CO2-1

Neutron resonance analysis technique in neutron time-of-flight for shielded nuclear materials

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INTRODUCTION: The Japan Atomic Energy Agency is developing various active neutron interrogation techniques [1] to supplement safeguard verifications. Neutron resonance analysis aims to combine the neutron resonance transmission analysis [2,3], together with the neutron resonance capture analysis [3,4] and the newly proposed neutron resonance fission neutron analysis (NRFNA) [5]. In a NRFNA method, using a pulsed neutron beam with the neutron time-of-flight (TOF) technique, prompt capture gamma rays and fast fission neutrons from fissile nuclides are measured at the sample location. The use of plastic scintillators with good n/γ pulse shape discrimination (PSD) is necessary to distinguish fission neutron events from gamma ray events. In this paper, we report a demonstration experiment for shielded nuclear materials performed at KURNS-LINAC.

EXPERIMENTS: The KURNS-LINAC was operated with an acceleration energy of ~30 MeV, and an average current of ~42 μA. Pulsed neutrons were produced with a repetition rate of 50 Hz and a pulse width of 2 μs. Natural uranium samples of 1.5, 3.0, and 6.0 mm thickness were placed in lead (Pb), polyethylene (PE), and borated polyethylene (B-PE) boxes. Each box has external dimensions of 7x7x7 cm^3 with a wall thickness of 2 cm. Six hexagonal PSD plastic scintillation detectors (Eljen, EJ-276) surrounded by lead shielding were used for NRFNA. The setup used is shown in Fig. 1, for the case of the Pb shielding box. The output signals of the detectors were sent to the CAEN digitizer V1730D (14bit, 500MSample/s); the data were recorded in list mode with 1h autosave.

RESULTS: Obtained data are being analyzed by the PSD technique [5]. Fig. 2 shows the TOF spectra of a 3.0 mm natural uranium sample inside the Pb box. In the gamma-ray spectrum, resonance peaks from ^{238}U(n,γ) reaction are clearly visible at 6.67, 20.87 and 30.68 eV; in the neutron spectrum, the characteristics peaks from ^{235}U(n,f) reaction are observed at all energies from 1 to 35 eV. Similar results can also be achieved with smaller sample. This shows a possibility to identify fissile nuclides in heavy material shielding container. Further analyses are ongoing to quantify the fissile content.

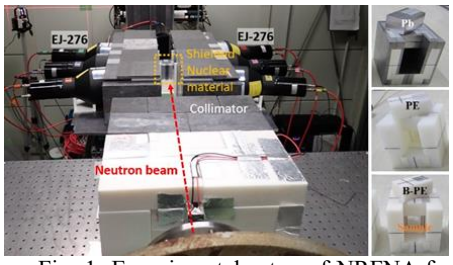

Fig. 1. Experimental setup of NRFNA for shielded materials (Pb, PE, and B-PE).

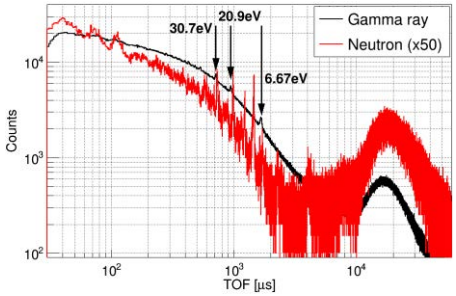

Fig. 2. TOF spectra of 3 mm natural uranium in Pb shielding.

ACKNOWLEDGEMENTS: This research was implemented under the subsidy for "promotion of strengthening nuclear security and the like" of the MEXT (the Ministry of Education, Culture, Sports, Science, and Technology of the Japanese government).

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[2] P. Schillebeeckx *et al.*, Nuclear Data Sheets (2012) 113.
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CO2-2

Neutron Activation Analysis for High-Purity Niobium Compound and Metal

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INTRODUCTION: Unlike typical atomic nuclei composed of both protons and neutrons, multi-neutron systems, especially tetraneutron, have attracted much attention [1]. We have been searching for tetraneutrons (^{4}n), which might be emitted in thermal fission of uranium, by means of the activation method. When we insert a sample containing a nuclide, ^{A}Z, into a reactor, tetraneutrons may interact with a nucleus and produce an isotope, $^{A+3}$Z, by the ^{A}Z(^{4}n,n) reaction. If the isotope $^{A+3}$Z is unstable and emits γ-rays after β decay, the amount of $^{A+3}$Z after the irradiation can be evaluated by γ-ray spectroscopy. This method resembles the neutron activation analysis, except for the reaction.

In the first irradiation experiment, we used ^{88}Sr-enriched strontium carbonate, and determined an upper limit of the emission rate of tetraneutrons in uranium fission [2]. We learned that a silica tube which was used to confine powder of strontium carbonate was a large source of background in the obtained γ-ray spectrum. To avoid using a sample in powder form, we have decided to focus on a different element. The target nucleus ^{A}Z should be chosen carefully, based on the neutron capture cross section and the half-life of $^{A+1}$Z, both of which directly affect the radioactivity of the sample after irradiation, and the half live of $^{A+3}$Z, as well as the chemical properties and the purity of the sample. A mononuclidic element is suited in view of the availability of a high-purity sample, and niobium will be one of the promising elements meeting the aforementioned criteria.

EXPERIMENTS: We prepared three kinds of niobium samples: (1) niobium pentoxide (Nb_2O_5), (2) and (3) niobium metals from different companies. In general, niobium metal contains tantalum, whose chemical properties are similar to those of niobium. On the other hand, niobium pentoxide does not contain tantalum. However, As niobium pentoxide was in powder form, we made a pellet by using a press machine so as to avoid using a silica tube. We irradiated samples at the pneumatic tube Pn-2 and the hydraulic conveyer Hyd in order to investigate the γ-rays emitted from the activated sample, which are mainly due to impurities in the samples. This measurement is important in determining a detailed plan of the next search experiment of tetraneutrons with a longer irradiation time.

PRELIMINARY RESULTS: Figure 1 shows the γ-ray spectra for the three samples. As expected, photopeaks from tantalum are seen for samples (2) and (3). Moreover, the background level is comparable between them, while that of sample (1) is sizably larger. A detailed analysis is in progress.

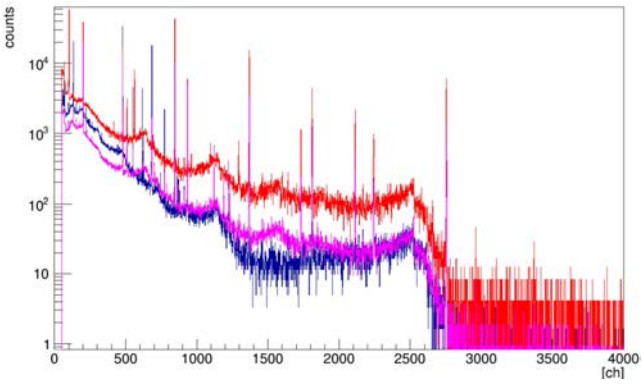

Fig. 1 γ-ray spectra for the three samples [(1): red, (2): blue (3): magenta]. The spectra are normalized for 1 gram of niobium.

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CO2-3

Development of thimble-type ionization chamber for an intense epi-thermal neutron beam

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INTRODUCTION: The need to ensure traceability to neutron standards at AIST for hospitals performing boron neutron capture therapy (BNCT) has been discussed. For this purpose, we are developing a real-time detector that can measure high-intensity neutrons used in BNCT. It is required that the detector can measure neutrons in appropriate measurement time at standard neutron fields whose fluxes are 4 to 5 orders of magnitude lower than those at BNCT as well.

EXPERIMENTS: A thimble type ionization chamber was developed as a neutron detector for high intensity neutrons. The ionization chamber is made of aluminum and has a dome shape with a diameter of 13 mm and a length of 15 mm. An aluminum needle-shaped electrode with a diameter of 1 mm is set in the center. The chamber was filled with a mixture of ^{3}He gas (0.5 atm) and Ar gas (1.4 atm). Characterization of the ionization chamber was performed using a pulsed white neutron source produced using an electron linac at the Institute for Integrated Radiation and Nuclear Science of the Kyoto University. The ionization chamber measured neutrons by two-dimensional measurements of neutron time-of-flight (TOF) and pulse height. In addition, in the current output mode, the measurement was performed by varying the applied voltage for the neutrons.

RESULTS: Figure 1 shows a TOF spectrum obtained from the ionization chamber in the pulse mode. Neutron components were successfully observed using the ionization chamber. Figure 2 shows the relation between the applied voltage and output current from the center electrode in the current mode. This figure indicate that the ion saturation region and proportional region are successfully obtained. However, signal to noise ratio was insufficient. In the future, we will evaluate the detection efficiency and stability. In addition, it is necessary to accumulate data for various gas mixing patterns to improve S/N.

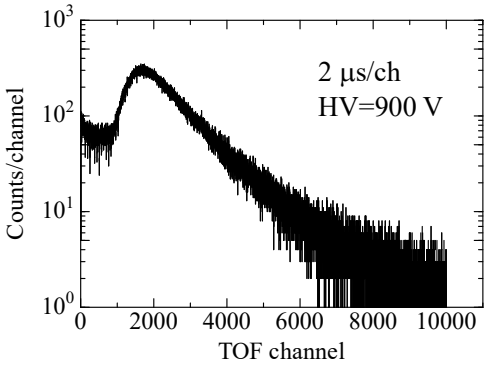

Fig. 1. TOF spectrum for the pulsed neutron source obtained from the electron linac. The thermal bump was clearly observed.

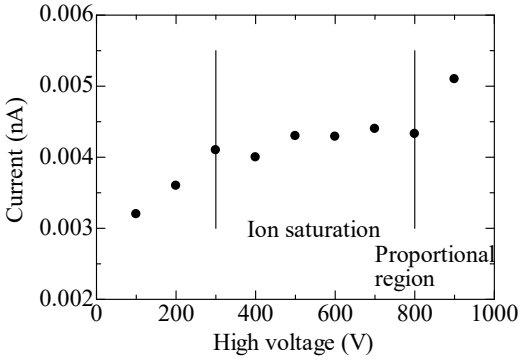

Fig. 2. Relation between applied voltage and output current.

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CO2-4

Identification of β-delayed γ and x-rays of ^{157}Nd using an Isotope Separator On-Line KUR-ISOL

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INTRODUCTION: The decay data of the fission products are important for nuclear engineering and nuclear physics. We have focused on the decay studies of A~150 region because of few decay information due to their short half-lives and low fission yields. Continuing from last year, we conducted study on the decay of ^{157}Nd to identify β-delayed γ-rays and KX-rays. The half-life of ^{157}Nd with β-counting and the level scheme of ^{157}Pm had been proposed in ref.[1,2], respectively. In August last year, the β-delayed γ-rays and a decay scheme using an on-line mass separator for ^{252}Cf spontaneous fission [3] have been proposed for the first time ahead of us. In this experiment, the β-delayed KX-ray was identified although there was less information on γ-rays than ref.[3].

EXPERIMENTS The amount of 72mg of 93% enriched $^{235}UF_4$ target was installed in the through-hole facility in Kyoto University Reactor. The nuclei of interest were produced by thermal neutron-induced fission and mass-separated with KUR-ISOL. They were collected on a computer-controlled thin Mylar tape and transported to the measurement position periodically. The clover detector, which has four large Ge crystals (80 mmø×90 mmL) and a through hole 15 mmø, and two semi-cylindrical β-particle detectors made of plastic scintillator set in the through-hole were used for β-γ coincidence measurements. The detectors were shielded with 10 cm thick lead and boron-doped polyethylene bricks to reduce the background radiation. Data were recorded on APV data acquisition systems (Techno AP Co.) with list mode including time information. The time interval of collection-measurement was set at 3 s-3 s, respectively.

ANALYSIS and RESULTS: To determine the half-lives, the measured data were reconstructed to each 0.5 s spectra to analyze with spectrum multi-scaling (SMS). Fig.1 shows the typical decay curves of the 66 keV γ-rays. The half-life was determined 1.2(5) s, approximately. The half-lives of the 255 and 1703 keV γ-rays also agree the value of ref.[3] with in the uncertainty. In KX-ray region, Sm KX-ray of the daughter nuclide were intensively observed than Pm KX-ray. Then the decay curves were analyzed with the time differential spectra of each 1 s. As shown in Fig.2, the Pm KX-ray was certainly observed in the first differential spectrum and almost disappeared in the next differential spectrum. The Pm KX-ray is considered to decay with about 1 s associated with the β-decay of ^{157}Nd. In these analyses, the three γ-rays and Pm KX-ray were identified as the β-delayed γ-rays and KX-ray with the half-life of about 1 s.

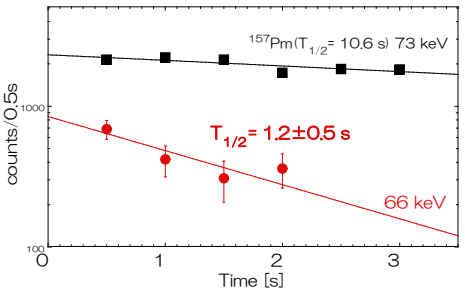

Fig. 1. Decay curve of the 66 keV γ-ray.

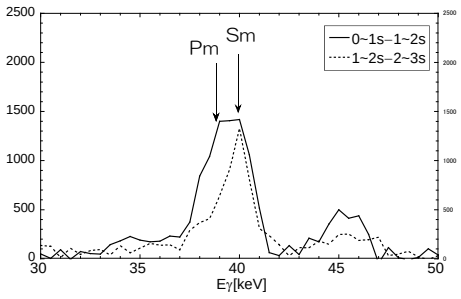

Fig. 2. The X-ray region of the time differential spectra.

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CO2-5

Experimental study on photo-neutron reaction of uranium by bremsstrahlung photons

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INTRODUCTION: Due to the social demands to enhance the nuclear security, non-destructive assay technology has been developed for detect the unauthorized transfer of nuclear material especially highly enriched uranium(HEU) by utilizing the photo-fission signals induced by bremsstrahlung photons [1]. Based on the numerical analysis, it was suggested the feasibility of HEU detection by different energy of bremsstrahlung photons with 7 MeV and 13.5 MeV injected electrons to the Ta target[1]. For the validation of the proposed principle, the experimental study has been started to detect the photo-neutrons signals from uranium target by using the bremsstrahlung photons produced by LINAC accelerator. In the fiscal year 2023, the preliminary experiments were performed for detector characterization and background noise reductions to clarify the experimental set-up requirement for uranium photo-neutrons signals.
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EXPERIMENTS: The LINAC electron beam energy was set as 13, 18, and 30MeV, and multiple target materials were examined such as natural uranium, lead, Iron and void. To detect the photo-neutron signals, liquid scintillator for gamma-neutron co-counting, He-3 tube detectors with poly-carbonate neutron moderator and Cd filter were installed. For the background noise reduction by the photo-neutron reaction by structure materials or others, the Cd and Poly-Block with B were introduced in the beam duct between the LINAC electron target room to the measurement room. As for the reference data, Cf-252 standard source was measured by the set-up detectors. The measurements were performed
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RESULTS: As preliminary results, the detector response of liquid scintillator and He-3 tube were well examined and found their detectability under the bremsstrahlung photon beam irradiation to the target materials(*Fig*. 1). The comparison between the target materials of uranium, lead and irons suggested the different the neutron detection responses. More data analysis and parametric studies will be needed to examine the photo-neutron reactions especially for uranium targets. As for the background noise reduction, the installation of neutron absorbers of Cd filter or Poly-Block with B in the beam duct have not shown the significant background neutron reduction, more analysis is needed including the effect of other structure materials. As a summary, we could achieve the fundamental data for planning the next experiments.
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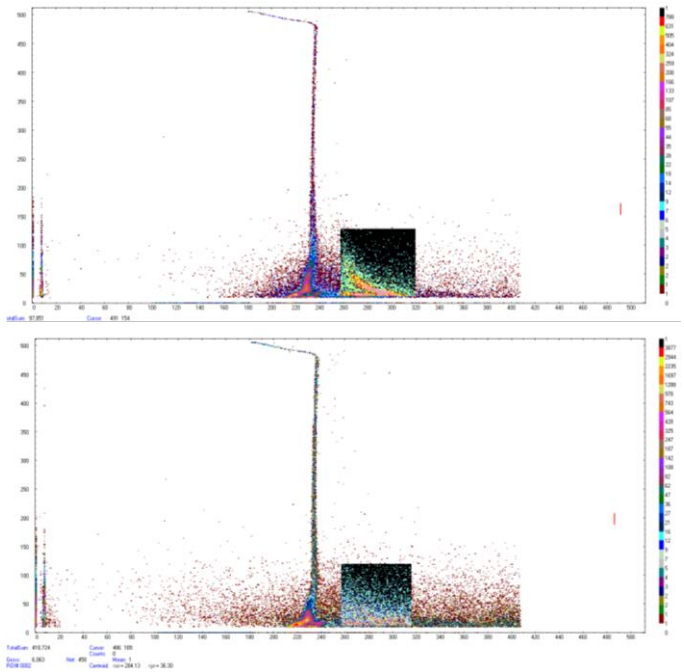

Fig. 1 Liquid Scintillator Response (Up: Cf-252 Neutron Source, Down: Bremsstrahlung photon irradiation to NU, Black area: ROI for neutrons)

REFERENCES:

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CO2-6

Validation Experiment of Thermal Neutron Scattering Law Data for Innovative Reactor

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INTRODUCTION: Small modular reactors (SMRs) with a solid hydride moderator have been studied [1]. Improvement of accuracy of thermal neutron scattering law (TSL) data of candidate solid moderators such as Zirconium-hydride (ZrH_2) and Calcium-hydride (CaH_2) is required for developing innovative reactor system. Therefore, the project entitled as "Development of Nuclear Data Evaluation Framework for Innovative Reactor" has been carried out. In the project, we preformed the integral experiments to validate TSL data of JENDL-5 using a pulsed neutron source.

EXPERIMENTS: The experiment was performed at the Institute for Integrated Radiation and Nuclear Science, Kyoto University (KURNS-LINAC). Pulsed fast neutrons from the photo neutron source were produced without neutron moderator. A beam width and a frequency were 5 μs and 50 Hz, respectively. The flight path used in the experiment is in the direction of 135 degrees to the electron beam. The pulsed neutron beam was collimated to 30 mm in diameter.

Each sample assembly was placed at about 10 m from the neutron source. The cylindrical containers made of aluminum 60 mm in diameter and 55 mm in height and were filled with powders of ZrH_2 or CaH_2. The weights of ZrH_2 and CaH_2 were 476 g and 150 g, respectively. The neutron spectrum by slowing-down in the sample was measured by a ^{6}Li-glass detector with a TOF method. The distance between the center of the container and the surface of the detector was 30 cm. The net thermal neutron spectrum was obtained from the difference between TOF spectra with and without Cd sheet of 0.5 mm thickness around the cylindrical container.

RESULTS: The comparison of the net experimental thermal neutron spectrum with the calculated ones using JENDL-5 [2] for ZrH_2 sample is shown in Fig. 1. The calculations were performed by the Monte Carlo simulation code MCNP-6.3 considered with and without the S(α,β) effect. The thermal bump was observed in about 65 μs of the calculation results with the S(α,β). The experimental values almost supported the calculation with the S(α,β) of JENDL-5.

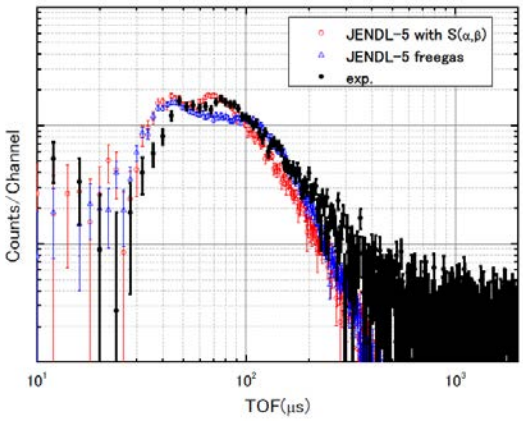

Fig. 1 Comparison of the experimental thermal neutron TOF spectrum with the calculated ones for ZrH_2 sample.

The present study includes the result of "Development of Nuclear Data Evaluation Framework for Innovative Reactor" founded by the Ministry of Education, Culture, Sports, Science and Technology of Japan.

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[2] O. Iwamoto *et al.,* J. Nucl. Sci. Technol., **60(1)** (2023) 1-60 .

CO2-7

Neutron cross-section measurement of molybdenum

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INTRODUCTION: Kyoto University Critical Assembly (KUCA) is currently preparing to use a new reactor fuel using uranium-molybdenum (UMo). This is the first reactor in the world to use this fuel, and it is expected to be used as a research reactor fuel worldwide in the future. In the core analysis of a reactor using UMo fuel, the reliability of nuclear data has been sufficiently confirmed for conventional uranium-aluminum alloy fuel, but core analysis using UMo fuel, especially molybdenum, is not sufficient, and it is necessary to improve the nuclear data of molybdenum.

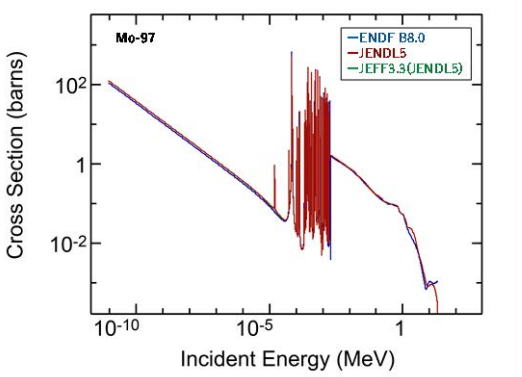

Fig. 1. Cross-section of Mo-97

In fact, when Mo-97 in JENDL5 and ENDF/BVIII are compared shown in Fig.1, there are some different points are observed. Therefore, this study aims to contribute to the improvement of nuclear data of molybdenum by measuring cross sections using a KURNS-Linac for some molybdenum nuclides with a view to upgrading the core analysis of KUCA.

EXPERIMENTS: In this experiment, natural Mo plate of different thickness were used as samples for the preliminary test. The thickness of Mo plate were 0.5 mm, 1.0 mm and 3.0 mm, and size were 18 x 18 mm. The samples for TOF method were set at the 90 degree and 10 m distance from the target. The target was surrounded by pack-man type water moderator. A BGO scintillator (2 inch diam. and 2 inch long) was used for measuring total energy absorption gamma-rays and set at 122 mm from the sample. Only LiF plate was set in front of the detector for neutron shield and no other shield was set. The conditions of the accelerator were as follows: short mode, average beam current was 16.7 μA, frequency was 50 Hz and pulse width was 100 ns. The size of beam is 50 mm diameter.

RESULTS: Fig. 2 shows the TOF measurement results for three different sample thicknesses. Each measurement result is normalized by the measurement time. One channel in this figure corresponds to 100 ns. As an example of the experimental results, there was no large difference in the resonance peak value of 44 eV with increasing thickness. This indicates that $n\sigma_{tot}$ is large enough to saturate the resonance capture due to the neutron self-shielding effect in the sample.

In the future, we plan to conduct cross-section measurement experiments using thinner samples by selecting appropriate sample thicknesses by preliminary calculations.

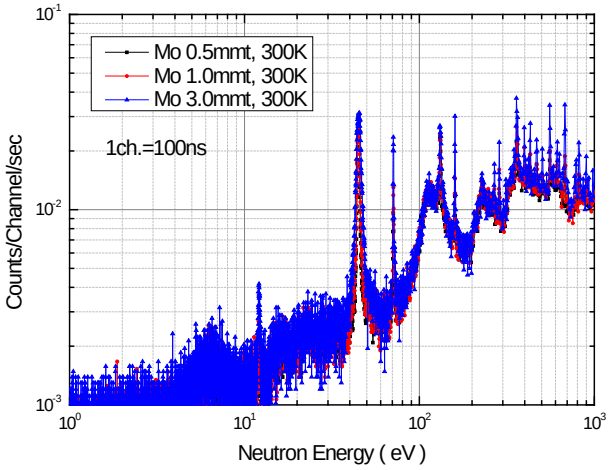

Fig. 2 Result of TOF

CO2-8

Measurement Neutron Reaction Rate Ratio using the TOF method in the KURNS-LINAC

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INTRODUCTION: In thorium fuel loaded nuclear reactor systems, it is important to have reliable evaluated nuclear data on the neutron capture reaction of the parent material Th-232 and the fission reaction of U-233. The authors have previously carried out differential experiments using the electron linear accelerator at the Institute for Integrated Radiation and Nuclear Science, Kyoto University (KURNS-LINAC) to measure the fission cross section of U-233 and the neutron capture cross section of Th-232. As a result, the followings have been observed from these measurements: 1) the U-233 fission cross section measurement, the experimental values are generally in good agreement with the JENDL-4.0 [1]. 2) the Th-232 neutron capture cross section measurements showed differences in the neutron widths of the 69 eV, 121 eV and 170 eV resonances compared to JENDL-4.0 [2]. On the other hand, these resonance parameters have not been revised in the latest JENDL-5 [3], released in December 2021. Therefore, in the present study, the Th-232 neutron capture cross section was remeasured.

EXPERIMENTS: The experiment was performed on the 12 m neutron flight path shown in Figure 1. The samples were Th-232 and B-10 for neutron flux measurement. Th-232 and its daughter nuclides emitted γ rays as background. To reduce the background γ rays, a 10-mm-thick piece of lead was placed between the Th-232 sample and the BGO detector. In the measurement of B-10 for neutron measurement, a single γ ray of 478 keV emitted by the (n,α) reaction between the B-10 sample and neutrons was measured, and the TOF information was observed with the Ta target in-cident of accelerated electrons as the start signal and the prompt gamma ray emitted by the reaction be-tween the sample and neutrons as the stop signal.

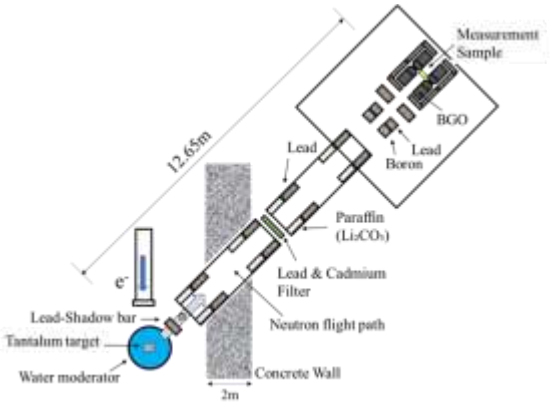

Fig.1 Experimental geometry

RESULTS: Figure 2 shows the observed TOF spectra (Th-232 sample and blank run). The TOF spectra in this figure have been processed for background and so on. The TOF spectra in this figure show that resonance is observed in the region above about 500ch (below 1 keV). In the next step, neutron capture cross section and resonance parameters will be to determine from the measured data.

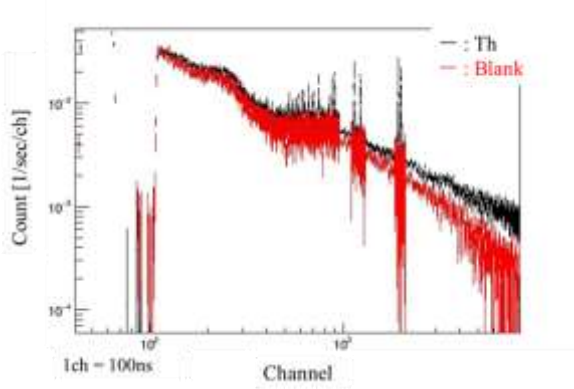

Fig.2 Obtained TOF spectrum

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CO2-9

Measurement of Neutrino Helicity using ^{152m1}Eu

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INTRODUCTION: Parity violation in the weak interaction was discovered in the beta decay process. Left-handed property (i.e., negative helicity) of neutrino is the origin of the parity violation. Another unique property of the neutrino is very small mass. We can interpret it as mass-less fermion in the most of decay processes, and we expect the helicity of -1 for the neutrino. Using a radio isotope irradiated in KUR, we measured the neutrino helicity to confirm this expectation.

EXPERIMENTS: This study was performed to measure the neutrino helicity by using neutrino emission process, $^{152m1}\mathrm{Eu} + e^- \rightarrow {}^{152}\mathrm{Sm}^* + \nu_e$. $^{152}\mathrm{Sm}^*$ promptly goes to the ground state with gamma-ray emission. Because $^{152}\mathrm{Sm}^*$ has non-zero momentum, the gamma ray energy to "forward" (or "backward") direction has approximately 1% higher (or lower) energy than the rest frame as illustrated in Fig. 1. We selected "forward" process using the similar setup as previous study [1]. The gamma ray loses its energy via the Compton scattering. In Sm_2O_3 scatterer, resonant interaction occurs when the gamma energy is equal to the excitation energy of ^{152}Sm, i.e., 963 keV. We measured the resonant gamma rays (841 keV or 963 keV) by using a scintillation counter. By changing the direction of magnetic field, we modulate the cross section of the Compton scattering with respect to the helicity.

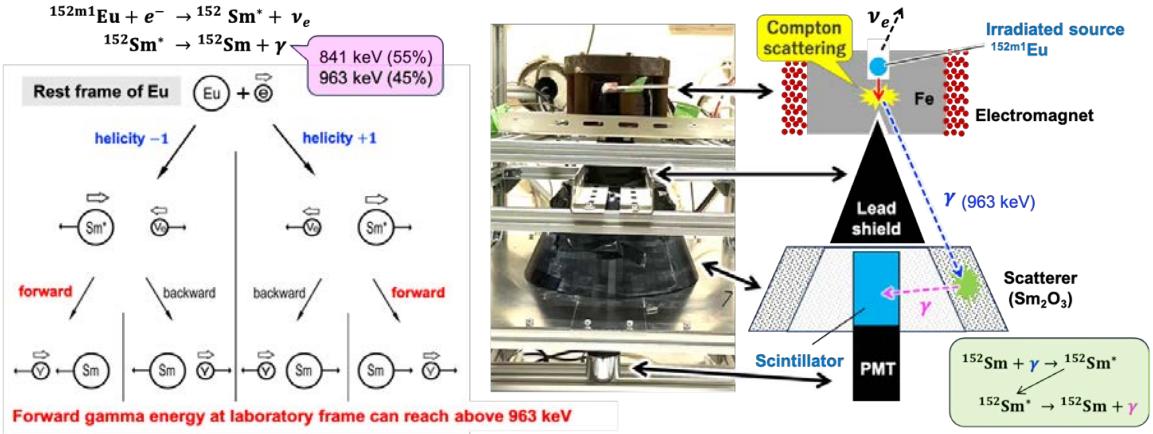

Fig. 1. Experimental principle and setup to measure the neutrino helicity.

RESULTS: Figure 2 shows measured asymmetry of the energy spectrum based on the modulation. This result favors negative helicity. The obtained helicity is –1.5 ± 0.9 which is statistically consistent with the previous study, –1.0 ± 0.3 [1].

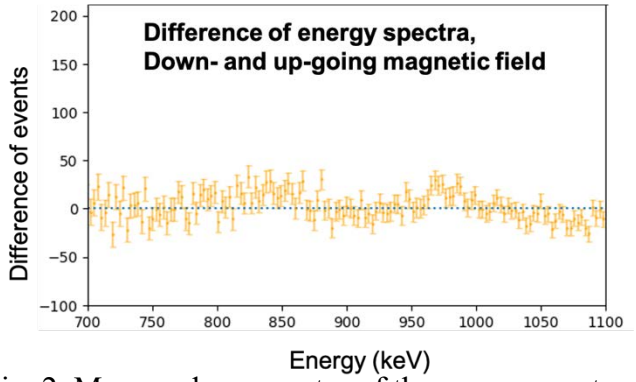

Fig. 2. Measured asymmetry of the energy spectrum.

REFERENCES:

[1] M. Goldhaber, L. Grodzins, and A. W. Sunyar, Phys. Rev., **109** (1958) 1015.