CO2-1 Prompt γ -ray Measurement of ³⁵Cl(n, γ) Using Total Absorption Type Ge Detector

H. Hayashi, Y. Kojima¹, M. Shibata¹ and A. Taniguchi²

Institute of Health Biosciences, The University of Tokushima Graduate School ¹Radioisotope Research Center, Nagoya University ²Research Reactor Institute, Kyoto University

INTRODUCTION: Prompt γ -ray analysis (PGAA) is one of the efficient microanalytical techniques. The prompt γ -ray spectrum is composed of many γ -rays up to 10 MeV. If these γ -rays were totally absorbed by the detector, the corresponding event had an energy of S_n (neutron separation energy). When the certain samples having unknown isotopes were measured with a total absorption detector, use of the S_n peaks was appropriate to the analysis. In order to develop a measurement method by means of a total absorption type Ge detector [1], we used neutron beams of KUR. Our Ge detector has a through-hole. The samples are put into the center position of the detector, and neutrons are introduced into the hole. The capture γ -rays emitted from the sample are absorbed by the detector with the large solid angle. In the previous study, a neutron guide was developed [2]. In the present paper, in order to examine the possibility to apply the method to PGAA, the total absorption spectra of typical nuclei were measured and compared with the general ones.

EXPERIMENTS: Samples of ²⁷Al(natural, 0.04g), ³⁵Cl(CH₂-CCl₂, 0.008g) and ¹⁹⁷Au(natural, 0.02g) were irradiated by thermal neutrons at the B-4 line. Neutron flux was 7×10^5 n/cm/s at the 1MW operation. Measurement times were 1-10 hours. The inset of Fig.1 shows a schematic drawing (side view) of the experiment. In order to compare the spectrum measured at the center position with the other one, we measured spectra based on two sample positions; one is a center position (X=0 cm), and the other is a surface position of the detector (X=6.5 cm).

RESULTS AND DISCUSSION: Fig. 1 shows the measured spectra of ³⁵Cl(n, γ). Differences between two spectra are clearly observed in the energy region around S_n =8.6 MeV. This fact indicates that the probability of totally absorbed events at X=0 cm is larger than that at X=6.5 cm. In order to analyze quantitatively, peak areas of the observed intense peaks were compared between the two spectra. Fig. 2 shows dependence of peak counts divided by an intensity of γ -ray (I $_{\gamma}$) [3] as a function of γ -ray energies. When the peaks are formed by a single γ -ray, the values of counts/I $_{\gamma}$ are proportional to the efficiency of the detector. Most of the data presented in the Fig. 2 shows a similar shape with the efficiency of the detector except for 2468 keV and 8578 keV at X=0 cm.

The 8578 keV is S_n of ³⁶Cl, and this peak is formed by not only a single γ -ray but also sum of a number of γ -rays. Therefore, the 8578 keV data at the X=0 cm shows approximately ten times larger than the other peaks. On the other hand, the 8578 keV data at the X=6.5 cm did not show markedly-elevated value. The peak of S_n is enhanced by a factor of ten, therefore we considered that the peaks corresponding to the S_n values are available to analyze a certain sample which consist of the unknown isotopes. It has possibility that our measurement method can efficiently identify the nuclei of interest compared to the other measurement method. In order to deduce the detection efficiency of S_n values for various nuclei, we plan to measure various nuclei.

CONCLUSION: Using the total absorption type Ge detector, we measured γ -ray spectrum of ${}^{35}Cl(n,\gamma)$, ${}^{27}Al(n,\gamma)$ and ${}^{197}Au(n,\gamma)$. The peaks of S_n were remarkably enhanced. This measurement method is effective to detect the nuclei of interest by analyze the peak of S_n .



Fig.1 Measured spectrum of ${}^{35}\text{Cl}(n,\gamma)$. The inset shows a cross sectional view of the detector. The solid and dashed lines show spectra measured with X=0 cm and X=6.5 cm, respectively.



Fig. 2 Relationship between counts/I_{γ} and γ -ray energy. The peaks of 2468 keV and 8578 keV are formed by sum of the number of γ -rays.

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Development of New Calibration Method for a Neutron Detector using a Pulsed White **CO2-2** Neutron Source from the KURRI-LINAC

T. Matsumoto, H. Harano, A. Masuda, C. Shoda¹,

Y. Fukumoto¹, J. Kawarabayashi¹, H. Tomita¹, T. Iguchi¹, A. Uritani¹, M. Takada², S. Kamada², J. Hori³ and

Y. Sakurai³

National Metrology Institute of Japan, National Institute of Advanced Industrial Science and Technology ¹School of Engineering, Nagoya University ²National Institute of Radiological Sciences ³Research Reactor Institute, Kyoto University

INTRODUCTION: Response functions are one of the key parameters of neutron detectors, which should be determined generally by calibration using monoenergetic neutrons. However, it is difficult to obtain monoenergetic neutrons in the epi-thermal energy region. In the present study, we have developed a new calibration method for neutron detectors by means of the time-of-flight method using a pulsed white neutron source with energy range from thermal to 10 keV at KURRI. In this calibration method, a precise detection efficiency curve of the neutron detector is experimentally determined by two-dimensional simultaneous measurements of the pulse height of detector outputs with the time of flight (TOF). We also developed an epithermal neutron camera consisting of GEMs and resonance filters for neutrons up to 10 keV.

EXPERIMENTS: Neutrons were obtained by the photoneutron reaction using a water-cooled tantalum target at the KURRI Linac [1]. The neutron spectral fluence was determined with BGO, ⁶Li-glass and liquid scintillators in previous measurements. Calibration tests were performed for a neutron survey meter "PRESCILA" composed of plastic scintillators and a ZnS(Ag) scintillator and a personal electric dosimeter composed of Si detectors. The response function of a prototype of epithermal neutron imaging camera was also measured using the white neutrons. The prototype detector consists of a silver plate as a resonance filter, a B₄C thermal neutron absorber, and a GEM with a neutron converter of ${}^{10}B$. The difference in observed counts from each readout electrode of GEMs between the measurements with and without the resonance filter gives the epithermal neutron image.

RESULTS: Figure1 shows the TOF spectrum measured with the PRESCILA. As shown in Fig.2, the energy response was successfully obtained from the TOF spectrum and the neutron spectral fluence. The energy response was obtained also for a personal dosimeter. Fig.3 show the epithermal neutron image obtained with the prototype of epithermal neutron imaging camera. The measured detection efficiency for the epithermal neutron in the energy range from 4.2 eV to 6.3 eV, corresponding to the resonance peak of 109 Ag, was $5x10^{-4}$ counts/neutron.







Fig.2 Efficiency curve of the PRESCILA obtained from the calibration method with the white neutrons



Fig.3 Epithermal neutron image obtained by the epithermal neutron imaging camera with GEMs

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採択課題番号 23044 熱中性子フルエンス率の測定の高度化と 共同通常 その国際標準化に関する研究 (産総研)原野英樹、松本哲郎、増田明彦(名大)井口哲夫、瓜谷章、河原林順、渡辺賢一、富田 英生、照田千尋、福本靖、中山元(放医研)高田真志、鎌田創(京大・原子炉)堀順一、櫻井良憲

CO2-3 Neutron Capture Cross section Measurement on ²⁴³Am with a pair of C₆D₆ detectors

J. Hori, H. Yashima and S. Nakamura¹

Research Reactor Institute, Kyoto University ¹Japan Atomic Energy Agency (JAEA)

INTRODUCTION: Accurate neutron capture cross section data of minor actinides (MAs) are required for transmutation study and design of innovative reactor system. Americium-243 is one of the most important MAs since it has a long half-life (7,370 yr) and the neutron capture reaction of ²⁴³Am produces higher-mass Cm isotopes. However, the present status of experimental data for ²⁴³Am is very poor both in guality and guantity. This is because its highly energetic gamma radiation from ²³⁹Np (a daughter nuclide of ²⁴³Am) causes severe problems in capture cross-section measurements. Therefore, there are only three available neutron-capture measurements with the time-of-flight (TOF) method. Wisshak and Käppeler measured the neutron capture cross section in the range from 5 to 250 keV using a Moxon-Rae detector and the ${}^{7}Li(p,n){}^{7}Be$ neutron source [1]. Weston and Todd measured the neutron capture cross section in the range from 0.26 to 92 keV using a C_6F_6 detector and the photo-neutron source [2]. Recently, we have measured the neutron capture cross section of ²⁴³Am with a 4π Ge spectrometer in the energy range from 0.01 to 400 eV at the KURRI-LINAC [3]. In this work, we have employed a C_6D_6 liquid scintillator which has a fast time response as a prompt gamma-ray detector in order to obtain the neutron capture cross section in the higher energy region.

EXPERIMENTS: The capture cross section measurement was carried out by the TOF method using the KURRI-LINAC. A photo-neutron target of Ta was adopted as a pulsed neutron source for the neutron TOF measurement. We employed a pair of C_6D_6 liquid scintillators for the capture gamma ray measurement. The decay gamma rays from ²⁴³Am sample were eliminated by covering the 6-mm-thick lead shield with the front end of each detector. The distance between the sample and the neutron source was 12.0±0.02 m. Output signals from the scintillators were summed up and stored with the Yoko-gawa's WE7562 multi channel analyzer as a two dimensional data of pulse height (PH) and TOF.

The americium sample, which was purchased from Russia Research Institute of Atomic Reactors, was AmO_2 powder of 0.128 g packed in an aluminum disk container of 30 mm in outer-diameter. The amount of ²⁴³Am con-

tained in the sample was determined as 868±15 MBq by measuring the decay gamma rays.

An aluminum disk container without the americium oxide was used as a dummy sample for the background determination. An enriched ¹⁰B sample was also used to measure the incident neutron flux. The sample of ¹⁰B was packed in an aluminum container. The energy dependent neutron flux was derived with the standard cross sections of the ¹⁰B(n, $\alpha\gamma$) reaction. The correction for the neutron self-shielding and multiple scattering in the sample was made by the MCNP-4C [4] with the nuclear data taken from JENDL-4.0 [5].

RESULTS: In this experiment, the shape of the ²⁴³Am neutron capture cross section was measured as a function of neutron energy. The relative yields were normalized at the 1.356-eV resonance area to the evaluated value in JENDL-4.0. Making use of a pair of C_6D_6 liquid scintillators, the neutron capture cross section of ²⁴³Am has been measured up to 40 keV as shown in Fig. 1. Further study such as application of pulse-height weighting technique will be made to determine absolute cross section value.



Fig. 1 Neutron capture cross sections of ²⁴³Am

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採択課題番号 23048 高速中性子領域における中性子捕獲断面積測定に関する研究 共同通常 (京大・原子炉)堀 順一、八島 浩(原子力機構)中村詔司

CO2-4 Reaction Rate Distributions in the Thorium-Loaded Accelerator-Driven System with 14 MeV Neutrons at the Kyoto University Critical Assembly

T. Iwasaki, N. Aizawa, H. Kubo, C.H. $\mathsf{Pyeon}^1,$ T. Yagi^1 and T. Misawa^1

Graduate School of Engineering, Tohoku University ¹Research Reactor Institute, Kyoto University

Introduction: The experimental study on the accelerator-driven system (ADS) in the Kyoto University Critical Assembly (KUCA) was focused on the development of a new neutron source. Among a series of the ADS experiments in KUCA, the experimental results of reaction rates of the thorium (Th) capture reactions were presented: the thorium-loaded ADS experiments were carried out by varying the neutron spectrum, and were aimed at measuring the Th capture reactions in the subcritical system. In the experiments, for the investigation of the effect of neutron spectrum, the moderator material was varied in fuel region. The objectives of this study were to investigate the neutronic characteristics of the thorium-loaded ADS with 14 MeV neutrons through the experiments on the reaction rates of Th capture reactions, and to evaluate the precision of the calculations by the MCNPX code with ENDF/B-VII.0 and JENDL-3.3 libraries.

Results and Discussion: The absolute values of measured reaction rates $(^{115}In(n, \gamma)^{116m}In \text{ reactions})$ revealed differently the variation of reaction rates at the position of the tritium target shown Fig. 1. The highest peak of reaction rates was found in the core region in cases of the natural-uranium-polyethylene core (NU-PE core) shown Fig. 2. The reaction rates in the thorium-graphite (Th-Gr) core were observed flattening in the entire core region due to the hard neutron spectrum caused by the graphite, comparing with others. Moreover the moderation effect of the high-energy neutrons in some cores (Th-PE, Th-Gr and NU-PE cores) was found around the boundary between the core and polyethylene regions. From these results, the effect of neutron spectrum on the Th capture reactions was observed remarkably in the thorium-loaded ADS experiments with 14 MeV neutrons. The numerical calculations were executed with the use of the MCNPX code coupling with ENDF/B-VII.0 and JENDL-3.3 shown in Fig. 3. The calculated reaction rates of the Th capture reactions reconstructed approximately the experimental ones, and the difference between ENDF/B-VII and JENDL-3.3 libraries was not found in the reaction rate distributions. Also, another comparison between the experiments and the calculations demonstrated good agreement in the thorium-loaded ADS including the Th-Be, experiments, Th-Th-Th-PE (Th*3-PE; Fig. 2) and NU-PE cores.



Fig. 1 Top view of Th-PE core configuration







Fig. 3 Comparison between the results in experiments and calculations of the In reaction rate distributions

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CO2-5 Measurements of Thorium-Fueled Core Characteristics and Replacement Worth of Thorium Plates to Aluminum Plates (2)

N. Takaki, A. Yamaguchi, H. Watanabe, T. Kitada¹, T. Fujii¹, K. Wada¹, Vu Thanh Mai¹ and H. Unesaki²

Department of Nuclear Engineering, Tokai University ¹Graduate School of Engineering, Osaka University, ²Kyoto University Research Reactor Institute

INTRODUCTION: Utilization of Thorium fuel in conventional reactor is known as one alternative approach to diversify fission resource and improve the core performances. However the accuracy of the cross section is quietly lower than that of Uranium because there were less experiments with Thorium and following validation studies are scarce. This low accuracy brings large uncertainty in calculation results of Thorium-fueled core performance such as k-eff, coefficients, conversion ratio etc..

The object of this experiment is to validate the Thorium cross section through the comparison of calculation and measured results for Thorium-fueled core characteristics and replacement worth of Thorium plates to Aluminum plates.

EXPERIMENTS: The assembled core is the first core to be critical with unit fuel cell named "10/8PETEETEE" which is shown in Fig. 1.



Fig.1. Unit Fuel Cell(10/8"PETEETEE).

In the experiment, critical approach was performed. The number of fuel assembly and control rod arrangements at critical were analyzed before the experiment so as to match the constraints (maximum excess reactivity and control rod worth), and each steps from initial core to critical core was also analyzed with the comparison of Uranium-fueled core and the Thorium-fueled core performed with other unit fuel cells. The core arrangements at each step are illustrated in Fig. 2.



Initial core were assembled by 29 fuel assemblies marked as "0" and the number marked in fuel assemblies in Fig.2 shows the fuel assemblies added at each step of critical approach. The number of fuel assemblies was slightly increased then the critical core were realized at last step:8 with 47 fuel assemblies. The inverse count rate ratio curve obtained through the critical approach is shown in Fig. 3 for the case of all control rods withdrawn (j=3).



Fig.3. Inverse count rate ratio curve obtained for B10/8"P17ETEETEE core.

The excess reactivity was measured by the period method and the value is about 0.08%dk/k, and the control rod worth for C1, C2 and C3 measured by rod drop method are 0.272, 0.365 and 0.315%dk/k, respectively. These values satisfy their constraints to operate the core.

The core shown in Fig.2 was slightly changed to low excess reactivity state so as to measure the replacement worth. Thorium plates were replaced to Aluminum plates to measure the replacement worth by the change in excess reactivity. The position of the replacement was the center fuel assembly named $\frac{1}{\sqrt{3}}$ 15 and the number of the replacement was two or four at around the center in core height. This measurement was done to check the C3 rod position at critical with different replacement situation, because the calibration curve of C3 rod obtained by fitting many experimental results (period method) was confirmed to have enough accuracy to check the excess reactivity. The results are summarized in Table 1 and the results are roughly good agreement in the analysis with continuous energy Monte-Carlo analysis with JENDL4.0.

Table 1. Results of excess reactivity at each replacement situation.

Core configuration	Excess reactivity []k/k]	Error
partial core	4.92E-04	6.55E-06
par+Al2 core	1.78E-03	3.65E-06
par+Al4 core	3.02E-03	1.05E-05

RESULTS: Thorium-fueled core with $H/^{235}U$ ratio=210 was assembled for the first time and criticality approach and related measurements were successfully performed. The detailed analysis of the results is in progress.

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CO2-6 Development of Subcriticality Measurement for Accelerator-Driven Reactor (VI)

K. Hashimoto, W. Sugiyama, A. Sakon¹, M. A. Binmaarof¹, T. Hasegawa¹, S. Tanaka¹, C. H. Pyeon², T. Misawa² and T. Sano²

Atomic Energy Research Institute, Kinki University ¹Interdisciplinary Graduate School of Science and Technology, Kinki University ²Research Reactor Institute, Kyoto University

INTRODUCTION: An accelerator-driven subcritical reactor system has been constructed in A-loading facility of the KUCA and a series of cross-power spectral analyses between time-sequence signal data from a neutron detector and a beam current meter have been performed to develop the methodology of on-line subcriticality monitoring. The preliminary results of the cross-power spectrum analyses are showed in this report

EXPERIMENTS: These measurements were performed in a reactor system referred to as A3/8"P36EU(3). A tritium target was placed outside polyethylene reflector and pulsed neutron beam was emitted from the target. As pulse repetition frequency, 20, 100 and 500Hz were employed. Not only cross-power spectral density between time-sequence signal data from two BF₃ counters placed in the core, but also the spectral density between the signal data from the neutron counter and a current meter for accelerator beam were analyzed to determine the prompt-neutron decay constant. The experiments were carried out in three subcritical states. The subcriticality of the state was adjusted by changing control rod pattern. These control rod patterns are showed in Table 1.

Table 1 Control rod patterns employed

Pattern	Ro	d Positior	ı	Reactivity*
	C1	C2, C3	S4~S6	[%Δk/k]
Α	L.L.	U.L.	U.L.	-0.279
В	L.L.	L.L.	U.L.	-0.668
С	L.L.	L.L.	L.L.	-1.573

L.L.:Lower Limit, U.L.: Upper Limit *Evaluated from reactivity worth of control rod

RESULTS: Figure 1 shows the magnitude of a cross-power spectral density measured in a subcritical system (rod pattern A) driven by pulsed neutron source, whose repetition frequency is 20Hz. The spectral density consists of a correlated reactor-noise component and many uncorrelated delta-function-like peaks at integral multiple of the repetition frequency (20Hz). We formulated the cross-power spectral density between accelerator-beam current and neutron detection signals to obtain the following formulae for magnitude (gain) and phase of the imaginary spectral density.

$$\left|\Phi(\omega)\right| = \frac{A}{\sqrt{\alpha_0^2 + \omega^2}},\tag{1}$$

$$\angle \Phi(\omega) = -\tan^{-1} \left(\omega / \alpha_0 \right) \tag{2}$$

In this figure, also, a least-squares fit of the above equation (1) to the point data of delta-function-like peaks is shown, where the other data, i.e., the data of correlated reactor noise component are masked. The decay constant 211.3 ± 12.6 (1/s) inferred from the fitting is consistent with 223.6 ± 5.2 (1/s). obtained by a pulse neutron experiment. The magnitude of the correlated component is too minute to infer the decay constant.

Figure 2 shows the prompt-neutron decay constant inferred from phase data near the integral multiple of pulse repetition frequency on the basis of Eq. (2). The decay constant decreases with an increase in frequency. In the higher frequency range, the phase analysis underestimates the decay constant. The underestimation originates from unnegligible contribution of spatial higher mode. However, no least-squares fitting is necessary for inference of the decay constant and the analysis of phase data is advantageous than that of magnitude. We should improve the underestimation in near future.







Fig.2 Evaluation of prompt-neutron decay constant from phase shift data near integral multiple of pulse repetition frequency

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(近大・原研)橋本憲吾、杉山 亘(近大院・理工)左近 敦士、ムハマド・アイマン・ビンマーロ フ、長谷川 喬、田中慎吾(京大・原子炉) 卞 哲浩、三澤 毅、佐野忠史