Measurement of the neutron capture cross-section of ^{237}Np using ANNRI at MLF/J-PARC

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Abstract. The neutron capture-cross section of ^{237}Np was measured using the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) with special emphasis in the region of interest for the core design of Accelerator-Driven Systems (ADS), from 0.5 to 500 keV. A neutron time-of-flight method was employed using the NaI(Tl) spectrometer in the ANNRI beamline at the Japanese Proton Accelerator Research Complex (J-PARC) together with the pulse-height weighting technique. The cross-section was determined by normalizing the results to JENDL-4.0 cross-section data at the first resonance of ^{237}Np. In the 0.5 to 500 keV range, the present preliminary results present better agreement with previous experiments of Weston *et al* [3]. Experimental data from Esch *et al* [6] is about 15% lower than the present results. In comparision with evaluated data, ENDF/B-VIII.0 offers better agreement from 0.5 to 10 keV than with JENDL-4.0. From 0.5 to 10 keV JENDL-4.0 underestimates the present results by 10-15%. Nontheless, over 10 keV energy JENDL-4.0 shows good agreement up to 500 keV. The present preliminary cross-section has uncertainties of about 5% from 0.5 to 35 keV, a value lower than the uncertainties present in JENDL-4.0 of 6-10%. However, over 35 keV the total uncertainties steadily increase and amount to 10% at 500 keV.

1 Introduction

Nuclear transmutation has been widely established as a solution in order to reduce the long term accumulation of the high-level component of the nuclear waste. High-level waste (HLW) includes minor actinides (MA) and long-lived fission products (LLFP). Currently available nuclear data are not suitable for the final designs of the neutron transmutation systems. More improvement is needed in terms of reduction of the present total uncertainties[1].

^{237}Np is the most abundant MAs present in HLW and also one of the main component of the Accelerator-Driven Systems (ADS) core, a sub-critical facility for nuclear transmutation. The region of interest for the core design is from 0.5 to 500 keV, where JENDL-4.0 [2] includes uncertainties for the capture cross-section of ^{237}Np from 6% up to 10%. Current uncertainties in the evaluated nuclear data are an important contributor to the ADS criticality uncertainty. Hence, it is of utmost importance to precisely determine the neutron capture cross section at such energy range in order to reduce the uncertainties.

Even though the neutron capture cross-section for ^{237}Np has been extensively reported in previous experiments, the available experimental data in the region of interest, from 0.5 keV to 500 keV, is scarce. There are four sets of data using time-of-flight (TOF) method that cover the mentioned region of interest, those of Weston *et al* [3], Esch *et al* [6], Kobayashi *et al* [16] and Shcherbakov *et al* [5]. However, only the reported data from Weston *et al* and Esch *et al* cover the whole region from 0.5 keV to 500 keV and they present divergences from 15% to 35%. In the 100-500 keV range, activation-method experimental data has been published but they differ from each other up to about 30-40% [7–10].

In this work, preliminary results of the neutron capture cross section for ^{237}Np are presented with special emphasis in the region of interest for the core design of ADS, from 0.5 keV to 500 keV. Details of the experimental setup and the data analysis are also provided.

2 Experimental Setup

The experiments were conducted at the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) from the Materials and Life Science Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC). An intense pulsed neutron beam was produced by the Japansese Spallation Neutron Source (JSNS) at MLF using the 3 GeV proton beam of the J-PARC facility. The repetition rate of the accelerator was 25 Hz and the proton beam power was 400 kW.

A NaI(Tl) spectrometer was employed which consisted of two NaI(Tl) detectors at angles of 90° and 125°. However, in order to produce the final results only the detected events measured with the 90° detector were used. The sample was positioned at a neutron flight path of 27.9

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m and a TOF method was employed. In this experiment, a FAST ComTec MPA4T multi-event time digitizer was utilized for fast data acquisition purposes. This module digitizes the time between a starting trigger signal, namely the signal coming from the JSNS as the incident proton impacts the spallation target, and successive multiple stop events, i.e. the measured capture γ-rays. The energy of the detected γ-rays was measured using traditional pulse-height method with the signal coming from the dynode of the photomultiplier tubes (PMT) of the NaI(Tl) detector. Simultaniously, in order to diminish the effect of the strong γ-rays emitted from the spallation reaction on the detected events in the keV region, the signal from the anode of the PMT was employed for pulse-width measurement to also determine the energy on the γ-rays. More information about this methodology can be found in the work of Kabtabuchi *et al* [11].

A 200 mg (5 MBq) ^{237}Np sample of 20 mm diameter and 1 mm thickness was utilized for the experiments. The ^{237}Np sample consisted of 227 mg of neptunium dioxide (NpO_2) powder binded with 624.5 mg of Al powder. The isotopic purity of ^{237}Np for the sample was 99.99%. The powders were packed into a 30 mm diameter Al pellet with 0.4 mm thick walls. An exact replica of the Al dummy container was used to derive the background induced by the Al case. The energy-dependance of the incident neutron flux was derived by gating the 478 keV gamma-rays emitted from the ^{10}B(n, $\alpha\gamma$)^{7}Li reaction. For this purpose, a 90% enriched ^{10}B sample of 10 mm diameter and 0.5 mm thickness was measured. Background events due to scattered neutrons were derived using a ^{nat}C sample with 10 mm diameter and 0.5 mm thickness. A ^{197}Au sample was employed as a standard sample for the neutron capture cross-section measurement.

3 Data Analysis

3.1 Pulse-width to pulse-height conversion

A correlation between the measured pulse-width and the pulse-height can be established using the values recorded for each gamma-ray. This conversion curve is essential as only the TOF-PW spectrum contains correct measurements of the gamma-rays for events induced by neutrons coming to the sample with a time-of-flight below 15 μs. This accounts for neutrons with an energy of about 25 keV and higher. Hence, the pulse-height spectrum is reconstructed using the conversion curve from the pulse-width measurement with correct values for the γ-rays measured within the first 15 μs after the spallation reaction. Further details of this procces is explained in [11].

3.2 Capture Yield

The pulse-height weighting technique (PHWT) was applied in order to derive the neutron capture yield [12]. A weighting function was derived using the detector response function which was calculated using the simulation code SG [13]. The ^{237}Np capture yield was obtained by subtracting the contributions from the Al case and the neutron scattered at the sample. The latter was derived from the ^{nat}C sample measurement using the following normalization:

$$Y_{Np}(E_n) = Y'_{Np}(E_n) - Y_C(E_n)\frac{\sigma^{ela}_{Np}(E_n)\frac{m_{Np}}{M_{Np}}}{\sigma^{ela}_{C}(E_n)\frac{m_C}{M_C}} \tag{1}$$

where Y_{Np} is the neutron capture yield from the ^{237}Np sample, Y'_{Np} and $Y_C(E_n)$ are TOF spectrum from the ^{237}Np sample (after the Al dummy case background subtraction) and the carbon sample respectively. $\sigma^{ela}(E_n)$, M and m are the elastic cross-section, the atomic mass and the sample mass seen by the beam for both ^{237}Np and carbon. Since the neutron beam had a diameter of 13 mm at the sample position, for the ^{237}Np that had a diameter of 20 mm, only 49% of the sample is seen by the neutron beam.

In addition, the capture yield was corrected for the dead-time count loss and for the self-shielding and multiple scattering effects using simulations with the PHITS code [14].

3.3 Neutron Flux

The energy-dependence of the incident neutron flux was derived using the ^{10}B and ^{197}Au measurements. In the case of the boron sample, the measured net TOF spectrum was gated for the 478 keV gamma-rays. This gated specutrum was divided by the reaction rate for the ^{10}B(n, $\alpha\gamma$)^{7}Li reaction. Likewise, the net TOF spectrum from the ^{197}Au sample was divided by the reaction rate for the ^{197}Au(n, γ)^{198}Au reaction. Both reaction rates were obtained from simulations of the measurements with the PHITS simulation code in which the sample characteristics were taken into account. The neutron flux derived from both measurements is shown in Fig. 1. As it can be seen, the neutron flux derived from the gold sample measeurement presents a resonance structure from the radiative cature over 10 eV. Hence, only the energy dependance obtained from the boron measurement was used in order to produce the cross-section results.

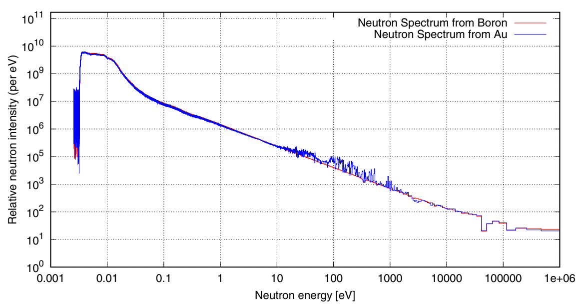

Figure 1. Energy dependance of the incident neutron flux derived from the boron measurement (red) and the Au measurement (blue).

3.4 Cross-section calculation

The derived neutron capture yield from the PWHT was divided by the energy dependence of the neutron flux in order to obtain a relative capture cross-section. For better results, the neutron flux obtained from the boron measurement was used. The absolute value for the cross-section was determined by normalizing the relative cross-section to the evaluated value for the full shape of the first resonance (0.49 eV) of JENDL-4.0 from 0.25 to 0.7 eV. This is the same procedure used in past TOF experiments of Weston *et al* [3] and Esch *et al* [6] as there is no steady value for the thermal cross-section between evaluated libraries. This procedure can be formalized in the following equation:

$$\sigma_{Np}(E_n) = N\frac{Y_{Np}(E_n)C(E_n)}{\phi_n(E_n)} \quad (2)$$

where $Y_{Np}(E_n)$ is the neutron capture yield, $C(E_n)$ is the energy dependent correction factor for self-shielding and multiple scattering, $\phi_n(E_n)$ is the incident neutron spectrum and N is the normalization factor using the first resonance of JENDL-4.0.

4 Results and Discussion

Preliminary results for the capture cross-section of ^{237}Np were derived with errors of about 5% from 0.5 keV to 35 keV. Over that energy, due to the contribution of the neutrons reacting with all the Al present in the beam-line and in the Al dummy case, the couting rate is severely reduced. Thus, from 35 keV the total uncertainty steadily increases up to 10% at 500 keV as a consequence of an augment in the statistical uncertainty.

The preliminary results of neutron capture cross-section for ^{237}Np from 0.5 to 500 keV are shown in Figure 2 together with previously reported experimental data and evaluated data from JENDL-4.0 and ENDF/B-VIII.0 [15] for comparison purposes.

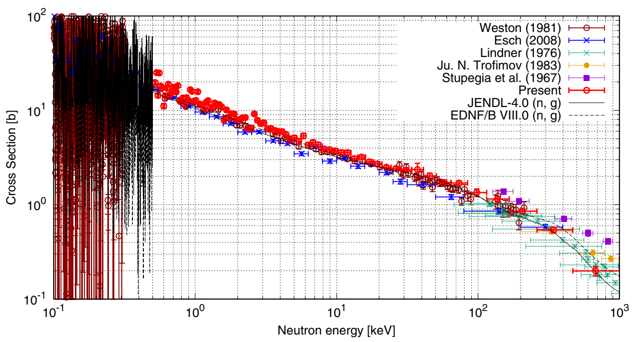

Figure 2. Neutron capture cross-section of ^{237}Np in the region of interest from 0.5 to 500 keV.

The preliminary results seem to agree more with the experimental data from Weston *et al* [3]. Experimental results from Esch *et al* [6] seem to underestimate the present results by about 15%.

In comparison with evaluated data, the present results are in reasonable agreement with the recomended values from ENDF/B-VIII.0, in particular from 0.5 to 100 keV. JENDL-4.0 underestimates our results by 10-15% from 0.5 to 10 keV, but provides fairly good agreement from 10 to 500 keV.

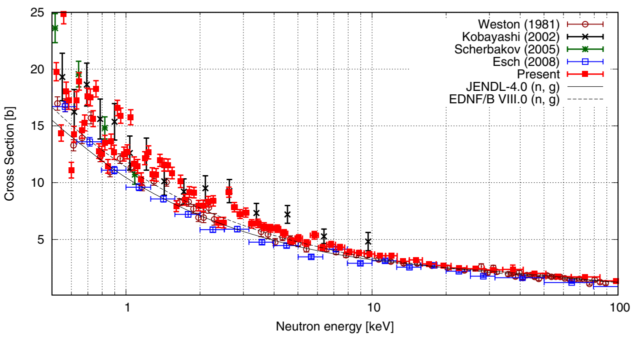

Figure 3. Unresolved resonance region of the neutron capture cross-section of ^{237}Np in more detail.

In Figure 3, the unresolved resonance region is portrayed up to 100 keV. The present preliminary results show what might resemble a resonance structure from 0.5 to 3 keV. The present results show good overall agreement within uncertainites with previously reported data from Weston *et al*, Kobayashi *et al* [16] and Scherbakov *et al* [17]. Results from Esch *et al* [6] are about 15% lower than the present results.

5 Conclusions

The ^{237}Np neutron capture cross-section was measured the Accurate Neutron-Nucleus Reaction Measurement Instrument at the Japan Proton Accelerator Research Complex with special emphasis in the region of interest from 0.5 to 500 keV. A relative neutron capture cross was dertermined using pulse-width analysis alongside pulse-height weighting technique. The obtained relative cross-section was normalized to the first resonance value of the JENDL-4.0. The present preliminary results agree within uncertainties with previous experiments of Weston *et al* [3], Kobayashi *et al* [16] and Scherbakov *et al* [17] but results from Esch *et al* [6] differ about 15%. As regards to evaluated data, ENDF/B-VIII.0 offers better agreement, in particular from 0.5 to 10 keV as JENDL-4.0 underestimates the present results by 10-15% in that energy range. Nonetheless, above 10 keV the preliminary results show good agreement with JENDL-4.0 up to 500 keV. The preliminary results present uncertainties of about 5% from 0.5 to 35 keV. This value is lower than the current uncertainties of JENDL-4.0 of 6-10%. However, over 35 keV the tota uncertainties steadily increase to 10% at 500 keV.

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