

VIII-II-1. Project Research

Project 2

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OBJECTIVE and RESEARCH SUBJECTS:

The main objective of this research project is to improve the accuracy of nuclear data (cross sections) for the design of innovative reactors through the experimental research for the MA and LLFP nuclides. This project consists of six research subjects from five institutions, as follows.

21P2-1: A Study on Neutron-Capture Reactions by High-Energy Gamma-ray Spectroscopy (H. Harada et al., JAEA)

21P2-2: Study on Neutron Capture Cross Sections of MA and LLFP Nuclei using Various Detectors (J. Hori et al., Kyoto Univ.)

21P2-3: MLPPAC(Multi-Layer Parallel Plate Avalanche Counter) for Measurements of Fission Cross-sections of Minor Actinides [Cross-section Measurements for Neutron-induced Fission of Curium Isotopes] (T. Ohtsuki et al., Tohoku Univ.)

21P2-4: Isotopic Analysis of ^{107}Pd Samples (T. Fujii et al., Kyoto Univ.)

21P2-5: Study on Nondestructive Analysis and Neutron Behavior Using Photonuclear Reaction (K. Ishibashi, et al., Kyushu Univ.)

21P2-6: Study on Neutron Energy Spectrum Measurements for Pulsed Neutron Source (K. Nakajima et al., Kyoto Univ.)

MAIN RESULTS and CONTENTS of this REPORT:

H. Harada et al. (21P2-1) have studied the resonance dependence of γ -ray intensity distributions using the 4π Ge spectrometer. The γ -ray intensity distributions were deduced by unfolding γ -ray pulse-height spectra with the response functions of the spectrometer. The response functions were calculated by using Monte Carlo simulation code EGS5. It was found that the gross structure of the γ ray intensity distributions does not change significantly for ^{238}U resonances, although some discrete γ ray yields strongly depend on the resonance.

J. Hori et al. (21P2-2) have studied on neutron capture cross sections of MA and LLFP nuclei using a pair of C_6D_6 detectors, a 4π Ge spectrometer and a total absorption BGO spectrometer. The TOF spectrum of ^{243}Am with a pair of C_6D_6 detectors was compared with that

with a 4π Ge spectrometer. The resonance analyses of the neutron capture cross sections of ^{105}Pd and ^{108}Pd obtained with the BGO spectrometer were carried out with the SAMMY code.

T. Ohtsuki et al. (21P2-3) have developed the multi-layered parallel plate ionization chamber (MLPPIC) to measure the fission cross sections for MA nuclides. In the present experiment, cross sections for curium isotopes, the third abundant minor actinide in the high level radioactive waste after neptunium and americium isotopes, were measured using the lead slowing-down neutron spectrometer at KURRI. The fission cross sections for ^{245}Cm and ^{248}Cm were successfully obtained, and they were compared with the past measurements.

T. Fujii et al. (21P2-4) have investigated the preparing method to obtain highly pure target samples of MA and LLFP nuclides for nuclear data measurements. In the present study, the chemical and isotopic purities of the raw materials of ^{107}Pd were analyzed by absorption spectrophotometry and mass spectrometry. Since the ^{107}Pd sample consists of stable isotopes, ^{102}Pd , ^{104}Pd , ^{105}Pd , ^{106}Pd , ^{108}Pd , and ^{110}Pd , and the LLFP ^{107}Pd , in which only estimated isotopic compositions are known. The results show that the sample includes no chemical impurities and contains 15.3% of the LLFP ^{107}Pd .

K. Ishibashi et al. (21P2-5) examined the feasibility of nondestructive analysis of "difficult to measure" nuclei in a nuclear waste. They proposed to measure the gamma-ray generated by photonuclear reactions using for the "difficult to measure" nuclides using an electron linac. In the present study, ^{99}Tc as a "difficult to measure" nucleus has been examined. The average photonuclear cross section of $^{99}\text{Tc}(\gamma,3n)^{96}\text{Tc}$ between 26 and 36 MeV was measured, and the shape of cross section (energy dependence) was predicted.

K. Nakajima et al. (21P2-6) have investigated to the method to reduce the signal distortion caused by the gamma burst generated when the pulsed neutron produced, in order to expand the energy range of measurements. In the present study, the digital data processing was adopted for the distorted signals. The results shows that the digital data processing can almost vanishes the distortion in the signal and hence the energy range of the TOF method expanded from 1keV to several tens of keV.

PR2-1 Study of Neutron-Capture Reactions by High-Energy Gamma-ray Spectroscopy

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INTRODUCTION: Accurate neutron capture cross sections are required for the study of an innovative nuclear system. To satisfy the demand, a 4π Ge spectrometer was developed, and its performances were studied in the course of capture cross section measurements using time-of-flight (TOF) techniques at the electron linear accelerator of KURRI. The 4π Ge spectrometer is a unique tool due to its superior energy resolution for studying properties on neutron capture reactions, not only capture cross sections but also γ -ray intensity distributions. Precise understanding of resonance dependence or neutron energy dependence of γ -ray intensity distributions is useful to reduce uncertainties of neutron capture cross sections. In this study, the γ -ray intensity distributions were deduced using the 4π Ge spectrometer for $^{238}\text{U}(n, \gamma)$ reaction.

EXPERIMENTS: The 4π Ge spectrometer was set at the 10 m neutron flight path of the electron linear accelerator of KURRI as shown in Fig.1 A natural Uranium sample (11.05g) was set in the center of the spectrometer system. The accelerator was operated with electron energy of 30MeV, an average beam current of 64 μA , a repetition rate of 50Hz and a pulse width of 3.0 μs .

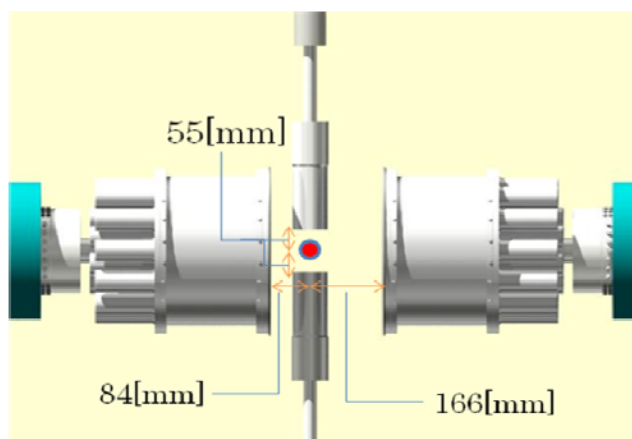


Fig. 1. Experimental setup.

RESULTS: Figure 2 shows γ ray spectra gated by the neutron resonances at 6.67, 20.87 and 36.68eV of ^{238}U . The gross shapes of the spectra are almost identical. Fig. 3 shows γ ray intensity distributions deduced by unfolding the spectra in Fig. 2 with the response functions of the 4π Ge spectrometer. The response functions were calculated by using Monte Carlo simulation code EGS5. It is informative that the gross structure of the γ ray intensity distributions does not change significantly for these ^{238}U resonances, although some discrete γ ray yields strongly depend on the resonance as was reported in Ref [1].

The response functions will be also used in future to deduce neutron capture cross sections in combination with a pulse-height weighting technique. As an example, the deduced weighting function using the response functions for the cluster Ge detector is shown with arbitrary unit in Fig. 2.

It was demonstrated that the 4π Ge spectrometer is a powerful tool to deduce γ -ray intensity distributions as a function of neutron energy.

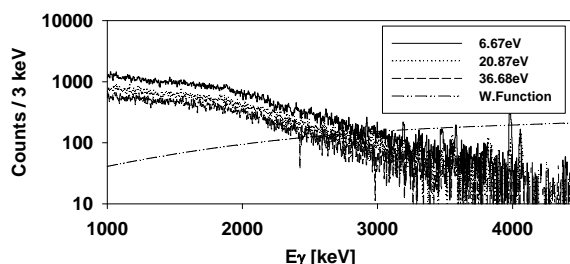


Fig. 2. γ ray spectra for ^{238}U resonances.

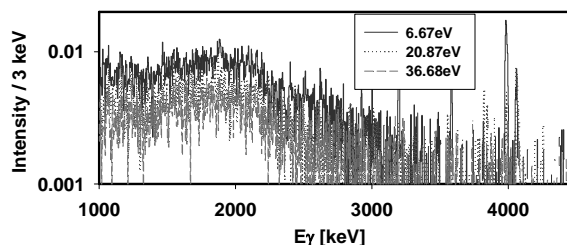


Fig. 3. γ ray intensity distributions for ^{238}U resonances.

Present study includes the result of “Study on nuclear data by using a high intensity pulsed neutron source for advanced nuclear system” entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

REFERENCES:

[1] S. Goko, *et al.*, Conference record of 2009 Fall Meeting of AESJ, J18 (2009) (Japanese).

J. Hori and H. Yashima

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INTRODUCTION: Accurate neutron capture cross section data of minor actinides (MAs) and long-lived fission products (LLFPs) are required for the study of transmutation system. However, accurate measurement of these nuclei is a very difficult task due to high radioactivity and/or low isotopic purity of sample. To overcome these difficulties, it is important to select a detector suitable for the objective nuclide.

In the case of MA with a high radioactivity, we have selected a pair of C_6D_6 detectors characterized by a small efficiency and a low sensitivity to the scattered neutron. It is expected that the dead time due to radioactivity and the background due to neutrons scattered from the sample can be reduced.

In the case of LLFP with a low isotopic purity, we have selected a 4π Ge spectrometer characterized by a high energy resolution. It can be applicable to eliminate backgrounds due to impurities in the sample. We have demonstrated the nuclide discrimination technique with a 4π Ge spectrometer as shown in Ref [1, 2].

In the case of a small amount of stable FP nuclide with a high isotopic purity, we have selected a total absorption $Bi_4Ge_3O_{12}$ (BGO) spectrometer characterized by a large efficiency and a high sensitivity to the scattered neutron. The experimental results were reported in Ref [1].

In this paper, we will report the experiment on ^{243}Am with a pair of C_6D_6 detectors and the resonance analyses on the capture cross sections of Pd stable isotopes obtained using a BGO spectrometer.

EXPERIMENTS and RESULTS: The measurements have been performed by the neutron TOF method using an electron linear accelerator (linac) at the KURRI.

A pair of C_6D_6 liquid scintillators, each 11 cm in diameter and 5 cm thick, was used for the measurement of capture γ -rays from the ^{243}Am sample, which was put in the neutron beam between the scintillators. Americium oxide (AmO_2) powder of 0.128 g (835 MBq) packed in an aluminum disk container of 30 mm in diameter was purchased from Russia Research Institute of Atomic Reactors. To decrease the constant background due to radioactivity, a 10 mm thick lead shield was set on the surface of each detector. The sample was placed at a distance of 12.0 ± 0.02 m from the Ta target.

The normalized TOF spectrum of ^{243}Am with a pair of C_6D_6 detectors is compared with the reported one [3]

with a 4π Ge spectrometer as shown in Fig. 1. It is found that the time dependent background is suppressed and the detection efficiency is about 100 times smaller than that of 4π Ge. For a MA sample with enough quantity and a high degree of radioactivity, the C_6D_6 detectors will be a useful tool.

In order to extract resonance parameters such as $g\Gamma_n$ and Γ_γ , the deduced neutron capture cross section values of $^{105,108}Pd$ reported in Ref. [1] were analyzed with the SAMMY code. An example of resonance analysis is shown in Fig. 2. Fifty-one resonances of ^{105}Pd and twelve resonances of ^{108}Pd were observed and analyzed.

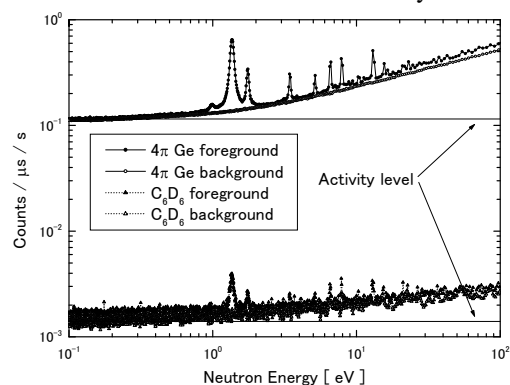


Fig. 1. Comparison of TOF spectra of ^{243}Am with 4π Ge spectrometer and a pair of C_6D_6 detectors.

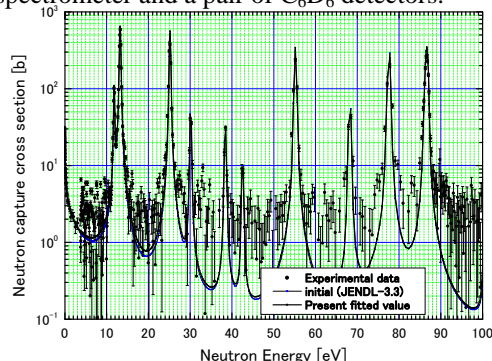


Fig. 2. Resonance analyses with SAMMY in the neutron low energy region of $^{105}Pd(n,\gamma)$ cross section.

Present study includes the result of “Study on nuclear data by using a high intensity pulsed neutron source for advanced nuclear system” entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

REFERENCES:

- [1] J. Hori, *et al.*, KURRI Prog. Rep. 2007 (2008).
- [2] J. Hori, *et al.*, KURRI Prog. Rep. 2008 (2009).
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採択課題番号 21P2-2 各種検出器を用いた MA、LLFP 核種の中性子捕獲断面積の研究 プロジェクト (京大・原子炉) 堀 順一、八島 浩 (原子力機構・基礎工学) 大島真澄、原田秀郎、小泉光生、古高和禎、中村詔司、北谷文人、藤 暢輔、木村 敦、金 政浩、後神 進史 (東工大・原子炉) 井頭政之、水本元治、片瀧竜也 (北大・工学研究科) 木野幸一、鬼柳善明

PR2-3 Cross-Section Measurements for Neutron-Induced Fission of Curium Isotopes

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INTRODUCTION:

In order to evaluate the feasibility of the transmutation or incineration of minor actinides in fast reactors and/or accelerator-driven subcritical systems, neutron-induced fission cross sections are indispensable. We have developed the multi-layered parallel plate ionization chamber (MLPPIC) for this purpose. In the present experiment, cross sections for the neutron-induced fission of curium isotopes, the third abundant minor actinide in the high level radioactive waste after neptunium and americium isotopes, were measured using the lead slowing-down neutron spectrometer at KURRI.

EXPERIMENTS:

The ^{245,248}Cm samples were obtained from Oak Ridge National Laboratory, USA. Each sample was electrodeposited on a stainless steel plate with an active spot of 12.7mm in diameter. The number of atoms in each sample was measured by α -spectrometry and estimated to be $(25.6 \pm 0.1) \times 10^{16}$ for ²⁴⁵Cm and $(91.4 \pm 0.3) \times 10^{16}$ for ²⁴⁸Cm.

Since it is difficult to count the number of incident neutrons on the samples, the reference method was employed. In this method, the curium sample and a reference of ²³⁵U are irradiated simultaneously at the same position. The cross section for the curium is obtained from that for the ²³⁵U reference sample as following equation:

$$\sigma_{245\text{Cm}(n,f)}(E_n) = \frac{Y_{245\text{Cm}(n,f)}(E_n)}{Y_{235\text{U}(n,f)}(E_n)} \frac{N_{\text{U}} \Omega_{\text{U}}}{N_{\text{Cm}} \Omega_{\text{Cm}}} \bar{\sigma}_{235\text{U}(n,f)}(E_n)$$

where Y is the fission yield, N is the number of atoms, Ω is the solid angle for the fission fragments.

RESULTS:

The cross section for ²⁴⁵Cm(n,f) is shown in Fig.1. The solid line is the evaluated value from JENDL-3.3 broadened according to the neutron energy resolution of the spectrometer. The result of Ref [1,2] are also shown for comparison. Our data show a good agreement with Ref [1], whereas the result of Ref [2] shows a slightly lower value in the energy region below 1 eV.

The cross section for ²⁴⁸Cm(n,f) is shown in Fig.2. Closed circles are the result of this work and compared with the other experimental data in Ref [3]. The contribution from the spontaneous fission of ²⁴⁸Cm was sub-

tracted using the data measured without the neutron beam. Although the background of the spontaneous fission was extremely large, the cross section was obtained successfully owing to the high neutron flux of the lead slowing-down spectrometer.

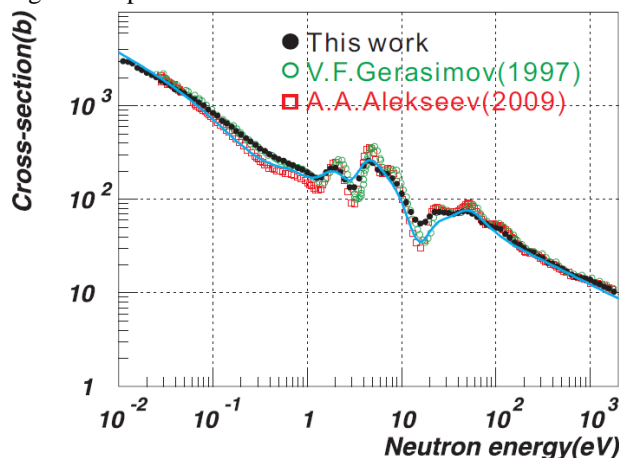


Fig. 1. Cross section for ²⁴⁵Cm(n,f). The result of this work is shown by closed circles and compared with the other experimental data from Ref.[1,2].

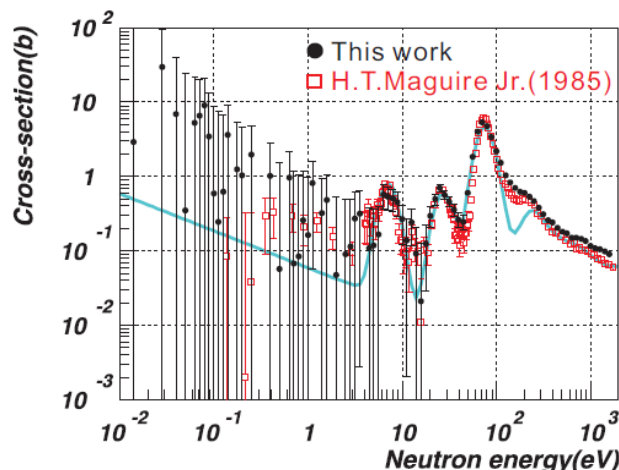


Fig. 2. The cross section for ²⁴⁸Cm(n,f). Previously obtained data from Ref [3] and the JENDL -3.3 evaluation are also shown.

Present study is the result of "Study on nuclear data by using a high intensity pulsed neutron source for advanced nuclear system" entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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- [2] A. A. Alekseev *et al.*, Atomic Energy **106** (2009) 134.
- [3] H. T. Maguire Jr. *et al.*, Nuclear Science and Engineering **85** (1985) 293.

T. Fujii, S. Fukutani, K. Takamiya, H. Yashima and J. Hori

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INTRODUCTION: In order to establish the future advanced nuclear system, the trustable nuclear data of long lived fission products (LLFP) and nuclides of transuranic (TRU) elements are necessary, and for this purpose, preparing highly pure target samples is important. The TRU samples of ^{244}Cm , ^{245}Cm , ^{246}Cm , and ^{248}Cm , and LLFP samples of ^{99}Tc , ^{93}Zr , and ^{107}Pd were prepared. The ^{107}Pd sample consists of stable isotopes, ^{102}Pd , ^{104}Pd , ^{105}Pd , ^{106}Pd , ^{108}Pd , and ^{110}Pd , and the LLFP ^{107}Pd , in which only estimated isotopic compositions are known. In this context, chemical and isotopic purities of the raw materials of ^{107}Pd were analyzed by absorption spectrophotometry and mass spectrometry.

EXPERIMENTAL: Two samples of ^{107}Pd were analyzed. Sample-1 (S1) is a brown flake and sample-2 (S2) is a black powder. Two samples were dissolved in 13.5 M HNO_3 at ~ 383 K. Most of S1 was dissolved, but an undissolved residue was found. S2 was completely dissolved. Supernatant of S1 and solution of S2 were diluted to prepare 1 M HNO_3 solutions and these were analyzed by UV/Vis spectrophotometry (3100PC, Shimadzu).

A portion of 1 M HNO_3 solution including $10\mu\text{g}$ Pd was loaded on a tungsten (W) filament. The portion was dried to be $\text{Pd}(\text{NO}_3)_2$. A rhenium (Re) filament was selected for a counter filament of double filament system. The W-Re set was loaded in a multiple-collector thermal ionization spectrometer (MC-TIMS).

Seven Faraday cups were arranged for ^{102}Pd , ^{104}Pd , ^{105}Pd , ^{106}Pd , ^{107}Pd , ^{108}Pd , and ^{110}Pd . The ionization filament (Re) was heated to obtain 10^{-12} A $^{187}\text{Re}^+$ beam. The evaporation filament (W) was heated to obtain Pd^+ beam ($\sim 10^{-13}$ A Pd^+ beam was obtained). Isotope ratio, $^{107}\text{Pd}/^{106}\text{Pd}$, was measured (20 ratios with 4 second integration time for each scan).

RESULTS: The UV/Vis absorption spectra of 1 M HNO_3 solutions of ^{107}Pd samples were shown in Fig. 1. A specific absorption peak corresponding to Pd(II) was found at 384-392 nm [1]. For both S1 and S2, the same absorption peak was confirmed. No chemical impurities could be found in the absorption spectra.

Isotopic compositions determined in this study were shown in Table 1. As a reference, a standard solution for absorption spectrometry (Wako Pure Chemicals) was analyzed. The analytical results showed that the isotopic composition of typical standard solution possesses that of naturally occurring isotopes of Pd.

It is clear that the isotopic compositions of S1 and

S2 are totally different from that of naturally occurring isotopes of Pd. This suggests that the raw materials of Pd were extracted from neutron irradiated materials like a spent nuclear fuel. Isotopic composition of S1 seems to be the same with that of S2. Though the chemical forms of S1 and S2 are different, the starting materials might be the same. Our analysis resulted in that both the samples S1 and S2 contain 15.3% of the LLFP ^{107}Pd .

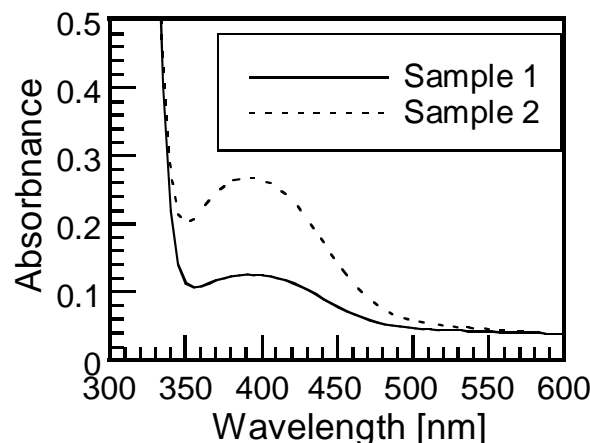


Fig. 1. UV/Vis absorption spectra of 1 M HNO_3 solutions of ^{107}Pd samples.

Table 1. Isotopic compositions (%) of Pd samples.

	^{102}Pd	^{104}Pd	^{105}Pd	^{106}Pd	^{107}Pd	^{108}Pd	^{110}Pd
Nat.	1.0	11.1	22.3	27.3	0.0	26.5	11.7
Std.	0.9	11.1	22.4	27.2	0.0	26.6	11.8
S1	0.0	1.6	48.8	23.1	15.3	8.6	2.6
S2	0.0	1.9	48.2	22.8	15.3	8.9	3.0

Nat.: Naturally occurring isotopes

Std.: A standard solution for absorption spectrometry (Wako Pure Chemicals)

S1 and S2: Samples 1 and 2.

REFERENCES:

[1] T. Fujii *et al.*, J. Radioanal. Nucl. Chem., **247(2)** (2001) 435-437.

A part of present study is the result of "Study on nuclear data by using a high intensity pulsed neutron source for advanced nuclear system" entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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INTRODUCTION: The activity density evaluation is important to dispose radiological waste containing transuranium nuclei and fission products. Indirect methods as the scaling factor method have been studied for quantitative evaluation of “difficult to measure” nucleus which emits only β particles. The methods need the destructive analysis of many samples. The measurements of gamma rays generated by photonuclear reactions for the “difficult to measure” nuclei has been proposed to examine activity densities by the “nondestructive analysis”. The photon-induced cross section data for some nuclei were measured at SACLAY [1], Lawrence Livermore [2] and evaluated by IAEA [3]. ^{99}Tc that is ones of typical “difficult to measure” nuclei.

EXPERIMENTS: The experiment was performed at the electron linear accelerator facility in Kyoto University Research Reactor Institute. Figure 1 shows the experimental arrangement. A ^{99}Tc was adopted as a “difficult to measure” nucleus. Bare gold foils were also set in front and back of each sample to obtain the neutron flux in front and back of the iodine sample. The sample was irradiated with continuous energy bremsstrahlung X ray beam for 60 minutes. The photon beam was generated by bombarding a thin Pt target with 36 MeV electrons. The gamma rays from each sample were measured for 5 minutes by a Ge detector after the irradiation and cooling.

RESULTS: The photon flux and energy distribution is necessary to obtain the amplitude of the (γ, n) reaction cross section. The photon flux and energy distribution at the sample position were calculated with the EGS 5 code [4] using the electron energy distributions and normalized by the activity of ^{196}Au produced by the $^{197}\text{Au}(\gamma, n)^{196}\text{Au}$ reaction illustrated in Fig. 2. The photons above the threshold energy of the $^{99}\text{Tc}(\gamma, 3n)^{96}\text{Tc}$ of 26 MeV were assumed to induce the reaction in the data analysis.

The yields of photonuclear the reaction product was obtained by counting the gamma-rays from the product. The average photonuclear reaction cross section between 26 and 36 MeV is 2.30 ± 0.17 mb. The photonuclear $(\gamma, 3n)$ cross sections gently change with the number of proton and neutron of the nucleus [3]. Considering the trends of the cross section for vicinal nuclide and the cross section of $^{98}\text{Mo}(\gamma, 3n)^{95}\text{Mo}$, a predicted shape of the cross section for $^{99}\text{Tc}(\gamma, 3n)^{96}\text{Tc}$ reaction is reproduced shown in Fig. 3.

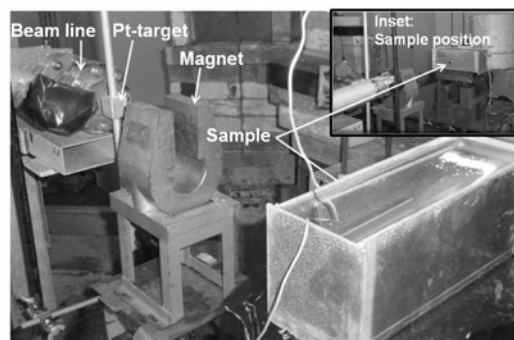


Fig. 1. Experimental setup.

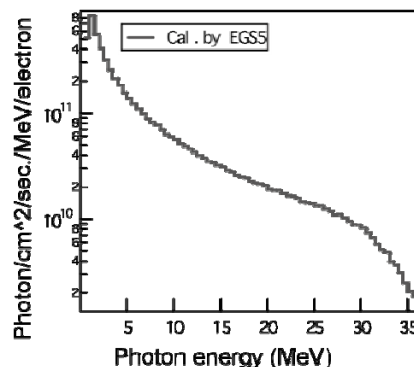


Fig. 2. Electron beam intensity.

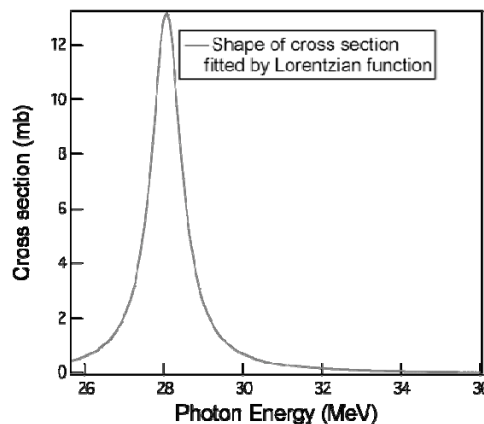


Fig. 3. Prediction of the shape of $^{99}\text{Tc}(\gamma, 3n)^{96}\text{Tc}$ reaction cross section.

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PR2-6 Study on Neutron Energy Spectrum Measurements for Pulsed Neutron Source

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INTRODUCTION: The pulsed neutron source using the electron linear accelerator at Research Reactor Institute of Kyoto University is widely used for nuclear science researches including nuclear data (neutron reaction cross sections) measurements [1]. It is important to understand the characteristics of the pulsed source, such as neutron spectra, for the proper use of the source. There are three conventional techniques to measure the neutron spectra, (1) Multi-foil activation method, (2) Time-of-Flight (TOF) method, and (3) Neutron counter measurement. Between these techniques, only the TOF method can obtain the differential data directly. However, the range of measurement is limited to about 1keV due to the distortion of signal by the gamma burst (gamma-flash) generated when the pulsed neutron produced [2].

In the present study, the method to expand the energy range of measurements using the digital data processing technique has been investigated.

MEASUREMENT METHOD: The TOF method measures the time of neutron flight from the pulsed neutron source to a neutron detector located at a fixed distance from the source. Since the flight time is proportional to the inverse of neutron speed, the energy of detected neutrons can be obtained easily. At the LINAC, the neutron spectrum leaked from the water moderator has been often measured. Fig.1 show a layout of TOF measurement. In this case, the B-10 sample was located at the end of flight tube (12.7m-length), and the gamma-ray of neutron capture reactions in the sample was detected by a multiple-BGO detector.

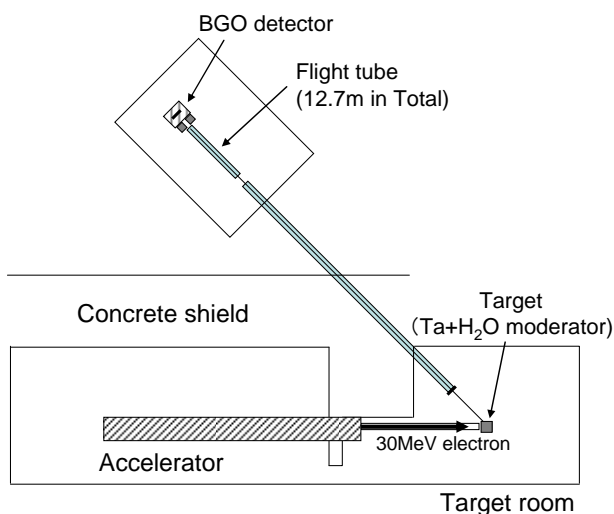


Fig. 1. Layout of Time-of Flight experiment at KURRI-LINAC.

Using this method, the differential data can be obtained directly. However, the range of measurement is limited to about 1keV. This is due to the distortion of signal by the gamma-flash. To avoid this effect, the digital data measurement and processing were performed. In this method, the output signal of the BGO detector was digitally recorded and the shape distortion due to the gamma-flash was subtracted for each measured data using the "typical distorted signal", which was previously obtained between the background measurement, then the effect of distortion almost vanished.

An example of the digital data processing is shown in Fig. 2. The solid line in the figure shows the measured signal of the detector, it is found that the large distortion caused by the gamma-flash continues to about 150-200ch (15-20ms). This distortion was subtracted using the typical distorted signal (the dotted line), and hence the corrected signal (the dashed line) was obtained. Then, the gamma-ray signal of neutron capture reaction at about 70 ch., which cannot be counted in the original distorted signal, can be counted after the correction. After all, the energy range of the TOF method expanded to several tens keV using the digital data processing technique. For further study, the accuracy of the method, in particular, the representivity of the typical distorted signal should be investigated.

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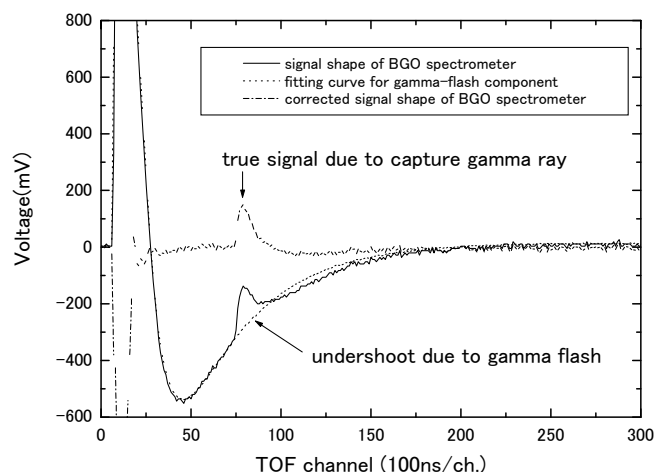


Fig. 2. An example of the digital data processing to avoid the effect of gamma burst.