

Measurement of Natural Uranium Samples Using Fast Fission Neutron Coincidence Counting

M. Maeda¹, M. Komeda¹, Y. Toh¹, Y. Takahashi², Y. Kitamura² and T. Misawa²

¹*Nuclear Science and Engineering Center, Japan Atomic Energy Agency*

²*Institute for Integrated Radiation and Nuclear Science, Kyoto University*

INTRODUCTION: We have been developing a Fast Fission neutron Coincidence Counting (FFCC) method applicable to nuclear materials containing neutron absorbers, such as fuel debris at the Fukushima Daiichi Nuclear Power Plant and concealed nuclear materials in the field of nuclear security [1]. In the FFCC method, samples are irradiated with fast neutrons, which are less affected by neutron absorbers, and the amount of nuclear material is estimated by measuring neutrons generated by fast neutron-induced fission. The method has previously been demonstrated experimentally for samples on the order of several tens of grams. In this study, measurements were conducted to investigate the response of the FFCC method for kilogram-scale uranium samples.

EXPERIMENTS: Figure 1 shows the experimental setup. A natural uranium sample was placed at the center, with four EJ-301 liquid scintillators (5-inch diameter and 5-inch length) arranged on both sides and a D–D neutron generator positioned behind the sample. A polyethylene shield was placed between the neutron generator and the scintillators to prevent direct neutron incidence. The sample was irradiated with neutrons, and the emitted fission neutrons were detected by the scintillators. Measurements were performed with a neutron intensity on the order of 10^5 n/s for uranium masses of 0 kg (blank), 1 kg, 5 kg, 10 kg, and 20 kg.

RESULTS: In the FFCC method, the time difference between neutron detection events for each detector pair is calculated [1]. Figure 2 shows a histogram of the time-difference event for a 10 kg uranium sample (time-difference spectrum). Fission neutrons produce a peak near 0 ns, while neutrons scattered between detectors (crosstalk) produce peaks shifted from 0 ns. As a result, three peaks appear in the time-difference spectrum. These results confirm that coincidence events originating from fission neutrons were successfully detected. In addition, measurements for other samples showed variations in the fission neutron count depending on the uranium mass. These results suggest that the FFCC method is applicable to uranium samples on the kilogram scale.

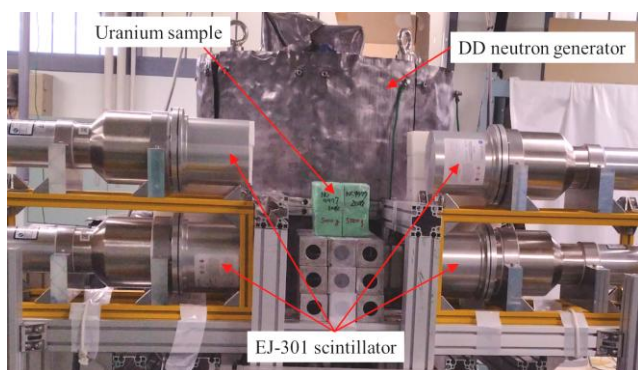


Fig. 1. Experimental setup.

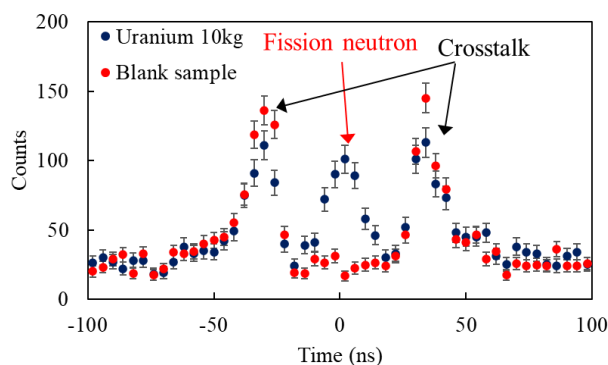


Fig. 2. Time-difference spectrum.

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Neutron resonance analysis technique in neutron time-of-flight for highly enriched uranium

F. Rossi¹, J. Lee¹, Y. Yoshimi¹, K. Hironaka¹, T. Shiba¹, K. Terada², J. Hori²

¹Integrated Support Center for Nuclear Nonproliferation, Security and Human Resource Development (ISCN), Japan Atomic Energy Agency (JAEA)

²Institute for Integrated Radiation and Nuclear Science, Kyoto University

INTRODUCTION: The ISCN of the JAEA is developing a combined neutron resonance analysis (NRA) system aims to identify and quantify fissile material in an unknown sample. Neutron resonance transmission analysis (NRTA) and neutron resonance capture analysis (NRCA) are used together with the proposed neutron resonance fission neutron analysis (NRFNA) [1,2,3]. 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 with plastic scintillators able to perform n/γ pulse shape discrimination (PSD). In this paper, we report a demonstration experiment for highly enriched uranium samples performed at KURNS-LINAC.

EXPERIMENTS: The KURNS-LINAC operated with an acceleration energy of ~30 MeV. Pulsed neutrons were produced with a repetition rate of 50 Hz and a pulse width of 2 μs. Enriched uranium (²³⁵U: 93%) foils were used with thickness up to 6 mm. Between the beam exit and the sample, a borated polyethylene and lead collimator with an aperture of 2.0 cm was used. At the sample position, an assembly of ten hexagonal PSD plastic scintillation detectors (Eljen, EJ-276D) were used for NRFNA and NRCA with a 3.0 cm lead shielding. Between the sample and the NRTA detector (⁶Li-glass, GS20), a second collimator with an aperture of 1.0 cm was placed. The setup used is shown in Fig. 1. 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.



Fig. 1. Experimental setup.

RESULTS: Obtained data are being analyzed by an advanced PSD technique [4]. Fig. 2 shows the TOF spectra of 6 mm thick enriched uranium foils. To be noted, the GS20 spectrum is shifted in time, due to the further position of the detector from the sample. As in the case of shielded samples [5], the characteristics peaks from ²³⁵U(n,f) reaction are observed at all energies in the neutron spectrum; a linear mass correlation can be obtained after normalization for beam, intensity variation. In the gamma-ray spectrum, the 6.67 and the 20.87 resonance peaks from ²³⁸U(n,γ) reaction are visible and can be used to evaluate sample self-shielding effects. This experiment confirmed the possibility not only to identify fissile nuclides, but, more importantly, to quantify them.

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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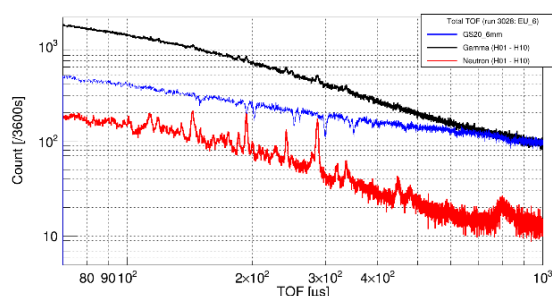


Fig. 2. TOF spectra of 6 mm thick enriched uranium: neutron (red), gamma (black), GS20 (blue).

Basic Study on Improving Experimental Accuracy in the Fast Energy Range of Neutron Cross Section Measurement Using KURNS-LINAC

T. Sano¹, J. Hori², H. Yashima², Y. Takahashi², K. Terada²

¹Atomic Energy Research Institute, Kindai University

²Institute for Integrated Radiation and Nuclear Science, Kyoto University

INTRODUCTION:

We have been conducting neutron cross section measurements for nuclear fuel materials and minor actinids by using KURNS-LINAC [1]. Those experimental studies have been performed with time-of-flight method (TOF method). Because KURNS-LINAC uses a photo-neutron source, measurements of neutron cross section above approximately 1 keV in the TOF method exhibited large uncertainties due to the influence of bremsstrahlung X-rays generated during photo-neutron production called “gamma-flash”. In particular, those uncertainties have a significant impact on the resonance analysis performed using the measurement data. Therefore, in this study, we focused on an iron filtering system to reduce gamma-flash and improve the accuracy of measurement data at energies of 1 keV or higher. The iron filtering system utilizes neutron cross-section windows in natural iron at 20, 73, and 82 keV to extract quasi-monochromatic neutron flux and had been installed the B-1 beam hole in KUR. In particular, the use of iron filter beam at JPARC MLF/ANNRI has yielded significant results in the study of neutron cross section measurement in the fast energy range [2]. In this study, an iron filtering system has been installed and characteristics of filtered neutron beam were measured.

EXPERIMENT:

The iron filtering system has been installed on end of 12m neutron flight path. The iron filter has 2 inch length. The filtered neutron beam was measured by TOF method. A neutron detector was employed Li-glass detector. In the experiments, a measurement of the iron filtered beam and the neutron beam with resonance filters (In, Ag, Mn, Co) were performed.

RESULTS:

Figure 1 shows obtained TOF spectra. A red line is the iron filtered beam and a blue line is the neutron beam with the resonance filters. Here, the TOF spectrum obtained using the resonance filter was used as a reference to determine the energy of the TOF spectrum obtained with the iron filtering system. Furthermore, the dips caused by saturation resonance (black arrow) are considered to represent the background level. As the results, the quasi-monochromatic neutron flux in the energies of 73 keV and 20 keV were obtained.

In those experiments, we successfully extracted quasi-monochromatic neutron beams with energies of 73 keV and 20 keV using the iron filtering system in the KURNS-LINAC pulsed neutron source.

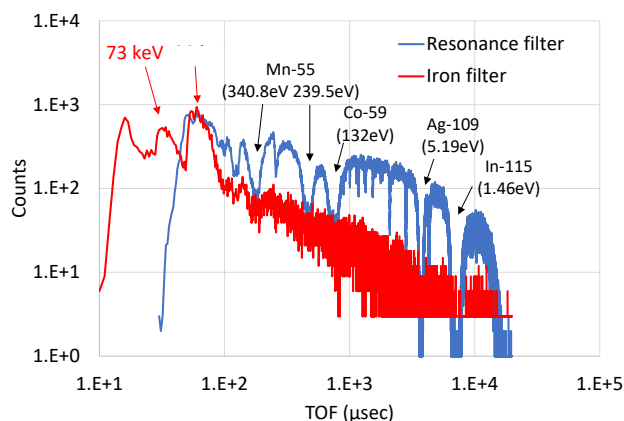


Fig.1. Measured TOF spectrum

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Search for Tetraneutron with an NAA-like Approach for Magnesium

H. Fujioka¹, S. Oikawa¹, K. Takamiya²

¹ *Department of Physics, Institute of Science Tokyo*

² *Institute for Integrated Radiation and Nuclear Science, Kyoto University*

INTRODUCTION: Compared to typical nuclei composed of both protons and neutrons, the tetraneutron is composed of only four neutrons. Motivated by a recent observation of a possible tetraneutron bound state [1], we have been searching for its experimental signature at KUR. A tetraneutron bound state, if it exists, would travel a sufficiently long distance, and have a chance to induce a ($^4\text{n},\text{n}$) reaction with a nucleus (AZ), leading to the production of ^{A+3}Z . This product can be identified by detecting γ rays from the β -decay of ^{A+3}Z with a germanium detector. This approach resembles neutron activation analysis (NAA), in which a nuclide (AZ) in a sample is identified and quantified by detecting γ rays from ^{A+1}Z produced via neutron capture.

Recently, theoretical calculations of tetraneutron-induced reaction cross sections have been made possible using an optical and statistical Hauser-Feshbach model code, CoH3 [3]. The calculations reveal that the $^{26}\text{Mg}(^4\text{n},2\text{n})^{28}\text{Mg}$ cross section is exceptionally large, around 10 barns, compared to typical ($^4\text{n},\text{n}$) cross sections.

EXPERIMENTS: We conducted a tetraneutron search experiment using two samples, ^{26}Mg -enriched ^{26}MgO powder and high-purity magnesium pellets. These samples were irradiated at the hydraulic conveyer (Hyd) for ten and one hours, respectively, at the KUR thermal power of 1 MW. As an example, the γ -ray spectrum for the ^{26}MgO sample is shown in Fig. 1. Although many photopeaks from impurities were identified, photopeaks from ^{28}Mg were not observed. Based on this result, we will derive an upper limit on ^4n emission rate in uranium fission.

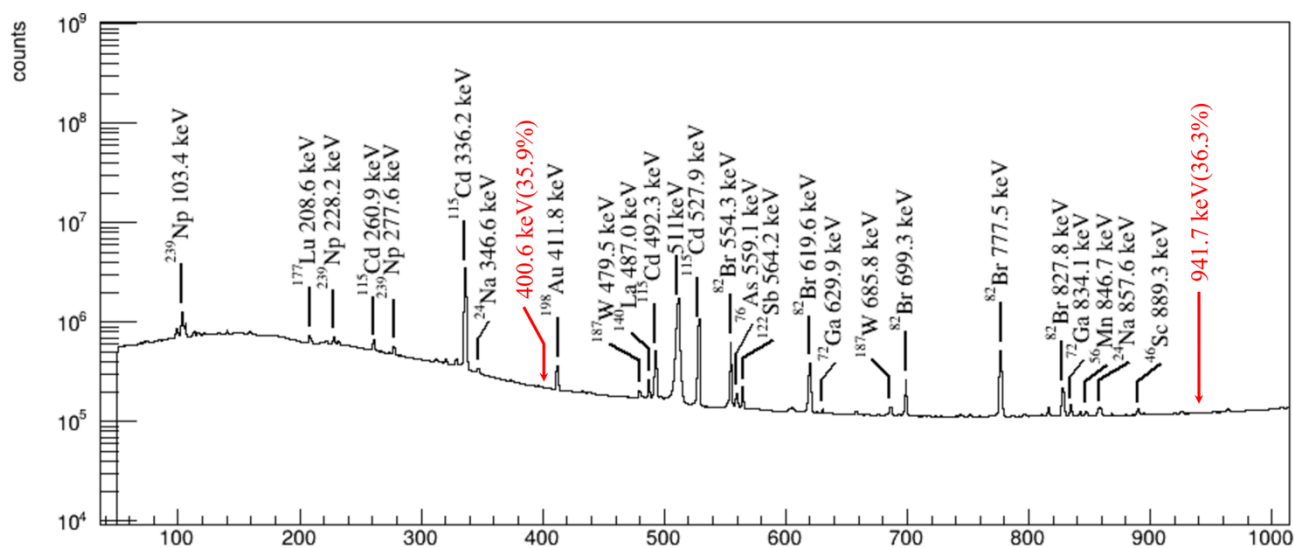


Fig. 1 The γ ray spectrum for the ^{26}MgO sample.

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

T. Matsumoto¹, A. Masuda¹, S. Manabe¹, H. Harano¹, T. Takata², H. Tanaka², K. Terada² and J. Hori²,

¹National Metrology Institute of Japan, National Institute of Advanced Industrial Science and Technology

²Institute for Integrated Radiation and Nuclear Science, Kyoto University

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. Then, we are developing an ionization chamber for measuring high-intensity epi-thermal neutrons with fluence rate of $10^9 \text{ cm}^{-2}\text{s}^{-1}$ or higher in the BNCT neutron field. As part of the characterization of these chambers, we measured their voltage characteristics.

EXPERIMENTS: The ionization chambers are thimble-type and consist of an aluminum chamber and an aluminum needle. One chamber is filled with ^3He (0.1 atm.) and Kr (0.9 atm.) gases, while another has ^6LiF deposited on its inside walls. Tests on these ionization chambers were conducted using a pulsed white neutron source based on an electron linac (KURNS-LINAC) and at the KUR heavy water facility. The relative energy responses were measured using the time-of-flight method with a pulsed white neutron source. The voltage characteristics were evaluated at KUR, where a high-intensity neutron flux is available.

RESULTS: Fig. 1 shows the results of applied voltage vs output current for ionization chambers with ^3He gas and ^6LiF foil. These results were obtained by applying a voltage to the outer electrode and extracting current from the central pin. It can be seen that the two types of ionization chambers exhibit the same trend for the same voltage. Previous measurements were conducted in the range of 100 V to 1000 V, and the results were found to be constant. A clear transition from the recombination region to the ionization region was observed. It was found that the device can be used as an ionization chamber at voltages generally 50 V or higher. Moving forward, we will evaluate the effects of dark current and gamma rays.

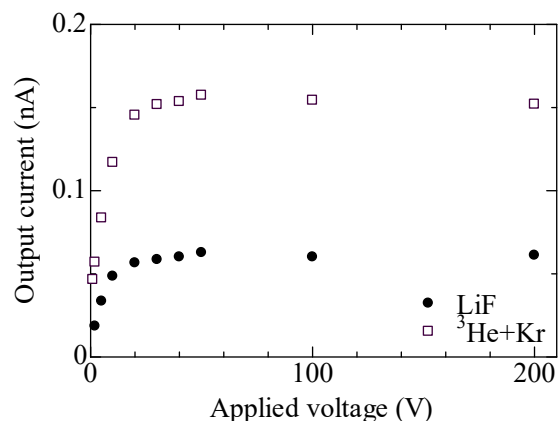


Fig. 1. Results of output current vs applied voltage for ionization chambers with ^3He gas and ^6LiF foil.

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Neutron Capture Cross-Section Measurements at TC-Pn in KUR for Tungsten among Nuclides in Decommissioning

S. Nakamura¹, Y. Shibahara², A. Kimura¹, S. Endo¹, R. Gerard¹

¹Nuclear Data Center, Japan Atomic Energy Agency

²Institute for Integrated Radiation and Nuclear Science, Kyoto University

INTRODUCTION: Regarding the nuclides of concern at clearance levels [1] in decommissioning, thermal-neutron capture cross-sections have been measured by an activation method using the thermal column pneumatic tube (TC-Pn) of KUR [2-7]. The present work selected ¹⁸⁶W nuclide and measured the thermal-neutron capture cross-section for production of ¹⁸⁷W with a clearance level of 10 Bq/g [1].

EXPERIMENTS: Tungsten (W) samples were prepared as 99.95% pure, 5- μ m-thick metal foils. Neutron irradiation for one hour using TC-Pn at 1-MW operation was performed three times as follows: I) The neutron flux was measured using Au and Co monitors to determine the ¹⁸⁶W cross-section. II) The measurement of ¹⁸⁶W was repeated using a simple comparison method with Au. III) To confirm the accuracy of the obtained ¹⁸⁶W cross-section, the W sample was irradiated as a neutron flux monitor together with Au and Co monitors. After neutron irradiation, the γ -rays emitted from each sample were measured using a high-purity Ge detector installed at the Hot Laboratory in KUR. The sample was placed at a distance of 110 mm from the front surface of the Ge detector. The γ -ray peak efficiencies of the Ge detector were measured beforehand with a mixed γ -ray source and a γ -ray reference source of ¹⁵²Eu. Gamma-rays emitted from ¹⁸⁷W were measured in sufficient yields. Six γ -rays emitted from ¹⁸⁷W were observed at γ -ray energies of 479, 551, 618, 625, 685 and 772-keV.

ANALYSIS AND RESULTS: The reaction rates of the flux monitors and W samples were derived from the γ -ray yields. Based on the *Westcott's* convention[8], the thermal-neutron capture cross-section of ¹⁸⁶W was derived using the neutron flux results and the reaction rates of ¹⁸⁶W. The result of cross-sections of ¹⁸⁶W were found to be 43.5 ± 1.2 barns for the irradiation I, which was 14% larger than the evaluated value of 38.1 ± 0.5 barns [9]. In irradiation II, a value of 43.1 ± 1.3 barns was obtained, confirming the reproducibility of the result from the irradiation I. Furthermore, when the neutron flux was measured using W as a monitor in irradiation III, the results were obtained as shown in **Figure 1**. The ¹⁸⁶W cross-section obtained in this study gives a neutron flux which is consistent with the neutron fluxes obtained with the Au and Co monitors.

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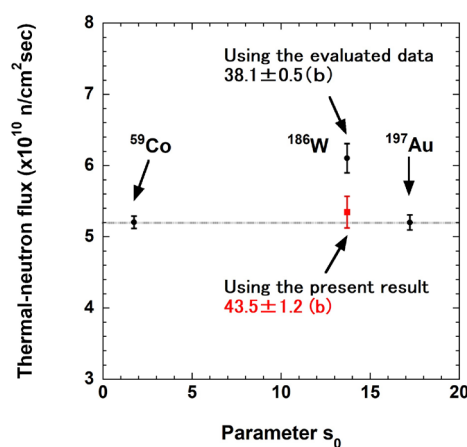


Figure 1 Thermal-neutron flux measured using the ¹⁸⁶W thermal-neutron capture cross section obtained in this study.

Progress in measurements of the response of a GAGG Scintillator to monoenergetic electron irradiation at KURNS-LUNAC

T. Niwase¹, T. Fujii², Y. Hirayama³, N. Kitagawa², N. Miyashita², G. Otsuka², S. Sakaguchi², R. Shikada², A. Taniguchi⁴, H. Watanabe², Y. X. Watanabe³ and Y. Yamanouchi^{2,3}

¹ Department of Physics, Meiji University, Kawasaki, Kanagawa 214-8571, Japan

² Department of Physics, Kyushu University, Nishi-ku, Fukuoka 819-0395, Japan

³ Wako Nuclear Science Center, Institute of Particle and Nuclear Studies, High Energy Accelerator Research Organization (KEK), Wako, Saitama 351-0198, Japan

⁴ Institute for Integrated Radiation and Nuclear Science (KURNS), Kyoto University, Kumatori 590-0494, Japan

INTRODUCTION: GAGG (Ce:Gd₃(Al, Ga)₅O₁₂) scintillators[1] have attracted considerable attention for radiation detection because of their high light yield, good energy resolution, and non-deliquescent nature. Owing to these properties, we are investigating the potential of GAGG scintillators for high-precision β -ray measurements aimed at elucidating the decay modes of nuclei in the termination region of the r-process.

EXPERIMENT: In this study, low-intensity electron beams with energies of 4–8 MeV, provided by KURNS-LINAC, were directly irradiated onto the GAGG scintillator in order to investigate its response to monoenergetic electrons.

RESULTS: The spectrum obtained with 4 MeV electron irradiation is shown in Fig. 1 (blue line). To suppress the γ -ray background in the experimental room, a plastic scintillator was placed in front of the GAGG scintillator. By requiring coincidence between the two detectors, electrons incident on the GAGG scintillator were identified. The spectrum obtained in coincidence with the plastic scintillator is shown in magenta in Fig. 1. By combining the experimental data with Monte Carlo simulations based on the Geant4 toolkit[2], we extracted the single-electron component while taking the contribution of each spectral component into account. Figure 2 shows the extracted single-electron spectra for incident energies of 4–8 MeV. Further analysis will be carried out to evaluate the detector response function in detail.

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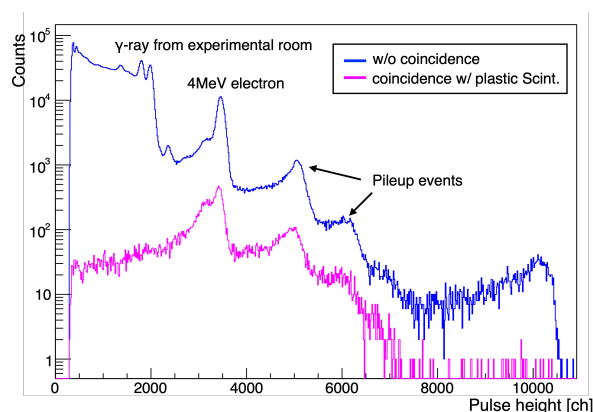


Fig. 1. Singles spectrum (blue) and coincidence spectrum with the plastic scintillator for 4 MeV electron irradiation.

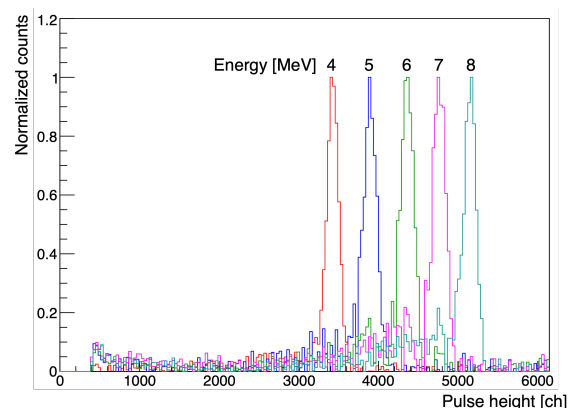


Fig. 2. Singles-electron spectra for incident energies of 4-8MeV, with normalized intensities.

Measurements of neutron capture cross sections of ^{78}Se at KURNS-LINAC

K. Terada¹, Y. Takahashi¹, H. Yashima¹, J. Hori¹, T. Sano²

¹ *Institute for Integrated Radiation and Nuclear Science, Kyoto University*

² *Kindai University*

INTRODUCTION: The disposal of long-lived radioactive waste generated from nuclear power plants remains a critical issue. Nuclear transmutation of long-lived fission products (LLFPs) via neutron capture reactions is expected to reduce the environmental burden associated with geological disposal [1]. Selenium-79, which has a half-life of 3.27×10^5 years [2], is one such LLFP targeted for transmutation. In transmutation systems without isotopic separation, stable Se isotopes ($^{74}, ^{76}, ^{77}, ^{78}, ^{80}, ^{82}\text{Se}$) coexist with ^{79}Se ; therefore, neutron capture cross section data for the Se stable isotopes are also required, as they influence the overall transmutation efficiency of the system. In this study, measurements of neutron capture cross section for ^{78}Se , one of the stable isotopes of selenium, were performed at the Kyoto University Institute for Integrated Radiation and Nuclear Science - Linear Accelerator (KURNS-LINAC).

EXPERIMENTS and RESULTS: We measured neutron capture cross sections of ^{78}Se using the time-of-flight (TOF) method at the KURNS-LINAC. Electron beams with an energy of 37 MeV, an average beam current of 10 μA , a pulse width of 100 ns, and a repetition rate of 100 Hz were bombarded onto a water-cooled tantalum target. Neutrons were produced by photo-nuclear reactions induced by bremsstrahlung X-rays and were moderated by a light water moderator (17 cm diameter, 30 cm height). Moderated neutrons were guided to the experimental area at 135 degrees relative to the electron beam direction. An enriched ^{78}Se sample (20 mm diameter x 1 mm thickness, 99.6% enrichment, net weight 2.0 g) was placed 12.1 m from the neutron source. Neutron capture gamma-rays emitted from the ^{78}Se sample were detected using an array of 12 BGO detectors (2-inch diameter \times 2-inch length). Detector signals were digitized using a CAEN V1730S module to record two-dimensional data of TOF and Pulse Height (PH). Figure 1 shows the measured TOF spectra of the ^{78}Se sample and the blank (without sample), respectively. A 383 eV resonance peak of ^{78}Se was observed at 44.5 μs . Furthermore, an enriched ^{10}B sample was used to determine the incident neutron energy spectrum by counting 478 keV gamma-rays via $^{10}\text{B}(n,\alpha)^7\text{Li}$ reactions. Neutron capture yields were subsequently derived by subtracting both sample-dependent and sample-independent backgrounds.

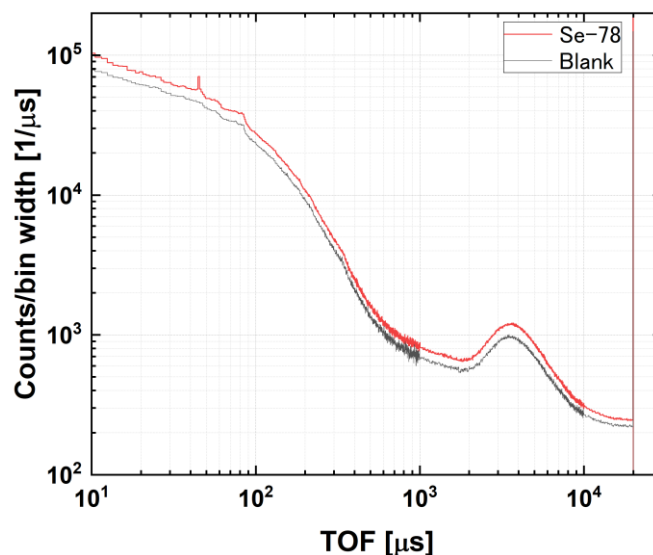


Fig. 1 TOF spectra of ^{78}Se (red) and Blank (black). The x-axis is TOF. The y-axis is counts per bin width.

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Sub-nanosecond Lifetime Measurements of Nuclear Excited States Using LaBr₃(Ce) Detectors

M. Shibata¹, Y. Okuda², A. Taniguchi³

¹ Radioisotope Research Center, Nagoya University

² Graduate School of Engineering, Nagoya University

³ Institute of Integrated Radiation and Nuclear Science, Kyoto University

INTRODUCTION: Unstable isotopes with β -decay half-lives of several days or longer are widely used in medical and scientific applications; however, for some nuclides, nuclear data have not been sufficiently validated. Sub-nanosecond lifetimes of excited states often show large uncertainties or inconsistencies among reported values. In this study, lifetimes of selected excited states were remeasured using β - γ and γ - γ delayed coincidence techniques with LaBr₃(Ce) detectors, which offer both high time and energy resolution. The target nuclides include ¹⁷⁷Hf, ¹⁴¹Pr, ⁷⁵As, and ¹⁰³Rh, aiming at improving the reliability of nuclear data.

EXPERIMENTS: The parent nuclides ¹⁷⁷Lu ($T_{1/2} = 6.6$ d), ⁷⁵Se ($T_{1/2} = 119.8$ d), ¹⁴¹Ce ($T_{1/2} = 32.5$ d), and ¹⁰³Ru ($T_{1/2} = 39.2$ d) were produced via (n, γ) reactions using the Kyoto University reactor (1 MW, neutron flux: 1.6×10^{13} n cm⁻² s⁻¹). Source activities during measurements were below 5 kBq. Measurements were conducted using two plastic scintillators (detectors 1 and 2: 50 mm \times 20 mm and 30 mm \times 30 mm \times 1 mm) and two LaBr₃(Ce) detectors A and B (38 mm \times 8 mm) as described in Table 1. The time resolution was 1–2 ns for 100 keV γ rays.

RESULTS: For the 113 keV level of ¹⁷⁷Hf, lifetimes of 0.550(5) ns were obtained, consistent with the evaluated value. For the 321 keV level, 0.780(7) ns was obtained, supporting the recent result of 0.792(17) ns [2] rather than the evaluated value [1] (Fig. 1). The 145 keV level of ¹⁴¹Pr was measured for the first time as 1.85(1) ns, in agreement with evaluation [3]. For ⁷⁵As (199 keV), 0.888(20) ns was obtained from γ -ray coincidence, consistent with previous results [4]. For ¹⁰³Rh (93 keV), the first LaBr₃-based measurement yielded 1.15(7) ns, also consistent with evaluation [5]. These results are preliminary but demonstrate improved precision using LaBr₃(Ce) detectors. Remeasurement with modern techniques is important for refining nuclear data.

Table 1. Target nuclides, excited levels, detector combinations, and evaluated values.

Nuclide	Level (keV)	Detector combination	Evaluated value (ns)
¹⁷⁷ Hf	113	plastic (1, 2) + LaBr ₃ (A)	0.541(14) [1]
	321	plastic (2) + LaBr ₃ (A)	0.665(16) [1]
¹⁴¹ Pr	145	plastic (2) + LaBr ₃ (A)	1.85(3) [3]
⁷⁵ As	199	LaBr ₃ (A, B)	0.885(30) [4]
¹⁰³ Rh	93	LaBr ₃ (A, B)	1.11(3) [5]

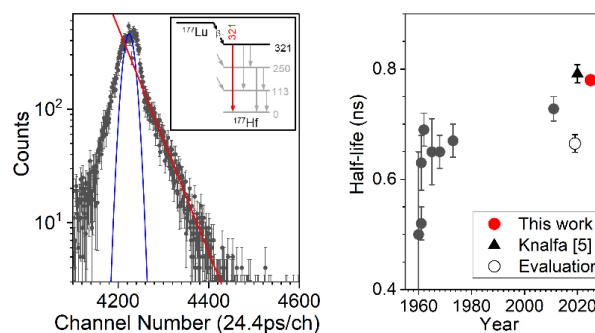


Fig. 1 Delayed coincidence time spectrum for the 321 keV level of ¹⁷⁷Hf (left) and comparison of the measured lifetime with previous and evaluated values (right).

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Study on Improving the Accuracy of Fission Cross-section Measurement

J. Hori¹, K. Terada¹, H. Yashima¹, Y. Takahashi¹ and T. Sano²

¹ *Institute for Integrated Radiation and Nuclear Science, Kyoto University*

² *Atomic Energy Research Institute, Kindai University*

INTRODUCTION: In the development of advanced nuclear reactors, improving the accuracy of fission cross sections for minor actinides (MAs) is one of the critical issues. However, the accuracy of fission cross-section data of MAs is currently insufficient to fully meet engineering requirements. That is because the fission cross-section measurement with a fission chamber has large uncertainties due to strong alpha decay background and the limitation of target mass imposed by the fission chamber. To overcome these difficulties, we aim to apply another technique that detects prompt neutrons emitted by fission reaction with a recoil proton type neutron detector for measurement of fission cross sections. We constructed a full solid-angle detector system and conducted performance tests on it at KURNS-LINAC TOF beam line.

EXPERIMENT: The linac was operated with a pulse width of 1 μ s, a repetition rate of 50 Hz, 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. A high enriched uranium foil (U_3O_8 -Al alloy, 93% U-235) used as a sample was set in the neutron beam line at a distance of 11.25 m from the neutron target. The sample was surrounded with five recoil proton type neutron detectors (EJ-276 plastic scintillator) and five BGO detectors. The signals from the detectors were digitized using a CAEN V1730D module. In order to distinguish between gamma and neutron signals, the Neutron/Gamma pulse shape discrimination (PSD) method was applied [1]. On the other hand, the 478-keV gamma rays from a ^{10}B sample were also measured with BGO detectors for determination of neutron flux at the sample position.

RESULTS: Typical two-dimensional histogram of pulse height and PSD for the high enriched uranium sample measurement is shown in Fig. 1. The PSD value was defined as a ratio of the differences in charge integration between short and long gates. Neutron-induced events could be separately observed in the PSD region from 0.35 to 0.45. By gating on the neutron-induced PSD region, we have obtained the TOF spectrum corresponding to the fission events of U-235. In this study, we implemented an advanced data-driven PSD method, enhancing the analysis of neutron and gamma signals for fission cross-section measurements.

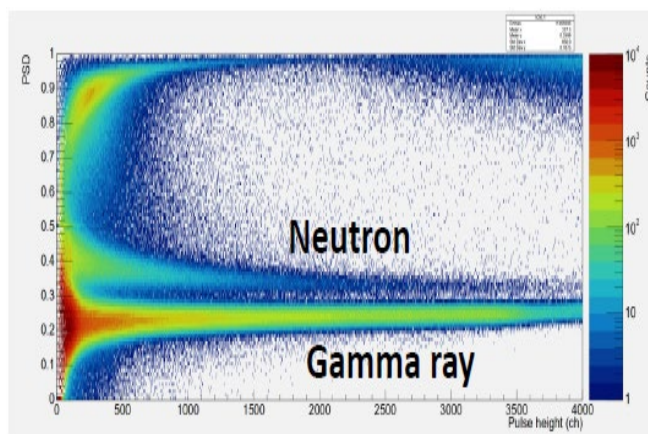


Fig. 1 2D histogram of pulse height and PSD

Acknowledgment

This work was supported by MEXT Innovation Nuclear Research and Development Program Grant Number JPMXD0224020564.

REFERENCES:

[1] CAEN S.p.A., Digital Pulse Processing for Pulse Shape Discrimination (DPP-PSD) User Manual (2017).