High-Energy Measurement of the Neutron Capture Cross Section of ^{237}Np

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Abstract

Neutron capture cross section measurements for ^{237}Np have been conducted with the Accurate Neutron Nucleus Reaction Measurement Instrument (ANNRI) at the Materials and Life Science Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC) using neutrons with energy ranging from thermal energy to 500 keV. Two normalization techniques were used to obtained the absolute cross-section with agreement within 2%. In the high energy region, from 0.5 to 500 keV, the cross-section results were obtained with errors of 4% or lower below 25 keV. Along with the cross section measurement, theoretical calculations were performed to reproduce the present results.

I. Introduction

Accurate nuclear data are of the essence for the nuclear transmutation of minor actinides (MAs) as the nuclear industry is set to tackle the issue of high-level waste (HLW) management. Nuclear transmutation facilities will take the task of converting HLW into short lived or even stable nuclei. Current evaluated nuclear data libraries are only suitable for the early stages of the design of nuclear transmutation systems. However, final designs and safety measures demand more precise nuclear data with a significant reduction in terms of their uncertainties [**?**, **?**].

^{237}Np has a long half-life of 2.14 x 10^6 years and it is one of the most abundant MAs present in spent nuclear fuel. ^{237}Np is also one of the main components of the Accelerator-Driven Systems (ADS) core, a subcritical reactor facility for nuclear transmutation. The region of interest for the core design is from 0.5 to 500 keV. Current uncertainties in JENDL-4.0 [**?**] for the neutron capture cross section of ^{237}Np (6-10%) are much higher than the requirements of 3%. Hence, it is of upmost importance to accurately determine the neutron capture cross section at such energy range to reduce the uncertainties.

In the region from 0.5 to 500 keV, the available experimental data is limited as there are only two sets of reliable data using TOF method, those of Weston *et al* [**?**] and Esch *et al* [**?**], but they diverge in the region of interest from 15% to 35% at some energies. Experimental data by activation method exists in the 100-500 keV range but they differ from each other about 30-40% [**?**, **?**, **?**].

In this paper, results of the neutron capture cross section for ^{237}Np are presented for incident neutron energy ranging from thermal energy to 500 keV with emphasis in the region of 0.5 keV to 500 keV. Moreover, in order to provide more accurate data, two normalization techniques were used to obtained an absolute value for the cross-section. The first technique was based on using the whole shape of the first resonance from JENDL-4.0 to determine a normalization factor. In the second technique, the energy-dependent cross-section was normalized using the total neutron flux obtained from a Au sample measurement in which the first resonance was completely saturated. In addition, the results of the experiments were complemented using the CCONE code in order to assess the reliability of the results. Details of the experimental setup and the data analysis are also provided.

II. Experimental Procedure

1. *Experimental Setup*

The experiments were performed using the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) at the Materials and Life Science Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC). Intense pulsed neutrons were produced by the Japanese Spallation Neutron Source (JSNS) in the MLF using the 3 GeV proton beam of the J-PARC facility. The proton pulses were shot at the spallation target every 40 ms and a beam power of 400 kW.

A TOF method was employed in the present experiment with a flight path of 27.9 m up to the sample position. Emitted γ-rays from the sample were detected by a NaI(Tl) detector. Detected capture events were stored sequentially in a computer as a list format data.

A multi-event time digitizer FAST ComTec MPA4T was employed for fast data acquisition purposes [?]. The time between the starting spallation trigger event and successive multiple stop from the detected γ-rays were digitized. The signal coming from the JSNS proton beam monitor was used as a trigger signal for the MPA4T module. At the same time, signals from the dynode of the NaI(Tl) detector were amplified, shaped and then fed into an analog-to-digital converter (ADC) for pulse-height measurement. However, strong γ-ray burst from the neutron source after the spallation reaction, this is known as gamma flash. These gamma flash events do not only induce noise in the TOF spectra but also saturate ADC modules for a significant period of time. Faster data acquisition is needed in such energy region. Hence, along with the pulse-height measurement, the pulse-width (PW) calculated from the time difference between the rising and the falling edges of the anode signal was recorded.

2. *Samples*

A 200 mg sample of ^{237}Np was used for the measurements. The sample consisted of 227 mg of neptunium dioxide (NpO_2) powder together with 624.5 mg of Al powder. The isotopic purity of ^{237}Np for the sample was 99.99%. The powders were packed into an Al pellet with a 20 mm diameter and 0.4 mm thick walls. A dummy container with the same measurements was also used for a background measurement. The incident neutron spectrum was reconstructed using γ-rays from the ^{197}Au(n, γ) reaction with a 20 mm in diameter and 1 mm in thickness gold sample and, also, using the 478 keV γ-rays from the ^{10}B(n, α)^{7}Li reaction with a boron sample containing enriched ^{10}B up to 90% and having a diameter of 10 mm and a thickness of 0.5 mm. Background events due to scattered neutrons were derived using a ^{nat}C sample with a 10 mm diameter and 0.5 mm thickness.

III. Data Analysis

1. *Pulse Width to Pulse Height Conversion*

Since pulse-width and pulse-height data were simultaneously recorded for all measured γ-ray, a conversion relation can be established by plotting both recorded data in a two-dimension histogram. More information about the pulse width analysis is described by Katabuchi et al [?].

2. *Background Removal*

In order to isolate the detected events coming from the ^{237}Np(n,γ)^{238}Np reaction, different measurements and techniques where applied to the recorded data from the ^{237}Np sample measurement run.

A dead time correction was applied offline to all measurements in order to estimate the count loss in the experiment [?]. The main cause for this count loss was the pile-up of two consecutive signals.

Capture events induced from slow neutrons coming from previous neutron bursts have to be subtracted. Every proton event induces neutron events with a frame length of 40 ms, as the proton beam repetition is 25 Hz. Overlapping background can be estimated using the operation pattern of J-PARC. A small part of the proton beam pulses from the 3-GeV synchrotron are injected into the 50-GeV synchrotron ring instead of JSNS. When no proton is shot into the JSNS, the measurable TOF is doubled since there is no trigger signal. Thus, the overlap background was estimated from the recorded events from 40 ms to 80 ms.

Blank background is subtracted using the data retrieved from a measurement with no sample. Likewise, the background events induced due to scattered neutrons at the sample and the events induced by the sample case are removed using the ^{nat}C and the TOF spectra obtained from the aluminum case respectively.

3. *Neutron Spectrum*

The neutron spectrum was estimated using the gold and boron samples. The obtained TOF spectrum from both runs was divided by the reaction rate simulated using the PHITS program [**?**]. Figure **??** shows a good agreement of the incident neutron distribution between the two samples except for the resolved resonance region of gold. Hence, for better performance, only the neutron spectrum from the boron measurement was used in the analysis.

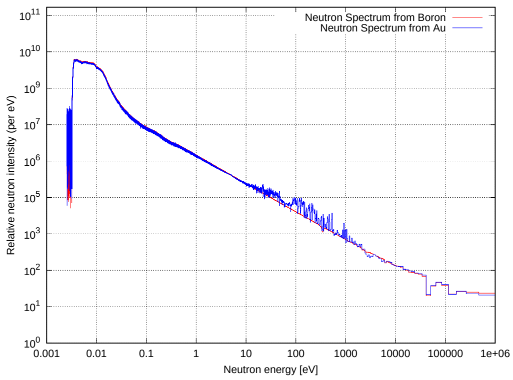

Figure 1: *Incident neutron spectrum calculated from the gold and boron samples. Over 10 eV the neutron spectrum from the boron measurement offers better results.*

4. *Data Normalization*

A relative capture cross-section can be obtained from the derived neutron capture yield and the the incident neutron spectrum. However, since the measurement times and reaction rates are different for the Np and the boron samples, the obtained energy dependence of the cross-section has to be normalized.

The first normalization process consisted on using the evaluated data from JENDL-4.0 for the whole shape of the first resonance, from 0.25 eV to 0.7 eV to normalize the cross-section results. The best fit for the normalization was obtained by minimizing the following residual:

$$\rho = \sum_{i=1}^{n} [y_i - N \cdot x_i]^2 \tag{1}$$

being n the number of energy points from 0.25 to 0.7 eV, y_i the evaluated cross-section values from JENDL-4.0, x_i the experimental results and N the normalization factor.

Alongside this method, the experimental data was also normalized using the saturated resonance technique to assess the reliability of the results. The gold sample used in the experiments was thick enough for the first resonance to be completely saturated. That means that all neutrons incoming with the energy of ^{197}Au(n,γ) first resonance (4.9 eV) react with the sample. This assumption can be seen in figure **??**. However, since the experimental setup has a detection threshold of 920 keV, a correction has to be made for the capture yield loss due to this threshold. The capture γ-ray spectrum for the ^{197}Au(n,γ)^{198}Au and the ^{237}Np(n,γ)^{238}Np reaction were calculated with CCONE at the energy of the ^{197}Au(n,γ)^{198}Au first resonance. The capture yield loss for Au was estimated as the total energy loss, namely the total γ-ray emitted under the 920 keV threshold, divided by the total excitation energy.

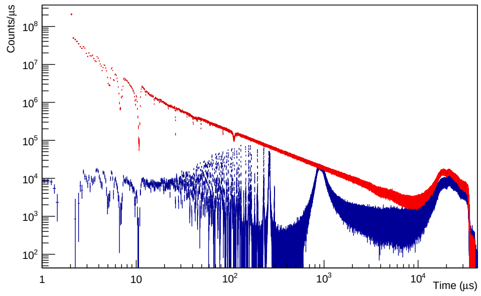

Figure 2: *^{197}Au neutron capture yield (blue) and indicent neutron flux (red).*

This correction for the saturated resonance method, N_{sat} can be expressed as:

$$N_{sat} = \frac{1 - \frac{E_{Np(0-920)}}{B_n^{Np}} / \frac{E_{Np(tot)}}{B_n^{Np}}}{1 - \frac{E_{Au(0-920)}}{B_n^{Au}} / \frac{E_{Au(tot)}}{B_n^{Au}}} \cdot \frac{S_{Np}}{S_{Au}} \cdot \frac{1}{t_{Np}} \tag{2}$$

being $E_{Np(0-920)}$ and $E_{Np(tot)}$ the energy sum of the capture γ-rays from 0 to 920 keV and total emitted, B_n^{Au} and B_n^{Np} the binding energies for ^{198}Au and ^{238}Np respectively. S_{Np} and S_{Au} are the proton bursts shot during the Np and Au measurements and t_{Np} the Np sample thickness in at/b.

5. *Uncertanty analysis*

In this experiment, alongside the statistical uncertainties related to the counting rate, several systematic uncertainties have been considered. These systematic uncertainties relate to the normalization, dead-time correction, frame overlap subtraction, self-shielding and multiple scattering corrections, neutron flux calculation.

The total error can be seen in figure **??** for both normalization techniques. The first resonance normalization offers better results as the normalization error is only of 0.5 %, where the error for the sat resonance normalization is of 2.54%. The results with the first resonance normalization present uncertainties below 3% from 0.5 to about 2 keV and maintain a value of about 4% up to 25 keV. From this energy point, the effects of the neutron scattering induced by ^{27}Al are visible as they reduce the incident neutron flux.

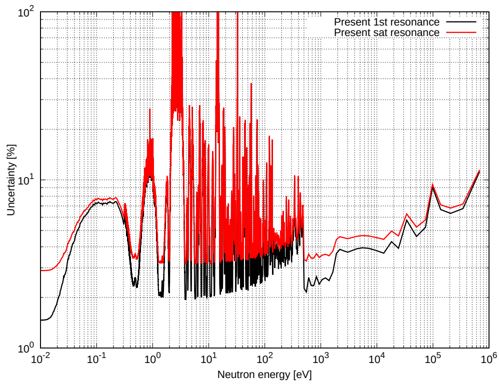

Figure 3: *Total uncertainty.*

IV. Cross-section Results

The neutron capture cross section obtained from both normalization techniques can be seen in figure **??**. The results from both normalization agree within uncertainties and offer good overall agreement with the evaluated data from JENDL-4.0.

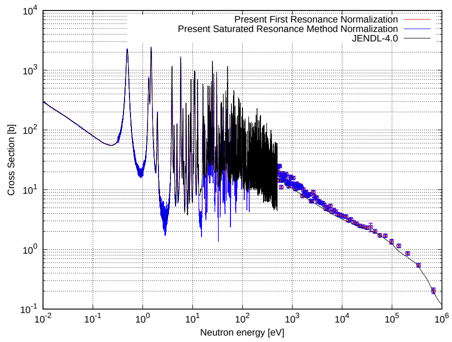

Figure 4: *^{237}Np neutron capture cross-section from 10 meV to 500 keV using the first resonance (red) and the saturated resonance method (blue) for normalization*

In the region of interest, from 0.5 to 500 keV, only the results form the first resonance normalization have been considered as they provide less uncertainty.

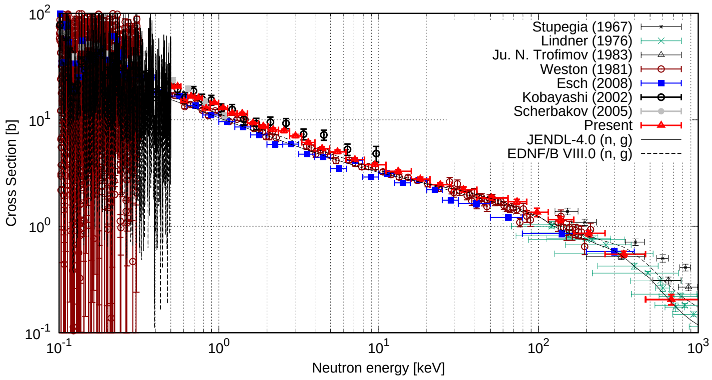

Figure 5: *Neutron capture cross-section of ^{237}Np in the region of interest from 0.5 to 500 keV.*

Figure **??** shows a comparison with previously reported experimental data in the high energy region. As can be seen, the present data holds better agreement with the experimental data by Weston [**?**] than with the data from Esch [**?**]. In the energy range below 20 keV, where the discrepancies with JENDL-4.0 were over 15%, data from Esch [**?**] provides even lower values than those from JENDL-4.0, specially between energies of 1 to 10 keV where the differences with the data by Esch amount to 25%. Over 100 keV, only activation data by Lindner [**?**] is consistent with the present data as activation data from Trofimov [**?**] and Stupegia [**?**] overestimate both the present data and that of Lindner and also evaluated data from JENDL-4.0 and ENDF/B VIII.0.

V. Theoretical Calculations

The cross-section results of the ^{237}Np neutron capture cross-section were evaluated by means of theoretical calculations using the CCONE code [**?**], Since discrepancies exist with the current JENDL-4.0 data library, the aim of this analysis is not only to assess the reliability of the results but to provide a new evaluation for the neutron capture cross-section of ^{237}Np.

The ^{237}Np neutron capture cross-section was derived by changing the E1 transition shape. For the rest of the calculations, the same parametrization as in the JENDL-4.0 were used from 0.5 keV to 1 MeV.

The final results for all reaction channels are shown in figure **??** together with evaluated results from JENDL-4.0. The present evaluation shows good agreement for the fission and inelastic channels with JENDL-4.0. For the ^{237}Np neutron capture cross-section, the results of the present evaluation provide better agreement with the experimental results in the whole energy range of the calculations.

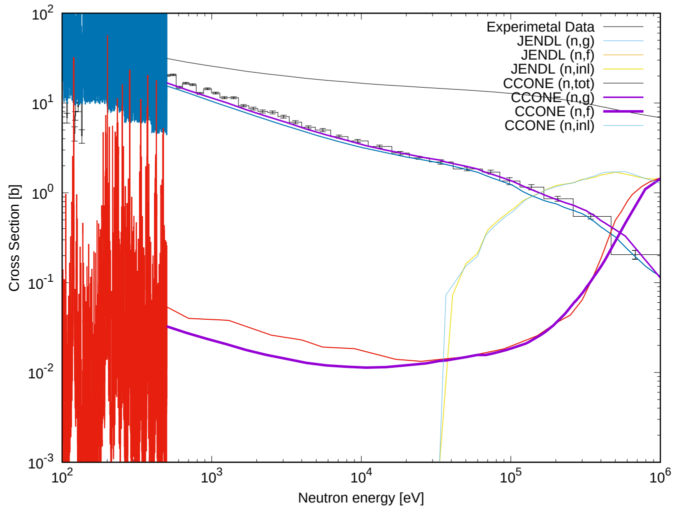

Figure 6: *Evaluated ^{237}Np neutron capture cross-section results from the CCONE calculation*

VI. Conclusions

The neutron capture cross-section of ^{237}Np was measured with incident neutrons ranging from 10 meV to 500 keV. A time-of flight method was employed using the NaI(Tl) spectrometer of the ANNRI beamline at J-PARC. In the high energy region, the cross-section results were obtained with errors of 4% or lower below 25 keV. However, over that energy the error increases to over 8% due to an increase of the statistical uncertainty. Nonetheless, for most of the energy region, the results offer a much lower total uncertainty than the error included in the JENDL-4.0 evaluated data library of 6-10%. In comparison with experimental data, the present data holds better agreement with the experimental data by Weston [**?**] as the results from Esch [**?**] underestimate the present results. Theoretical calculations were performed with the CCONE code to reproduce the experimental results. The new calculations provided by the present analysis offer better agreement with the experimental data than JENDL-4.0, specially in the region from 0.5 to 20 keV where the differences between JENDL-4.0 and the present experimental amounted to 10-25%.

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