tl) detector with and without the thick ^{nat}B total absorption sample.

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CO2-4 Measurements of Americium Isotopes by Activation Method at KURRI-Linac

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INTRODUCTION: Neutron capture cross section measurements have been conducted for Minor Actinides (MAs) under the research project entitled by "Research and development for Accuracy Improvement of neutron nuclear data on Minor ACtinides (AIMAC)".

Since americium-241 and -243 have relatively long half-life as 432yr and 7370yr respectively, these isotopes are important nuclides among MAs in terms of environment load reduction. This is why these two americium isotopes were selected in the present measurements.

EXPERIMENTS: Americium-241, 243 samples were sealed on Al plates with 22mm in diameter and 0.5mm in thickness, respectively. Their active areas were 20mm in diameter. The amounts of the samples were 583kBq for ^{241}Am, and 2MBq for ^{243}Am, respectively. Each Am sample was packed into an Al target holder. Then, sets of monitors of Au and Co foils were sealed at adequate positions on the Al holders. Then, the target holders were set near the moderator tank at the target room of KURRI-Linac.

Figure 1 shows the irradiation set-up of the ^{243}Am target near the moderator tank at the target room of KURRI-Linac. The ^{241}Am target was irradiated for 35 hours under beam conditions: repetition rate 100pps, beam width 4μs, beam current 220μA, and 6-kW power operation, while the ^{243}Am target was irradiated for 40 hours under another beam conditions: repetition rate 50pps, beam width 2.5μs, current 67μA, and 2-kW power operation.

Fig. 1 Set-up of the ^{243}Am target at the target room of KURRI-Linac

After the irradiations, induced activities of the samples were measured with an α-ray spectrometer, and a high purity Ge detector. **Figure 2** shows an example of α-ray spectrum obtained by measurements of the irradiated ^{241}Am sample for 11 days. On the other hand, the ^{243}Am sample was measured by the Ge detector. The γ-ray spectrum of the ^{243}Am sample was measured for 20 hours, and is shown in **Fig.3**.

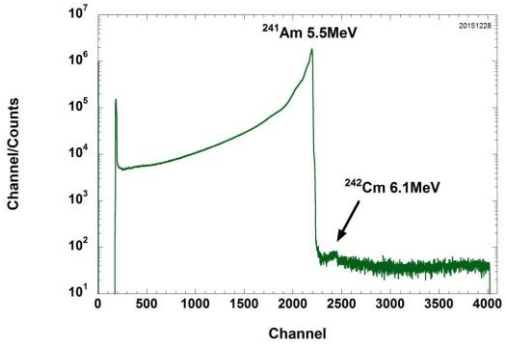

Fig.2 An example of α-ray spectrum of the irradiated ^{241}Am sample

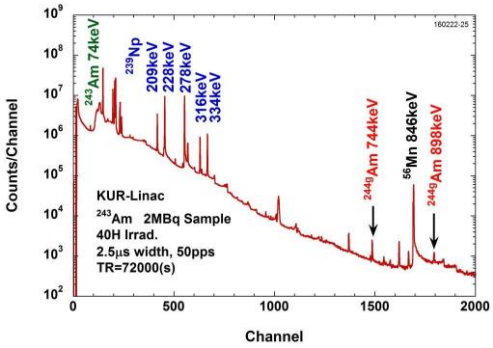

Fig.3 An example of γ-ray spectrum of the irradiated ^{243}Am sample

ANALYSIS and RESULTS: As for the ^{241}Am experiment, the neutron flux was approximately 1.4×10^{8} (n/cm^{2}s) on the basis of the Westcott's convention. From the peak counts of ^{241}Am and ^{242}Cm in Fig.2, the rate of ^{241}Am(n,γ)^{242g}Am reaction was obtained as $(8.63\pm0.80)\times10^{-14}$(/s). As for the ^{243}Am experiment, the neutron flux was approximately 9.0×10^{7}(n/cm^{2}s) at the target position. The decay γ-rays of 744 and 898 keV of ^{244g}Am (10.1h) were observed as shown in Fig.3. The target amount of the ^{243}Am sample was obtained with the yields of 228 keV γ-ray emitted from ^{239}Np in equilibrium with ^{243}Am. From the ratio of γ-ray yields of 744 keV of ^{244g}Am and 228 keV of ^{239}Np, the ^{243}Am(n,γ)^{244g}Am reaction rate was obtained as $(1.14\pm0.03)\times10^{-15}$(/s), which only included statistical errors.

Furthermore, detailed analyses are underway for the ^{241}Am and ^{243}Am experiments carried out at KURRI-Linac.

The authors would like to thank staffs of Kyoto University Research Reactor Institute for their support.

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