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Neutron capture cross section measurements on ^{237}Np with a 4π Ge spectrometer

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Abstract. Neutron capture cross sections on minor actinides are important in the design work for innovative nuclear reactor systems. In order to improve data accuracy, a 4π Ge spectrometer system surrounded with large BGO detectors for Compton suppression has been developed. Capture cross sections of ^{237}Np were obtained and resonance analyses using the code SAMMY were performed for several low energy resonances. Measurements were carried out at the electron Linac of KURRI (Kyoto University Research Reactor Institute) with an electron energy of 30 MeV.

1 Introduction

Accurate data of neutron cross sections are important in the design work for innovative nuclear reactor system that will be utilized to produce economical carbon free nuclear energy to meet growing electricity demand in the world. In particular, the closed fuel cycle system is envisaged to require the development and deployment of technologies that enable recycling and consumption of long-lived radioactive waste.

Neutron capture cross sections of minor actinides (MA) and long-lived fission products (LLFP) have attracted attention recently in the field of nuclear systems such as transmutation of radioactive waste and various innovative reactor concepts proposed for the Generation-IV. However, accurate measurements of these cross sections are very difficult due to high natural radioactivity of these samples. ^{237}Np is one of the most important nuclei in the minor actinides. In the past, several groups have measured the neutron capture cross section of ^{237}Np in the thermal neutron and resonance energy range [1–4].

The previous measurements were performed by the neutron time-of-flight (TOF) method using an electron Linac in the resonance region [1–4] as a pulsed neutron source and used C_6F_6, C_6D_6 or BGO detectors. To reduce the high background caused by γ-rays of the radioactive sample, the pulse-height bias corresponding to 0.5 ~ 1.0 MeV was used and it was necessary to insert rather thick lead shields between the sample and detectors or to arrange coincidences between the detectors in order to obtain a good signal-to-noise ratio.

In spite of the fact that there exist relatively large amounts of data, there are still discrepancies remained among the evaluated nuclear data files generated on the basis of the previous data [5]. New concepts for data taking methods may be necessary to improve the precision in the neutron capture cross section measurements in this research field. In the present experiment, a newly developed detector system with a 4π Ge spectrometer is introduced to improve data accuracy for MA and LLFP capture cross section measurements.

2 Experiments

2.1 4π Ge spectrometer and TOF facility

When a nucleus captures a neutron, the nucleus is excited in a capture state and followed by emitting γ-rays. The good energy resolution of a Ge detector is suitable for high-resolution measurements with ~2 keV for 1-MeV γ-rays. Although the efficiencies of scintillation detectors are higher than those of Ge detectors, the energy resolutions of scintillation detectors are not enough to resolve the energy differences of neutron capture states and γ-rays from the various sources of the background. On the other hand, the Ge detector system has to cover the sample with a solid angle as large as possible, and the efficiency of the detector should be high enough to detect all the γ-rays emitted from the excited nuclei. A project has been proposed and carried out to use such a 4π Ge detector for the MA and LLFP neutron capture cross section measurements to implement new and accurate experimental methods with better signal to noise ratio [6]. The characteristics of the detector system are presented in some detail elsewhere in this conference [7, 8].

The neutron capture detector system was set at a TOF neutron beam line of the KURRI electron LINAC. The electron beam irradiated a neutron target of laminated Ta plates surrounded with a water moderator. A Pb shadow bar was placed in front of the Ta target to stop the γ-flash. To reduce the background due to γ-rays and neutrons from the collimators, the neutron flight path was covered with paraffin blocks, Pb blocks, boron-mixed polyethylene rings, and B_4C shields. The Linac was operated at a beam energy of 30 MeV, an average current of 30 μA, a pulse width of 0.1 μs and a repetition rate of 100 Hz. The distance between the sample position and the moderator surface was about 10 m.

The schematic drawing of the 4π Ge spectrometer is shown in figure 1. It is composed of two cluster detectors and four clover detectors. The spectrometer was covered with BGO Compton suppression detectors, which eliminated background Compton events. The individual efficiency of each module of

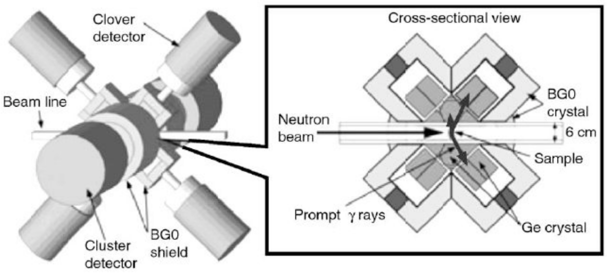

Fig. 1. Schematic drawing of the 4π Ge spectrometer.

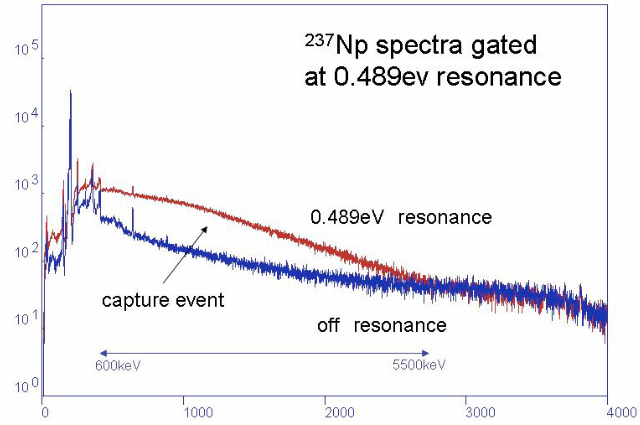

Fig. 2. Gated γ-ray spectra from the 0.489 eV ^{237}Np resonance.

the cluster Ge detector and that of the clover Ge detector were estimated to be 57% and 22%, respectively [9, 10].

2.2 Data acquisition system

The data acquisition (DAQ) system for the 4π Ge spectrometer is required to deal with a large amount of signals from the spectrometer which consists of 40 γ-ray signals from the Ge crystals and more than hundred signals from BGO anti-Compton shields. Since MA samples are highly radioactive and the DAQ has to manage high event rates, a high-performance DAQ system based on a digital data processing technique is developed [7]. The new DAQ system consists of three modules: main ADC modules, fast timing modules, and coincidence modules. Each module has a dual port memory and a main digital signal processor (DSP), which is a programmable processor.

Time intervals between an electron beam pulse and detected γ-rays were measured by the coincidence module. The time resolution of the time interval was 5 ns. The information on the pulse amplitudes and the detection times were stored on the dual port memories that were transferred to the memories. The events from the 30 Ge detectors were stored with the list mode (event recording mode). About 10 Gbyte data for the ^{237}Np sample were stored. In addition, the same amounts of data were stored for flux measurement and background determination.

2.3 Samples

The capture samples were set in the center of the detector. Two samples of ^{237}Np were used: 0.227 g (5.18 MBq) and 1.128 g (26 MBq) of neptunium oxide (NpO_2) powder packed in an aluminum disk container 30 mm in diameter and 0.4 mm thick walls. The thicknesses of the samples were 0.0637 g/cm^2 and 0.316 g/cm^2, respectively. The dummy sample of identical Al case without the neptunium oxide powder was used for the background determination. The Pb scatterer was also used to simulate the effects from scattered neutrons from the sample and surrounding materials. The incident neutron flux shape was measured with the ^{10}B sample made of boron powder encapsulated in a cylinder thin-walled (0.2 mm) aluminum container 25 mm in diameter with a sample material thickness 0.947 g/cm^2. The ^{10}B enrichment was 93%.

3 Data processing and analysis

3.1 Capture γ-ray yield

The data were measured with event-by-event mode (list mode) storing the information of γ-ray pulse height (PHA), time-of-flight (TOF) and time-interval-of-coincidence (FER) from the detectors for various samples, ^{237}Np, ^{10}B, Pb scatterer, Al case, NaCl powder and standard γ-sources (^{152}Eu, ^{60}Co, ^{137}Cs). In addition, the background information was also obtained with a black resonance filter method, in which thick metallic samples of In, Ag, Co and Mn were inserted upstream of the sample in the flight path. The data were sorted according to the PHA and TOF information, and two dimensional data sets (matrix) were generated. The sorting was also done by taking into account the FER information to eliminate the contamination from the spurious signals due to the γ-flash or radio-frequency noises from the accelerator. The energy calibration of the PHA signals was made using the well known γ-ray energies emitted from the neutron capture of chlorine in the NaCl sample. Neutron energy calibration was made using the narrow resonances observed in the Ag resonance filter. Figure 2 shows the gated γ-ray spectrum (PHA) data of the 5 MBq ^{237}Np sample for the prominent 0.489 eV resonance and near-by off-resonance region. The difference between two spectra was net counts from the neutron capture gamma-ray events. On the other hand, the TOF spectra were obtained by taking the events gated with the PHA region between 600 keV and 5.5 MeV.

3.2 Dead time correction

The dead time correction was the most important correction factor for the present experiments because ^{237}Np sample was highly active and 40 sets of Ge detectors produced many gamma-ray events. The several tens of 10^3 counts/s events were expected even with a rather high bias level of 400 keV with thick 5 mm Pb shields between the sample and detectors, which caused unavoidable dead time loss of the system. The dead time was dependent on very complicated data acquisition procedure. In the present experiment, an external random pulser was prepared and the pulse signals corresponding to the

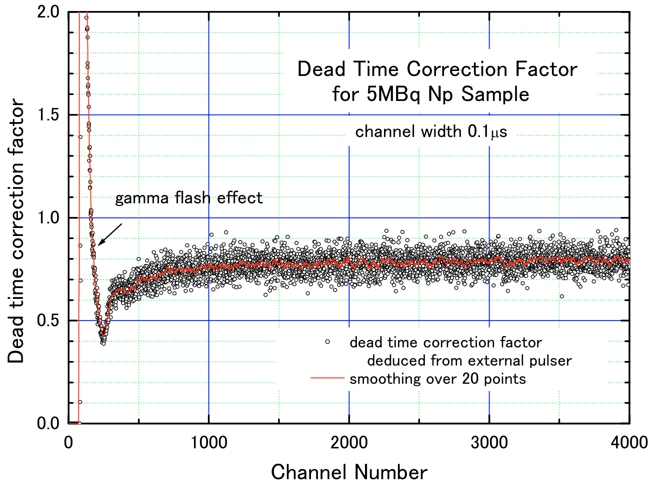

Fig. 3. Dead time correction factor for capture gamma-ray yield from the ^{237}Np 5 MBq sample.

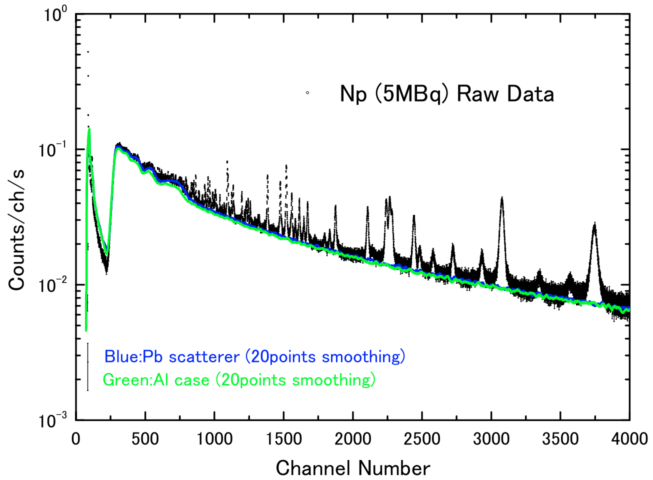

Fig. 4. Neutron capture gamma-ray yields from 5 MBq ^{237}Np samples, Pb scatterer and Al case.

PHA value of about 0 MeV were fed into the input terminals of the 40 individual preamplifiers. The stored counts were used for the dead time correction by comparing the counts of input pulses with actually stored pulses. The time dependent (TOF) dead time was obtained directly from this method. In our experiment, the dead time correction factor of about 10% ~ 20% was obtained as shown in figure 3.

3.3 Background determination

The capture gamma-ray yields from Pb scatterer and Al case were measured to obtain the background information. In this experiment, the counts from Pb scatterer and Al case were the same in the neutron energy range below 20 eV.

The effect from overlapping neutrons to the consecutive neutron pulse was also deduced from the counts in the TOF time range from 6 ms to 10 ms by extrapolation with the exponential function. The overlap neutron effect was less than 1% above ev region. The counts from ^{237}Np, Pb scatterer and Al case are shown in figure 4. The counts smoothed

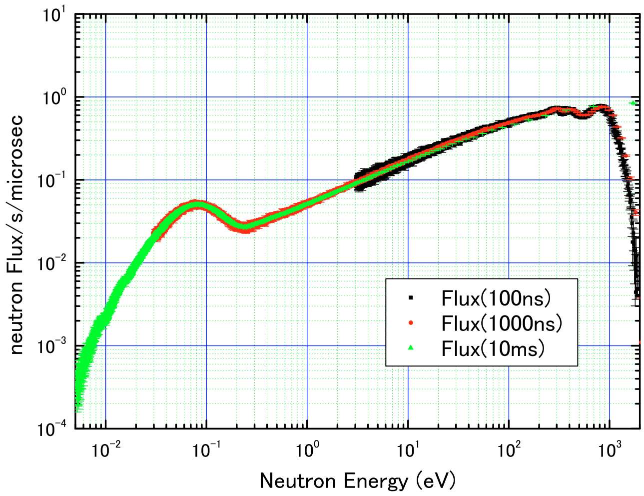

Fig. 5. Neutron flux shape obtained using 478 keV γ-rays of ^{10}B(n,$\alpha\gamma$) and JENDL-3.3 evaluated cross section.

over 20 points from Pb scatterer were taken as the energy dependent background. For the ^{10}B sample, the TOF (energy) dependence of the background was obtained by interpolation between the values observed in the transmission minima measured with black resonance filters: 336 eV (Mn), 132 eV (Co), 5.19 eV (Ag) and 1.457 eV (In).

3.4 Neutron flux

The neutron flux was deduced from the TOF spectrum of the 478 keV γ-ray, which was emitted in the ^{10}B(n,$\alpha\gamma$) reaction. The neutron capture cross-section data were taken from the Japanese Evaluated Nuclear Data Library (JENDL) version 3.3 [11]. Figure 5 shows the relative neutron flux shapes with the different energy bins.

4 Results and discussion

The shape of the ^{237}Np capture yield was obtained as a function of neutron energy in the present experiment. To obtain absolute value of the present capture cross section, the previous experimental and evaluated data were referred. The data by Weston and Todd [1], Kobayashi et al. [2], Harada et al. [4], the evaluated data of JENDL-3.3 [11], and the recommended value by Mughabghab [12] are 180 ± 6, 158 ± 3, 169 ± 4, 161.73 and 175.9 ± 2.5, respectively, which are within about 10% variation. By referring these values, the present data were normalized to the reference value of 179 b at 0.025 eV. The 0.489 eV resonance was first analyzed by the SAMMY code [13] and checked the normalization based on the large peak cross section region, because the present sample was rather thick (~90% of saturation) and gave the indication of the absolute value. By using this information, normalization could be reliable to be about less than 10%. The result of the present capture cross section is shown in figure 6 together with the evaluated value of JENDL-3.3. Our statistical uncertainly was 2% ~ 15%, which was not included in the error from the

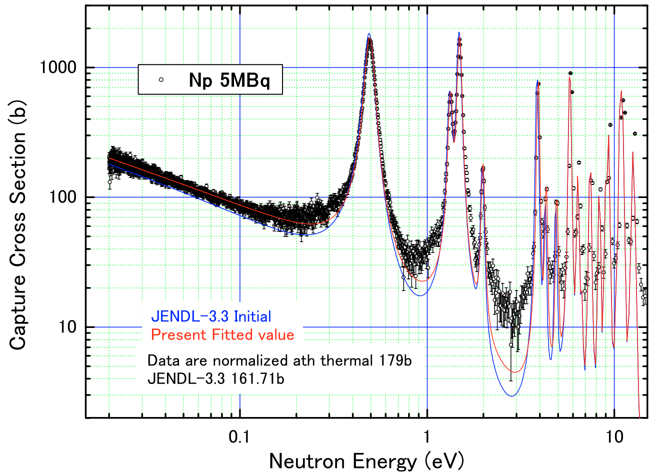

Fig. 6. Deduced capture cross section of ^{237}Np and comparison to JENDL3.3.

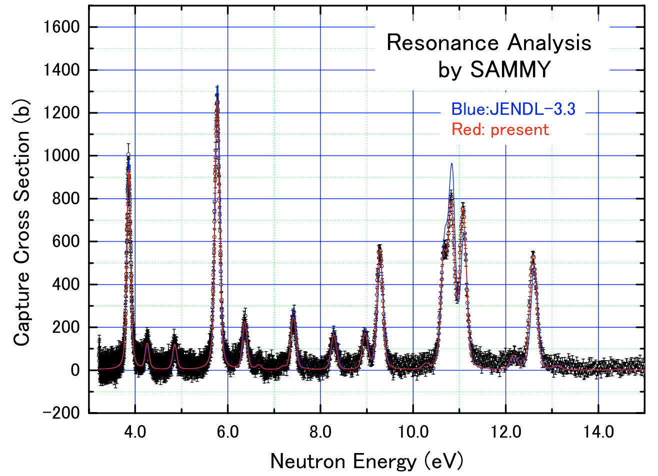

Fig. 7. Resonance analyses with SAMMY in the neutron low energy region.

background determination. The overall error of the data was so far estimated to be 5% ~ 10%.

The resonance analyses of the deduced cross section values were carried out for the low energy resonance below 20 eV. An example is shown in figure 7 and the comparison between JENDL3.3 and present values is shown in figure 8. The self-shielding effects were corrected with the SAMMY code. The overall agreement between two sets of data was within 1% indicating the normalization was valid.

5 Summary

The new experimental facility for neutron capture cross section measurements on MA and LLFP and the 4π Ge spectrometer were built with excellent gamma-ray energy resolution to obtain better signal to background ratio. The development of a new DAQ system based on an advanced digital processing technique could handle a large amount of the capturing events

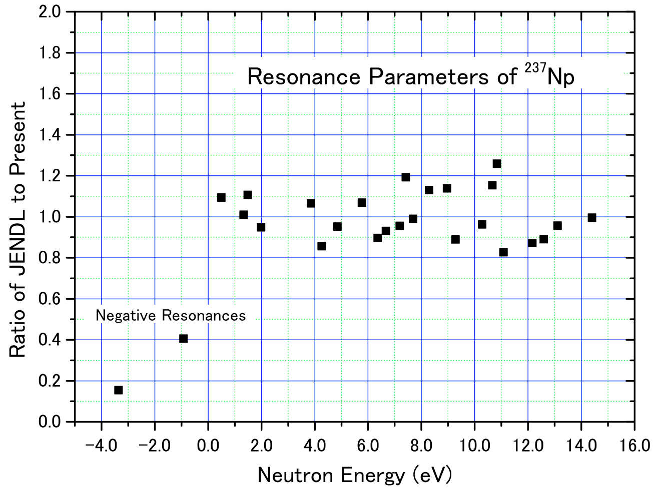

Fig. 8. Comparison of resonance parameters between presents and JENDL-3.3 for ^{237}Np.

to provide the precise capture gamma-ray spectral information. Capture cross section data of ^{237}Np were measured at this new experimental facility. The data are so far deduced on the basis of the thermal cross section value. The resonance analyses were made for several low energy resonances. The 0.496 eV prominent resonance was analyzed and used to check the reliability of the normalization of the thermal cross section value. Further study will be made to determine more accurate values such as the Ge detector efficiency to obtain the absolute cross section value.

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