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読者の広場

「新博士誕生」 My Doctoral Research and Life Experience in Japan

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1. Introduction

The need for more accurate nuclear data for neutron-induced reactions on minor actinides (MAs) has been quantified in these decades. The reason for this is the high radiotoxicity and long half-lives that MAs present. Through many years of nuclear reactor operation, MAs have long accumulated constituting a major part of high-level nuclear waste (HLW). In order to diminish the current amount of MAs, nuclear transmutation in which fissionable radioisotopes with long half-lives, namely MAs, are converted into short-lived or even stable nuclides via neutron-induced reactions is regarded as the most viable option. Currently available evaluated nuclear data from libraries such as JENDL-4.0 can be used for the early stages of the design of nuclear transmutation systems. However, the core designs and safety measures of transmutation facilities require much more precise nuclear data for MAs. A reduction of the neutron capture cross-section uncertainty is particularly important for criticality assessments [1].

^{237}Np has a long half-life of 2.144 x 10^6 years and it produces an intense α-emitter of ^{238}Pu by neutron capture and a subsequent β decay. Moreover, ^{237}Np is the most abundant MA present in the core of Accelerator-Driven System (ADS), a sub-critical facility for nuclear transmutation, and is responsible for the most of the neutron captures as the capture reaction competes against fission reaction in the operational energy range of ADS facilities. Thus, it is of utmost importance to precisely characterize the neutron capture cross-section of ^{237}Np from 0.5 to 500 keV. At that energy, JENDL-4.0 presents uncertainties that range from 6% to 10%, much higher than the demands of 2-3%.

Following, a brief summary of my doctoral research is presented which consisted on the measurement of the neutron capture cross-section of ^{237}Np.

2. Experimental Setup

The experiments were conducted using the NaI(Tl) spectrometer of the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) beam-line at the Japan Proton Accelerator Research Complex (J-PARC). An intense pulsed neutron beam was produced by the Japanese Spallation Neutron Source (JSNS) using the 3 GeV proton beam of the J-PARC facility. In the present experiment, a time-of-flight (TOF) method was employed with a flight path of 27.9 m up to the sample position. Emitted capture γ-rays from the ^{237}Np sample were detected by the NaI(Tl) detector. The measured capture events were sequentially stored with its corresponding TOF value from the incident neutron and the energy of the detected γ-ray. The neutron capture yield was derived by applying the pulse-height weighting technique [2].

A multi-event time digitizer FAST ComTec MPA4T [3] was employed with the goal of fast data acquisition. The incident neutron energy was determined from the time between the starting spallation trigger event and successive multiple stop from the detected γ-rays. The signal coming from the JSNS proton beam monitor was used as a trigger signal for the MPA4T module. The energy of the detected γ-rays was measured using the pulse-height method with the signal coming from the dynode of the photomultiplier tubes (PMT) from the NaI(Tl) detector. Simultaniously, in order to cut down 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 of the γ-rays. More information on the pulse width measuring technique employed at ANNRI can be found in Ref. [4].

A 200 mg (5 MBq) ^{237}Np sample with a 20 mm diameter and a 1 mm thickness was employed for the experiments. The sample was packed into a 30 mm diameter Al pellet with 0.4 mm thick walls. An exact replica of the Al container was used as a dummy container to derive the background induced by the Al case. 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% with 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.

3. Resonance Analysis

The neutron capture resonances of ^{237}Np in the resolved resonance region were analyzed using the least-square fitting code `REFIT` [5]. The initial resonance parametrization data expressed by resonance energy E_λ, neutron width $\Gamma_{\lambda,n}$, radiation width $\Gamma_{\lambda,\gamma}$ and fission width $\Gamma_{\lambda,f}$ along with the values for spin and parity for each resonance were taken from JENDL-4.0 [6]. The fitted parameters were used for the analysis of the ^{237}Np neutron capture resonance peaks from the first resonance (0.49 eV) to 110 eV. An averaged value for radiation width $\Gamma_{\lambda,\gamma}$ was established from 17 resonances under 22 eV of $\langle\Gamma_\gamma\rangle = 40.3 \pm 0.5$ meV. Examples of the fitting process are shown in Figs. 1 and 2. The fitted parameters for all the resonances can be seen in Ref. [7].

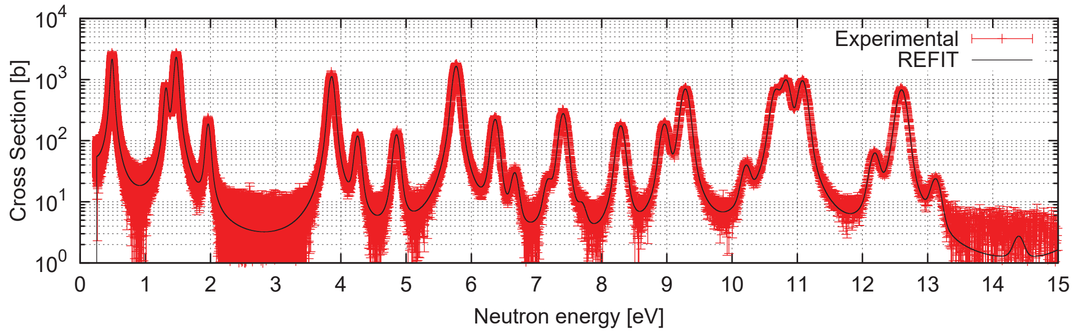

Fig. 1 (color) n + ^{237}Np resonance fitting with `REFIT` from 0.2 to 15 eV.

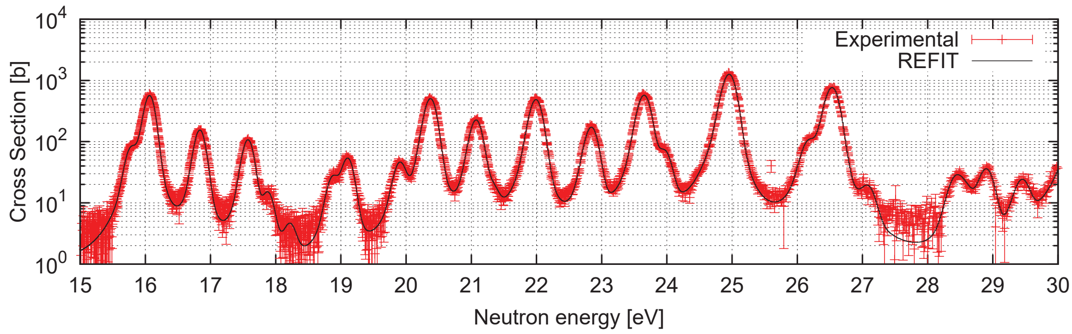

Fig. 2 (color) n + ^{237}Np resonance fitting with `REFIT` from 15 to 30 eV.

By means of an statistical approach, the properties of the nuclear levels in the compound nucleus through neutron-nucleus interactions can be derived from a neutron resonance analysis. The distribution of two consecutive levels or so called level spacing and the neutron strength function were derived to be $\langle D_0 \rangle = 0.60 \pm 0.02$ eV and $10^4 S_0 = 1.02 \pm 0.12$ respectively.

4. Results and Evaluation

The experimental results were normalized using two normalization techniques. The first method 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. Together with this method, the experimental data were also normalized using the saturated resonance technique. The gold sample used in the experiments was thick enough for the first resonance to be completely saturated. Since the resonance is saturated, the total netron flux can be determined as all neutrons incoming with the energy of ^{197}Au(n,γ) first resonance (4.9 eV) interact with the sample. 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 yield loss for Au was estimated from calculations using the CCONE code [8] as the total energy loss, namely the total γ-ray emitted under the 920 keV threshold, divided by the total excitation energy. This correction for the saturated resonance method, N_{sat} can be expressed as:

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

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

The absolute cross-section obtained from both normalization can be seen in Fig. 3. There is good agreement within 2% between both normalizations.

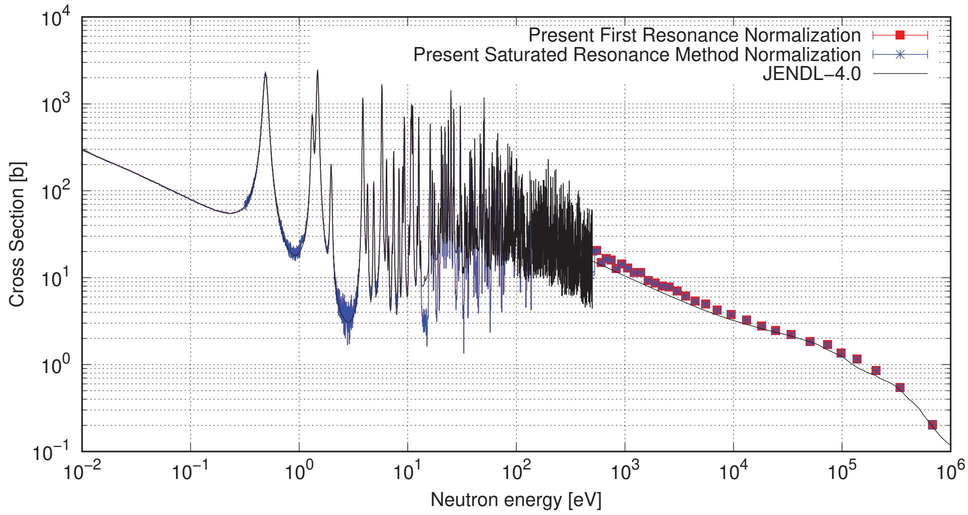

Fig. 3 ^{237}Np neutron capture cross-section from 10 meV to ! MeV using the first resonance (red) and the saturated resonance method (blue) for normalization.

For the thermal cross-section, values of 177.6 $\pm$ 4.2 b and 176.2 $\pm$ 6.2 b were obtained with the first resonance and the saturated resonance normalization processes respectively. Weston *et al* [9], Esch *et al* [10] and Hirose *et al* [11] provide experimental results that agree within 2% with values for the thermal cross section of 180 $\pm$ 6 b , 177 $\pm$ 5 b and 176.7 $\pm$ 5.2 b respectively. Moreover, JENDL-4.0 (178.08 b) [6] and ENDF/B.VIII.0 (175.43 b)[12] evaluated nuclear data libraries also present similar values. However, these values differ much from the thermal cross-sections obtained through activation method experiments.

In the high energy region, only the cross-section data from the first resonance normalization was considered as it provided lower uncertainties than those from the saturated resonance method. The present results contain uncertaintes of 4% or less from 0.5 to 25 keV. However, over that energy the error increases with the incident neutron energy and amounts over 8% at 500 keV. Nonetheless, from 0.5 to 25 keV the present errors are much lower than the current uncertainty of JENDL-4.0 of 6-10%.

Figure 4 shows a comparison with previously reported experimental data in the high energy region and with the recommended values of JENDL-4.0 and ENDF/B-VIII.0. As can be seen, the present data holds better agreement with the experimental data by Weston *et al* [9] than with the data from Esch *et al* [10]. In the energy range from 0.5 to 20 keV, where JENDL-4.0 underestimates the present results by over 10%, data from Esch *et al* [10] provides even lower values than those from JENDL-4.0. In comparison with evaluated data libraries, there is a 10-25% discrepancy with JENDL-4.0 data from 0.5 to 20 keV. However, over that energy there is good agreement within uncertainties up to 500 keV with the recommended values by JENDL-4.0. The agreement with ENDF/B VIII.0 is better from 0.5 to 20 keV as it provides a higher value than JENDL-4.0.

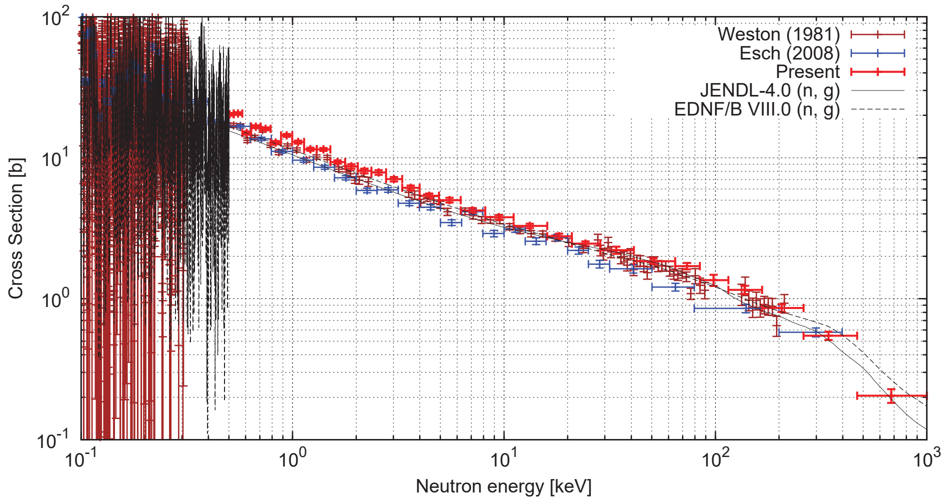

Fig. 4 Neutron capture cross-section of ^{237}Np in the region of interest from 0.5 to 1000 keV.

In the last part of the doctoral thesis, the cross-section results were evaluated by means of theoretical calculations using the CCONE code. The neutron capture cross-section for ^{237}Np was derived using mostly the same parametrization as provided in the JENDL-4.0 file. The only difference was that the parameters derived from the resonance analysis were used as input instead of the reference values provided by the RIPL-3 library. The final fit was obtained by changing the gamma strength function for the E1 transition. The other channels open in the same energy range, namely the fission and inelastic channel, were also calculated using the information present in the JENDL-4.0 file. The present calculation results are presented in Fig. 5 alongside the present experimental results and the values of JENDL-4.0. The present evaluation provides better agreement with the experimental results in the whole energy range of the calculations.

5. Conclusion

The neutron capture cross-section of ^{237}Np was measured with incident neutrons ranging from 10 meV to 500 keV. The absolute cross-section was determined by normalizing the results to JENDL-4.0 cross-section data at the first resonance of ^{237}Np and using the saturated resonance method using a ^{197}Au sample measurement in which the first resonance was completely saturated.

The resolved resonance region was analyzed with the REFIT code to derive the resonance parameters for the resonances up to 110 eV. An average radiation width $\langle \Gamma_\gamma \rangle = 40.3 \pm 0.5$ meV, a mean level spacing $D_0 = 0.60 \pm 0.02$ eV and a neutron strength function $10^4 S_0 = 1.02 \pm 0.12$ were obtained.

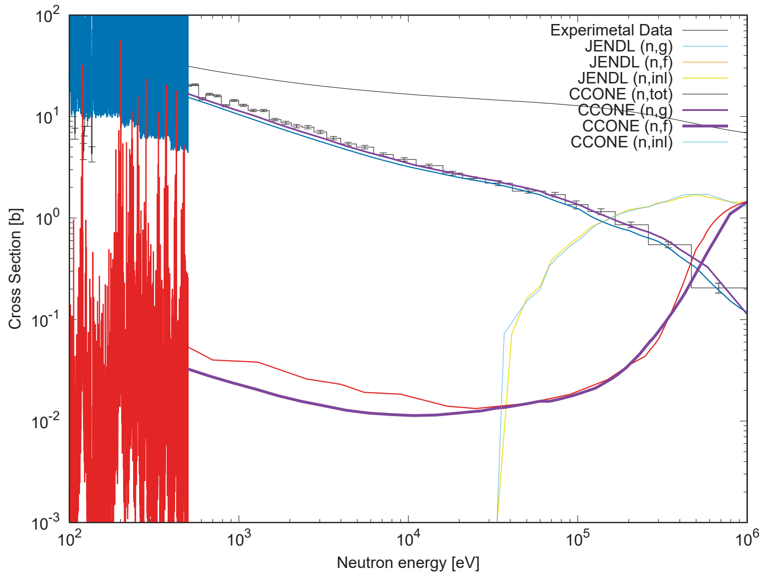

Fig. 5 CCONE calculation results for the different channels open in the 0.1 to 1 MeV energy range compared with JENDL-4.0 and the present experimental data.

The thermal cross-section was measured to be 177.6 ± 4.4 b and 176.2 ± 6.2 b with the first resonance and the saturated resonance normalization processes respectively. 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. In comparison with experimental data, the present data holds better agreement with the experimental data by Weston [9] as the results from Esch [10] underestimate the present results, specially between energies of 1 to 10 keV where the differences with the data by Esch amount to 25%.

The results in the high energy region were evaluated using theoretical calculations with the CCONE code to reproduce the experimental results. The evaluation provided by the present analysis offers 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%.

6. Life in Japan

My first experience in Japan was in May 2015 when I became an intern at the Nuclear Data Center at JAEA. Coming from Barcelona, life at Tokai was very different for me and a bit difficult at the beginning as I was used to living in a big city. However, I really enjoyed working in a professional environment at JAEA and I thrived thanks, in great part, to the people at the Nuclear Data Center.

My experience in 2015 was great and I was determined to come back to Japan to pursue my doctoral degree. Fortunately, I was accepted by professor Tatsuya Katabuchi at the Tokyo Institute of Technology from April 2017. From here on started what I can only describe as an amazing experience that allowed not only for my professional growth but for my personal growth as well.

I started living with my girlfriend, who I met in 2015, and really enjoyed the lifestyle of Tokyo as it also resembles the style of Barcelona. I also relished the atmosphere at Tokyo Tech. I think that the laboratory system is a very successful way to ensure proper learning through tutoring by the professors. Unfortunately, this system is not common in Spain.

Moreover, as soon as I arrived to Tokyo I managed to get to know the local Catalan comunity. We regularly host events and celebrate catalan festivities such is the one you can see in Fig. 6. This allowed me to stay in touch with my local culture and tradition while I was submerging into Japanese culture. Also, as much as I love japanese food I still needed to eat local Spanish and Catalan food. Luckily, I found two good restaurants with real local Spanish food. The first one is called ”El Castellano”, エル・カステリャーノ, near Shibuya which is the oldest Spanish restaurant in Tokyo. The second is my favourite and is called ”Fonda Sant Jordi”, フォンダ・サン・ジョルディ, in Ebisu. This restaurant allowed me to eat real local Catalan food and paella whenever I want. I really recommend it if anybody is interested.

After three years, I leave behind a great deal of friends and collegues after I completed my doctoral degree.

I have recently moved back to Tokai and I stared working as a postdoctoral reseacher in the Nuclear Data Center at JAEA. This time will be a little bit different as I am not alone. I recently got married in March after my graduation. I really look forward to the next years, who knows how many, at JAEA. Hopefully, it will continue to be a great experience.

Fig. 6 Catalan house event in Feburary 2018

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