

## CO2-1 Measurement for thermal neutron capture cross sections and resonance integrals of the $^{243}\text{Am}(n,\gamma)^{244g}\text{Am}$ , $^{244m+g}\text{Am}$ reactions

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**Introduction:** Americium-243 has the half-life of 7367 yr [1] and contributes to the production of  $^{244}\text{Cm}$  (18.11 yr [1]) through neutron capture reaction. The neutron capture cross-section data of  $^{243}\text{Am}$  are important in considering the amount of  $^{244}\text{Cm}$  accumulated in spent nuclear fuels. For the prediction of  $^{244}\text{Cm}$  production, a difference of 10% between calculated and experimental values was reported.[2] Thus the neutron capture cross sections of  $^{243}\text{Am}$  were measured by a neutron activation method.

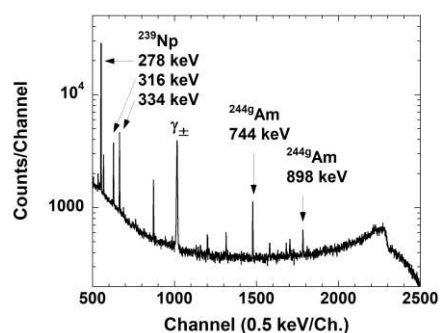
**Experiments:** Nitric acid solutions of  $^{243}\text{Am}$  equivalent to 1 kBq were dropped onto fiber filters and dried. To monitor neutron fluxes, a set of Au/Al and Co/Al alloy wires was attached to the Am sample as an irradiation target. Neutron irradiation was performed using irradiation systems of the KUR. Gamma- and alpha-ray spectrometry were applied to measurements of irradiated samples.

First, for the  $^{243}\text{Am}(n,\gamma)^{244g}\text{Am}$  reaction, measurements of emission probabilities of  $\gamma$  rays emitted from the ground state  $^{244g}\text{Am}$  (10.1 h[1]) had been performed under the project 25P1-16 in 2014. The previous 29% uncertainty [3] in the emission probability had been improved to 2% uncertainty [4]. The  $^{243}\text{Am}$  targets were irradiated in 1 MW power operation with the pneumatic tube Pn-2. The bare target was irradiated for 30 min, and that with a Gd filter for 5 min. Gamma rays emitted from irradiated Am samples and alloy wires were measured with a high-purity Ge detector.

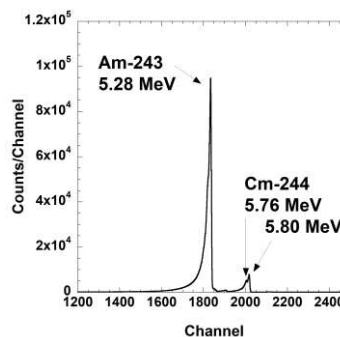
Next, for the  $^{243}\text{Am}(n,\gamma)^{244m+g}\text{Am}$  reaction, the amounts of  $^{244m}\text{Am}$  and  $^{244g}\text{Am}$  produced via neutron capture reaction were determined from the  $\alpha$ -ray yield of their daughter nucleus  $^{244}\text{Cm}$ . The  $^{243}\text{Am}$  targets with and without a Gd shield were irradiated for 6 hours by the hydraulic conveyer in 5 MW power operation. After irradiation, a cooling time for 5 days caused  $^{244g}\text{Am}$  sufficiently to decay to  $^{244}\text{Cm}$ . At this time, the amount of  $^{244g}\text{Am}$  was reduced to about 1/4000. Irradiated fiber filters were rinsed with 3M  $\text{HNO}_3$  solution and the solution was centrifuged. The separated supernatants were dropped onto Petri dishes, and dried to make  $\alpha$ -ray samples. Alpha-ray measurements were done with the model 7401 alpha spectrometer (Canberra, Inc.).

**Analysis:** Neutron fluxes at the irradiation positions were obtained from the induced activities of the monitor wires.

**Figure 1** shows an example of  $\gamma$ -ray spectrum due to the decay  $\gamma$ -rays emitted from  $^{244g}\text{Am}$  produced by the  $^{243}\text{Am}(n,\gamma)^{244g}\text{Am}$  reaction. Its measurement time was 16 hours. For the  $^{243}\text{Am}(n,\gamma)^{244m+g}\text{Am}$  reaction, **Figure 2** shows an  $\alpha$ -ray spectrum obtained by 168-hours measurement. Reaction rates of the  $^{243}\text{Am}$  were obtained from amounts of the  $^{243}\text{Am}$  samples and produced nuclides. Their reaction rates and neutron flux information were analyzed on the basis of Westcott's convention [5] to derive neutron capture cross sections. Data analysis is in progress.



**Fig.1** An example of  $\gamma$ -ray spectrum of irradiated  $^{243}\text{Am}$  sample.



**Fig.2** An example of  $\alpha$ -ray spectrum of irradiated  $^{243}\text{Am}$  sample.

### Acknowledgement

Present study includes the result of “Research and Development for accuracy improvement of neutron nuclear data on minor actinides” entrusted to the Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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## CO2-2 Quantitation of Gamma Ray Emission from Capture Reaction of Uranium-238

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**INTRODUCTION:** For the criticality control and the safeguards of fuel debris formed in the Fukushima Dai-Ichi nuclear power station, quantification of the mass of nuclear materials and its residual fissile enrichment is desirable. However, non-destructive quantification techniques have never been developed even for intact spent nuclear fuel assemblies. In the conventional fuels for light water reactor utilization, the dominant nuclide of the fuel mass is <sup>238</sup>U. Thus, we have focused on measurement of  $\gamma$  rays from capture reactions of <sup>238</sup>U by neutron induced  $\gamma$  ray spectroscopy (NIGS). We have succeeded in detection of 4060 keV  $\gamma$  rays from capture reaction of <sup>238</sup>U from a sub-critical core [1]. Furthermore, we found that the count rate ratios of the 4060 keV  $\gamma$  ray to fission prompt ones are consistent with <sup>238</sup>U/<sup>235</sup>U ratios in two sub-critical cores of different average <sup>235</sup>U enrichments [2]. However, the  $\gamma$  ray spectrum from the reaction has been reported to vary with the incident neutron energy [3]. To utilize the NIGS spectrum for the quantification of the nuclear material and the enrichment, the  $\gamma$  ray spectrum measurement was conducted in this work.

**EXPERIMENTS:** The measurement was conducted in LINAC pulsed neutron source facility in KURNS. A tungsten (W) target was irradiated by accelerated electron and the Bremsstrahlung photons were radiated. Then the photon interacted with W and photo nuclear reactions were induced. The beam frequency was adjusted to 50 Hz and the pulse width was 1  $\mu$ s (nominally). The beam current was stabilized to 27  $\mu$ A. The target was located on center of a cylindrical tank of 20 cm in diameter and 30 cm in height. The tank was filled with light water. The neutrons generated in the W target was moderated in the water. We used the neutron at “12 m room” which is located on 135 deg axis from the electron beam line. In the room, a square shaped sample of metallic U was irradiated by the moderated neutron. Its geometry was 4 cm  $\times$  4 cm and the thickness corresponds to 2.91 g/cm<sup>2</sup>. A HP-Ge detector of 35% relative efficiency was set on 90 deg axis from the neutron flying axis. The distance from the center of the sample to the head of the detector was 5 cm. The detector was carefully shielded from the  $\gamma$  - flash from the target. The  $\gamma$  ray pulse height in the detector was acquired together with the time of flight (TOF) information. The pulse height was calibrated with additional measurement of <sup>35</sup>Cl(n, $\gamma$ ) reactions. The energy of the neutron inducing the <sup>238</sup>U(n, $\gamma$ ) reaction was deduced from the TOF information. TOF spectra was calibrated with additional transmission measurements through Mn, In,

Ag, Cd. The effective length of the flight path was deduced to be 11.3m.

**RESULTS:** In figure 1, the  $\gamma$  ray pulse height spectra are given for several neutron energy regions: the thermal, the epi-thermal, and the resonances of <sup>238</sup>U(n, $\gamma$ ) reactions. In the thermal and epi-thermal regions, <sup>238</sup>U(n, $\gamma$ )  $\gamma$  rays were measured simultaneously with prompt and delayed ones from fission reactions of <sup>235</sup>U. For the resonance energies, it is considered that the <sup>238</sup>U(n, $\gamma$ )  $\gamma$  rays are dominant. In every region, 4060 keV  $\gamma$  ray was detected. The energy resolution of this experiment was better so that 3982 and 3991 keV components are resolved, which has never been done with 4 $\pi$  type HP-Ge detector [3]. 3583 and 3197 keV  $\gamma$  rays were measured especially for the En=6.67 eV resonance. However, 3567, 3541 and 3482 keV  $\gamma$  rays [3] were not clearly resolved due to the counting statistics and the single escape peaks. In CapGam [4], yield of 3296.5 keV  $\gamma$  ray is evaluated larger than that of 4060 keV  $\gamma$  ray for the thermal neutron capture of <sup>238</sup>U. However, the 3296.5 keV  $\gamma$  ray peak was not found for reactions for every neutron energy region.

The  $\gamma$  ray spectra were observed differently for incident neutron energies. On account of the difference, ratio of some resonances to the thermal <sup>238</sup>U(n, $\gamma$ ) reaction might be unfolded by spectra obtained by NIGS. Thus, NIGS would be a new validation tool for neutron spectrum calculation in the reactor physics field, which has never been achieved. For the purpose, <sup>238</sup>U(n, $\gamma$ )  $\gamma$  ray measurements with better statistical accuracy is further desired as well as nuclear data evaluation.

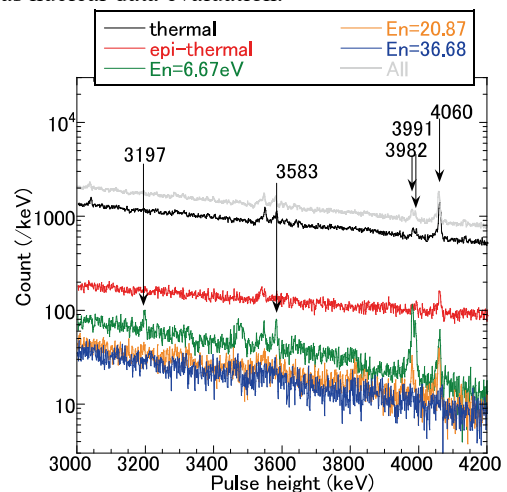


Fig. 1 Measured pulse height spectrum for U sample.

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**INTRODUCTION:** Evaluation of neutron fluence and neutron dose equivalent for the epi-thermal neutron region is very important to manage workplaces with neutron sources or nuclear fuels as well as irradiation fields in a boron neutron capture therapy (BNCT). In the pre-sent study, we have developed a current-mode neutron detector [1] that can be used in intense neutron flux field such as BNCT fields. The neutron detector is composed of a pair of <sup>6</sup>Li-glass and <sup>7</sup>Li-glass scintillators (10-mm diameter and 2-mm thickness). The current outputs from photomultipliers are directly used to evaluate the neutron fluence in the detector. We have tried to apply the current-mode detectors to sensors used in a neutron detection device with a moderator. We fabricated four pairs of the current-mode detector (four <sup>6</sup>Li-glass and four <sup>7</sup>Li-glass scintillation detectors). The individual difference was evaluated to precisely measure neutrons using the neutron detector device with the moderator. Moreover, we fabricate a prototype of <sup>6</sup>Li-glass scintillation detector with a photodiode (Fig. 1). We also experimentally verified the <sup>6</sup>Li-glass scintillation detector with the photodiode

**EXPERIMENTS:** A collimated neutron beam with 30 mm diameter was obtained by the photo neutron reaction using a tantalum target with a water moderator at the KURRI Linac [2]. Figure 2 shows the experimental setup. The detector was set at 12 m away from the target. An enriched <sup>10</sup>B<sub>4</sub>C sample with 10-mm thickness was also used to subtract background thermal neutrons in the measurement room. The <sup>10</sup>B<sub>4</sub>C sample was set between the detector and end of the collimator in the beam line. A BF<sub>3</sub> proportional counter was used as a neutron monitor. All data were normalized using counts obtained with the BF<sub>3</sub> proportional counter. Characteristics of the neutron detection were experimentally evaluated by means of the time-of-flight (TOF) method.

**RESULTS:** Figure 3 shows the relative sensitivity of the <sup>6</sup>Li-glass and <sup>7</sup>Li-glass scintillation detectors for thermal neutron region, respectively. The thermal neutron sensitivities for the <sup>6</sup>Li-glass scintillation detectors were agreed within 0.5 %. The sensitivities for the <sup>7</sup>Li-glass scintillation detectors had variation within 6 %.

It is guessed that the neutron sensitivity of the <sup>6</sup>Li-glass scintillation detector with the photodiode is lower than that with the photomultiplier, because of difference of photon sensitivities of the photon detectors for 470 nm photon from the <sup>6</sup>Li-glass scintillator. In the first experiments of 2019, signals due to neutrons were not observed

clearly, although signals due to gamma flash from the target were successfully observed.



Fig. 1. Prototype of the <sup>6</sup>Li-glass scintillation detector with the photodiode.

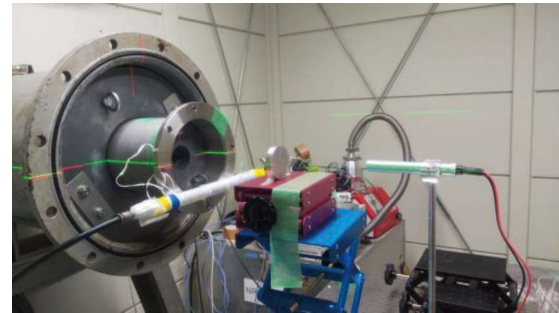


Fig. 2. Experimental setup for the <sup>6</sup>Li-glass scintillation detector. The detector was set at 12 m away from the target.

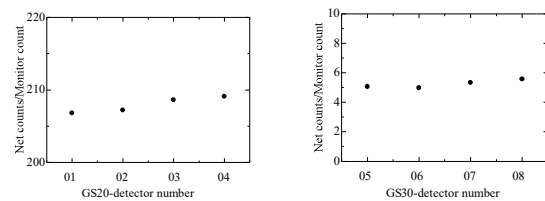


Fig. 3. Results of neutron sensitivities for the <sup>6</sup>Li-glass (GS20) and <sup>7</sup>Li-glass (GS30) scintillation detector.

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This work was supported by JSPS KAKENHI(JP16K21679, JP16K09030, 19K12638).

## CO2-4 Measurements of thermal neutron total and scattering cross section of moderator materials

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**INTRODUCTION:** The high temperature gas-cooled reactor is a Generation IV reactor concept that use a graphite moderator. The design takes advantage of the inherent safety characteristics with specific design optimizations. The treatment of thermal scattering caused by crystalline is important for graphite to improve the prediction accuracy of graphite moderated core analysis. There is sufficient thermal scattering law for single crystal graphite, but there is no method has been established to reconstruct thermal scattering cross section for polycrystalline graphite. On the other hand, a new concept of small modular reactor using CaH<sub>2</sub> as solid moderator has been developed [1]. The experimental and evaluated data of the thermal scattering law for CaH<sub>2</sub> are insufficient. In order to provide basic data for thermal neutron scattering law evaluation, the total and scattering cross sections of moderator materials were measured.

**EXPERIMENTS:** The total and scattering cross sections of moderator materials were measured in the incident neutron energy region from 0.001 to 10 eV by transmission and scattering experiments at the KURNS-LINAC. An experimental arrangement is shown in Fig. 1. Pulsed neutrons were produced from a water-cooled Ta-target by (γ,n) reaction with a pulsed electron beam. The incident neutron spectrum on a sample, the transmitted and scattering neutron spectrum were measured by means of a time-of-flight (TOF) method with a <sup>6</sup>Li-glass scintillation detector. A 5.0 mm diameter by 5.0 mm thick <sup>6</sup>Li-glass was located 12.0 m from the neutron source. The scattering neutrons were measured by a 50 mm diameter by 5.0 mm thick <sup>6</sup>Li-glass scintillation detector. The scattering neutron spectra were observed at angle of 45 degrees with respect to the neutron beam direction.

The characteristics of the samples are shown in Table 1. The graphite and polyethylene sample are used as a moderator in the KUCA facility.

**RESULTS:** The total cross sections of moderator materials were derived in the incident neutron energy region from 0.001 to 10 eV. Figure shows the scattering neutron spectra. The result of graphite had a structure caused by thermal scattering crystalline. Figure 3 shows the scattering neutron spectrum with the resonance filter (Cd, In, Ag, Mn). It was shown that it is possible to measure quasi-monochromatic neutrons from a white neutron source.

**ACKNOWLEDGEMENTS:** This research is a collaborative effort of Toshiba Energy Systems & Solutions, Kindai University, Kyoto University and Tokyo Institute of Technology.

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- [1] R. Kimura and S. Wada, Nucl. Sci. Eng., Vol.193, p.1013-1022, (2019).

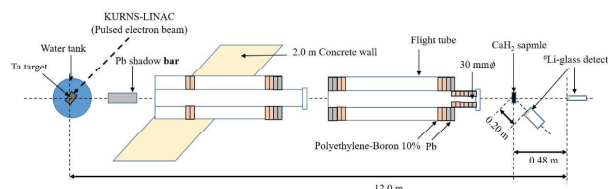


Fig 1. Experimental arrangement.

Table 1. Characteristics of samples.

Sample	Polyethylene	Graphite	CaH <sub>2</sub>
Structure	High molecular	Polycrystalline	Powder
Sample shape	Square plate	Square plate	Disk in Al case
Density [g/cm <sup>3</sup> ]	0.949	1.754	1.200
Size [mm]	50.8 × 50.8	50.8 × 50.8	φ50
Thickness [mm]	3.18	12.7	5.0
[atoms/b]	1.26 × 10 <sup>-2</sup>	1.12 × 10 <sup>-1</sup>	8.63 × 10 <sup>-3</sup>

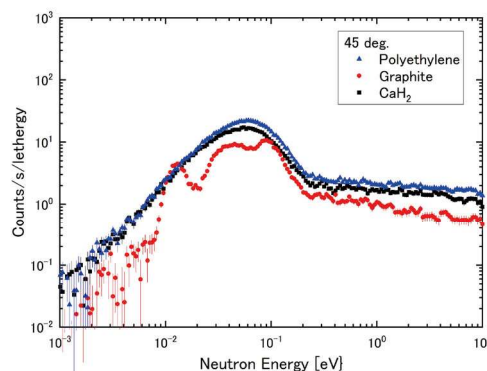


Fig. 2. Scattering neutron energy spectra.

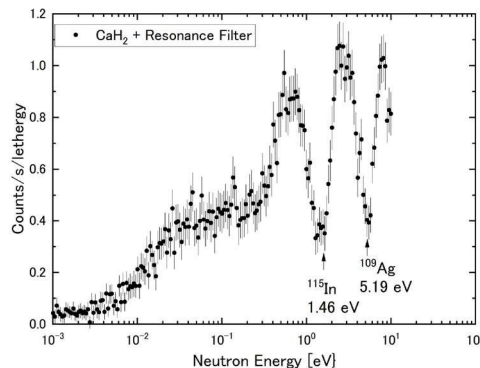


Fig. 3. Scattering neutron energy spectrum with resonance filter (Cd, In, Ag, Mn).



# CO2-5 Measurement of Temperature depended Thermal Neutron Spectrum in Solid Moderator

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## INTRODUCTION:

In recent years, small modular reactors (SMR) have been studied intensively. Those SMR are employed existing technologies such as light-water moderator and coolant and so on. On the other hand, a new concept of SMR using solid moderator has been developed [1]. Here, the moderator in a nuclear reactor is common material such as light-water, researchers and engineers could calculate the accurate neutron spectrum in the moderator by using evaluate nuclear data libraries such as the JENDL-4.0 [2]. However, if evaluate nuclear data libraries has poor data for moderator materials, the calculations of accurate neutron spectrum in the moderator will be difficult. The new concept of SMR is employed CaH<sub>2</sub> as solid moderator [1].

Therefore, in order to perform a nuclear design for the reactor with CaH<sub>2</sub>, it is necessary to experimentally investigate the temperature-dependent neutron spectrum in the moderator. In last year, a preliminary experiment of temperature-dependent neutron spectrum measurements was carried out [3]. In the present study, the experimental geometry and condition were optimized based on the results of preliminary experiment and the temperature-dependent neutron spectrum measurements were performed.

## EXPERIMENTS:

To measure the temperature-dependent thermal neutron spectrum in CaH<sub>2</sub> sample, a preliminary experiment was conducted method in KURNS-LINAC. The CaH<sub>2</sub> sample was set in a heater to be increased the sample temperature from 21°C to 470°C. Figure 1 shows an experimental geometry of present study. The heater was installed at a position of 12.6 m from the pulsed neutron source at KURNS-LINAC. In this experiment, a GEM type neutron detector was employed. The neutron flight length between the CaH<sub>2</sub> and the GEM detector was 60 cm. Table 1 shows the experimental conditions and Table 2 is the sample information. The transmitted neutron flux from the sample was observed by the TOF method in the present study.

## RESULTS:

The obtained thermal neutron TOF spectrum of the CaH<sub>2</sub> at room temperature (21°C) and 470°C are shown in the Fig. 1. The difference of thermal neutron spectrum between 21°C and 470°C was clearly observed.

## ACKNOWLEDGEMENTS

This research is a collaborative effort of Toshiba Energy Systems & Solutions, Kindai University, Tokyo Institute of Technology and Kyoto University.

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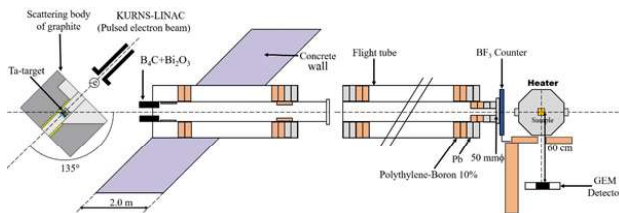


Fig.1. Experimental geometry.

Table 1 Experimental condition.

Sample	LINAC operational condition (Frequency, pulse width, current)	Sample temperature (degree)	Measuring time (h)
CaH <sub>2</sub>	35Hz, 3 μs, 53 μA	21	10
CaH <sub>2</sub>	35Hz, 3 μs, 53 μA	470	16
Blank	35Hz, 3 μs, 53 μA	21	5.5

Table 2 Sample information.

Chemical formula	CaH <sub>2</sub>
Density	1.33 g/cm <sup>3</sup>
Geometry	70 mm × 70 mm × 60 mm

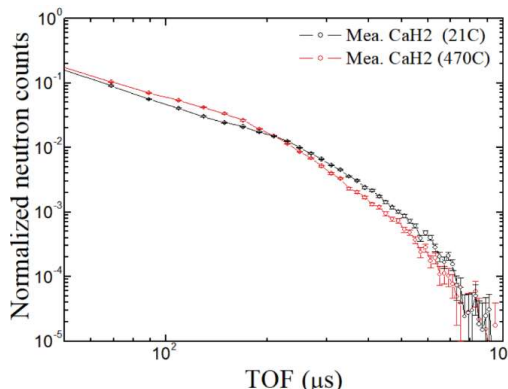


Fig.1. Obtained thermal neutron TOF spectrum of CaH<sub>2</sub> sample at 21°C and 470°C.