Gamma-Ray Spectrum Measurement from Capture Reactions of Uranium-238 for Thermal and Resonance Energy Neutrons

Yasushi Nauchi¹*, Jun-ichi Hori², Kazushi Terada², and Tadafumi Sano³

¹Central Research Institute of Electric Power Industry, Energy Transformation Research Laboratory, 2-6-1, Nagasaka, Yokusuka, Kanagawa 240-0196 Japan.
²Kyoto University, Institute for Integrated Radiation and Nuclear Science, 2, Asashiro-Nishi, Kumatori-cho, Senann-gun, Osaka 590-0494 Japan.
³Kindai University, Atomic Energy Research Institute, 3-4-1, Kowakae, Higashi-Osaka, Osaka 577-8502 Japan.

Abstract. γ-ray spectrum from the capture reaction of 238U is measured for the thermal and resonance energy neutrons at KURNS-LINAC facility. Prominent primary γ-ray of 3.297 MeV listed in CapGam is not observed for the thermal energy neutrons. The measured γ-ray spectrum is varied with neutron energy so that we can expect neutron spectrum inducing 238U(n,γ) reactions in critical / subcritical cores could be deduced by the γ-ray spectroscopy for the cores. Preliminary deduced ratio of 4.060 MeV γ-ray emission to capture for the thermal energy neutron agrees with CapGam so that the deduction scheme is considered adequate.

1 Introduction

When power of a reactor core is escalated, the earliest phenomena to bring negative reactivity into the core is the doppler effect. The mechanism of the effect is the broadening of the energy width of resonance capture reactions, which results in the increment of neutron absorption rate. In light water reactors, resonance capture reactions in the energy range less than 100 eV are significant [1]. Whereas 235U(n,γ) is the reaction to generate 239Pu which enables extension of burn-up of fuel. For the reason, the capture reaction of 235U is significant both for the reactor safety and its economical use [2].

From those viewpoints, a validation of calculation of 235U(n,γ) reaction rate in a reactor core is desirable. One of the authors has succeeded in γ-ray spectroscopy in a critical core with highly enriched uranium fuel [3]. He has also succeeded in detection of 4.060 MeV γ-rays in sub-critical systems [4]. That is one of the most frequently emitted primary γ-rays from 235U(n,γ) reactions for the thermal energy neutrons, according to CapGam data base [5]. Thus, γ-ray spectroscopy for an integral system is available nowadays. Focusing on differential experiments, Harada et al. has reported that the γ-ray spectrum varies with the incident neutron energy of the 235U(n,γ) reaction [6]. Accordingly, it is expected that the neutron energy spectrum inducing 235U(n,γ) reactions in an integral system might be deduced by the γ-ray spectroscopy. For the purpose, we need the data of ratios of γ-ray emission to capture in 235U(n,γ) reaction for each neutron energy.

For the reason, we have started the measurement of the γ-ray emission for neutron energies in differential experimental manner [7]. In this manuscript, outline of the measurement and the preliminary data processing are reported.

2 Experimental

The differential measurements were conducted at the LINAC facility in Institute for Integrated Radiation and Nuclear Science, Kyoto University (KURNS). Schematic view of the experiment is shown in Fig. 1.

2.1 Neutron Source

In the facility, electrons accelerated up to 30 MeV by the LINAC are injected onto a tantalum target located in a cylindrical aluminium container of 20 cm in diameter and 30 cm in height. In the target, the electrons are slowed down radiating the Bremsstrahlung rays. The rays induce the photo-nuclear reactions radiating neutrons. The neutrons are moderated in the container filled with light water. The repetition rate of the electron beam was 50 Hz and the pulse width was 1 μs. The beam current was nominally 27 μA.

2.2 Geometry and data acquisition

A metallic sample of uranium of the natural 235U enrichment was placed effectively 11.3 m distant from the tantalum target. The areal density and the size of the sample were 2.61 g/cm² and 4 cm × 4 cm, respectively.
The γ rays emitted by reactions induced by the neutron of the white energy spectrum were measured with a HPGe detector which has a nominal 35% relative efficiency. The normal axis of the sample was inclined to 45°, to enable easier corrections for neutron attenuation in the sample and to mitigate dependence of γ-ray detection efficiency to the axis. The nominal distance from the sample centre to the detector surface was 5 cm. The HPGe detector should be shielded from the neutrons and the γ-ray flash. For that, the neutrons passing through a collimator was used and the HPGe was located off the axis of the collimator. Besides, filtering material was put upstream of the collimator as needed.

The pulse height signals from the HPGe detector for γ rays were acquired together with the time of flight (TOF) event by event in the list mode. The TOF information in reference to the pulsed electron beam was taken to determine the incident neutron energy. The data acquisition diagram is shown in Fig. 2.

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### 2.3 Measurement for calibration

To determine the neutron flux, a boron sample of 1.48 g was placed instead of the U sample. The 10B enrichment of the boron sample was 97.24 at%. 10B(n,α)7mLi de-excitation ray of 478 keV from the boron sample were measured. The detection efficiency of the 478 keV γ-ray was given by γ-ray measurement of a standard mixed γ-ray source. To calibrate the pulse height response to the γ-ray energy and to determine the relative efficiency in the energy range from 2.7 to 4.5 MeV, a NaCl sample was placed and the 35Cl(n,γ) γ rays were measured for the thermal energy neutrons. To calibrate the TOF data to the neutron energy, so called filter notch method was used where 10B(n,α)7mLi 478 keV γ-ray was measured for the boron sample irradiated by neutron filtered by cadmium, indium, silver, cobalt plates.

### 3 Data Processing and Results

#### 3.1 Determination of neutron energy

Experimental campaign is currently under way so that preliminary data processing scheme is described. The overall TOF spectrum is shown in Fig. 3. A TOF channel corresponds to 2 μs. A widely spread peak from 1000 to 5000 channels corresponds to the thermal energy neutron induced events. The spectrum is flat between 300 and 10000 channels and there are sharp peaks from 32 to 200 channels.

![Fig. 3. Time of flight spectrum of neutrons for Uranium sample.](image)

In Fig. 4, the filtered TOF spectrum for the boron sample is shown. The notches by captures of 54Co (132 eV), 109Ag (5.10 eV) and 115In (1.46 eV) are remarkable as well as cut off of thermal energy neutrons by 113Cd (0.18 eV). Instead of the remarkable notches, we used rather shallower dips for calibration of TOF to the neutron energy for better accuracy. The used resonances are listed in Table 1 referring [8]. By the calibration, the resonance peaks in Fig. 3 are found to correspond to $E_n = 6.67, 20.87, 36.65$ eV. In this experiment, neutron spectrum above 50eV have not yet been measured.

#### Table 1. Resonance Energy to calibrate TOF.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Neutron energy (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>107Ag</td>
<td>16.30, 30.60, 51.56, 71.0</td>
</tr>
<tr>
<td>109Ag</td>
<td>5.19, 55.80</td>
</tr>
<tr>
<td>113In</td>
<td>14.6</td>
</tr>
<tr>
<td>115In</td>
<td>3.82, 9.07, 12.0, 22.73, 39.56</td>
</tr>
</tbody>
</table>
Fig. 4. Time of flight spectrum of neutrons for uranium sample.

3.2 Pulse height spectrum

The pulse height spectra were sorted corresponding to the thermal, epi-thermal, and resonance (6.67, 20.87, and 36.68 eV) energy neutrons. The pulse height data were calibrated to $\gamma$-ray energy referring the $^{35}$Cl(n,$\gamma$) $\gamma$-ray spectrum for the thermal energy neutron. The peak energies of the $^{35}$Cl(n,$\gamma$) reaction were referred to [5]. In this study, “primary” $\gamma$ rays from the capture reactions are focused on since we want to apply $\gamma$-ray spectroscopy not only for fresh fuels but also for spent ones. To identify the target $\gamma$ rays from FP $\gamma$s, the focused energy region is from a few to several MeV. For the purpose, $\gamma$-ray spectrum from $^{35}$Cl(n,$\gamma$) is suitable. The measured pulse height response was almost linear to the peak energies but a cubic equation was used to interpolate.

The measured spectrum is shown in Fig. 5. A photo-electric (PE) peak of 4060 keV is found except for the resonance of $E_n = 36.68$ eV. The peak to continuum ratio is smaller for the thermal and epi-thermal energy neutrons due to the fission prompt $\gamma$-ray component. Contrarily, the ratio is larger in the resonances of $E_n = 6.67$ and 20.87 eV. In addition to PE peaks, there are single (SE) and double escape (DE) peaks.

In Fig. 6, the primary $\gamma$ rays of larger yield from $^{238}$U(n,$\gamma$) for thermal energy neutrons listed in CapGam are shown. 4060, 3991, and 3982 keV $\gamma$ rays are found both in Fig. 5 and 6. However, the most prominent one in CapGam, 3297 keV, is not found in our data (Fig. 5). Accordingly, the data of 3297 keV should be reviewed.

Harada had measured $\gamma$-ray peaks for resonance neutron energies by suppressing DE and SE components. The peaks Harada detected are listed in Table 2. Since we could not suppress the SE and DE components, we cannot identify 3541 and 3482 keV components. On the other hand, the energy resolution is superior so that we succeeded in resolving 3982 and 3991 keV. In the current work, bump structures are found near 3817 keV. It was also measured in the Harada’s work [6] but the energy was not listed.

The $\gamma$-ray spectrum varies with the incident neutron energy. Only for $E_n = 36.68$ eV, 4067 keV component is found although the counting statistics is less. The peak count ratio of (3991 + 3982 keV) to 4060 keV is drastically changed between the thermal energy and $E_n = 6.67$ and 20.87 eV. The events induced by $E_n = 20.87$ eV neutrons might be distinguished by finding 3197 eV $\gamma$ rays. Then checking the ratio of (3991 + 3982 keV) to 4060 keV, the events induced by the thermal and resonance energy neutrons might be quantified in relative manner.

![Fig. 5 $\gamma$-ray spectrum for each neutron energy.](image)

![Fig. 6 Relative $\gamma$-ray yield from $^{238}$U(n,$\gamma$) for thermal energy neutrons listed in CapGam [5].](image)

Table 2. Peak $\gamma$ rays identified for resonance neutron energy.

<table>
<thead>
<tr>
<th>$E_n$(eV)</th>
<th>6.67</th>
<th>20.87</th>
<th>36.68</th>
</tr>
</thead>
<tbody>
<tr>
<td>4067</td>
<td>C+H</td>
<td>C+H</td>
<td>C+H</td>
</tr>
<tr>
<td>4060</td>
<td>C+H</td>
<td>C+H</td>
<td>C+H</td>
</tr>
<tr>
<td>3991</td>
<td>C</td>
<td>C</td>
<td>C</td>
</tr>
<tr>
<td>3982</td>
<td>C+H(+3991)</td>
<td>C+H(+3991)</td>
<td>C+H(+3991)</td>
</tr>
<tr>
<td>3583</td>
<td>C+H</td>
<td>C+H</td>
<td>C+H</td>
</tr>
<tr>
<td>3567</td>
<td>H</td>
<td>H</td>
<td>H</td>
</tr>
<tr>
<td>3541.3</td>
<td>H</td>
<td>H</td>
<td>H</td>
</tr>
<tr>
<td>3481.7</td>
<td>H</td>
<td>H</td>
<td>H</td>
</tr>
<tr>
<td>3197.2</td>
<td>H+C</td>
<td>H+C</td>
<td>H+C</td>
</tr>
</tbody>
</table>

C+H: Peak is found both in current work and [6]  
H: Peak is found only in [6]
3.3 Deduction of \( \gamma \)-ray emission per capture

Measurement and data processing with the pulse-height weighting method is under way to deduce the capture reaction rates, experimentally. In this work, the reaction rates were numerically calculated and the ratio of \( \gamma \)-ray emission to capture was preliminary deduced.

Based the count rate of 478 keV \( \gamma \)-ray from the boron sample irradiated by the neutron and the count rate of the \( \gamma \) rays from the standard mix \( \gamma \)-ray source, the emission rate of 478 keV \( \gamma \) rays were quantified. The attenuation of the neutron flux in the boron sample was corrected by the neutron transport calculation with the MCNP-5 code [9] and the AcelibJ40 library [10]. By the calculation, the \(^{10}\text{B}(n,\alpha)\) reaction rate per source intensity was given. With the deduced emission rate and the reaction rate per source, the source intensity and the flux were determined for the thermal and resonance energy neutrons. Using the thermal energy neutron flux, we calculated the \(^{35}\text{Cl}(n,\gamma)\) reaction rates in the NaCl with MCNP-5 and AcelibJ40. Then we deduced the number of \(^{35}\text{Cl}(n,\gamma)\) \( \gamma \)-ray emission. Based on the measured count rates of the photo-electric peaks and the number of \( \gamma \)-ray emission, the detection efficiency was given. The difference of neutron attenuation in the NaCl and the uranium samples were numerically estimated with MCNP-5 and Acelib-J40. With the deduced \( \gamma \)-ray count rate from the uranium sample and the efficiency, the number of \( \gamma \)-ray emission from \(^{238}\text{U}(n,\gamma)\) was given. There, background was subtracted using count rates in the TOF regions next to each peak.

Whereas the \(^{238}\text{U}(n,\gamma)\) reaction rates for the thermal and resonance energy neutrons were calculated with MCNP5 +AcelibJ40 taking the measured neutron flux into account. Finally, the ratio of \( \gamma \)-ray emission to capture was deduced.

The ratios are shown in Fig. 7. For the most prominent primary peak of 4060 keV, the ratio listed in CapGam agrees with the currently deduced one based on the measured data. It indicates that the data processing scheme is credible.

4 Summary

To develop an estimation technique of neutron spectrum inducing \(^{238}\text{U}(n,\gamma)\) reactions inside integral systems, we have measured \(^{238}\text{U}(n,\gamma)\) \( \gamma \)-ray spectrum for the thermal and resonance energy neutrons at LINAC neutron source facility in KURNS. Compared to Harada’s work [6], the spectrum was obtained with better energy resolution although some photo-electric peaks were overwrapped by SE and DE peaks. By the measurement, the most prominent primary \( \gamma \) ray of 3297 keV listed in CapGam was not found. Our data would be helpful to improve such database. The ratio of \( \gamma \)-ray emission to capture was preliminary deduced using numerically calculated reaction rate of \(^{238}\text{U}(n,\gamma)\) and the measured \( \gamma \) rays. The ratio of 4060 keV for the thermal energy neutrons agrees with the CapGam data so that the data processing scheme is credible.

As the future work, we would like to check the reproductivity of the data by multiple measurement. Beam current would be enhanced more for the better accuracy paying attention to the \( \gamma \) flash. Besides, back grounds due to the fission prompt \( \gamma \) rays and delayed \( \gamma \) rays shall be considered. Additionally, the pulse height weighting measurements with a BGO detector would be conducted and the reaction rate data would be deuced to determine experimentally the ratio of \( \gamma \) ray emission to capture.

References