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Absolute and relative cross section measurements of ^{237}Np(n,f) and ^{238}U(n,f) at the National Physical Laboratory

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Abstract. Cross section measurements in the fast energy region are being demanded as one of the key ingredients for modelling Generation-IV nuclear power plants. However, in facilities where there are no time-of-flight possibilities or it is not convenient to use them, using the ^{235}U(n,f) cross section as a benchmark would require a careful knowledge of the room scatter in the experimental area. In this paper we present measurements of two threshold reactions, ^{238}U(n,f) and ^{237}Np(n,f), that could become a standard between their fission threshold and 2.5 MeV, if the discrepancies shown in the evaluations and in some experimental data can be solved. The preliminary results are in agreement with the present ENDF/B-VII.1 evaluation.

1. Introduction

The need for better cross section values to improve accuracy when modelling Gen-IV nuclear power plants has been highlighted recently by a sensitivity analysis [1]. Experiments on fission cross sections are generally performed relative to ^{235}U(n,f). At fast neutron energies fission threshold isotopes (e.g. ^{238}U, ^{237}Np) might be more suitable for benchmarking. However, major concerns have been raised recently about the neutron-induced fission cross sections of ^{238}U and ^{237}Np. For the first isotope, ^{238}U, even though considered a secondary standard above 2 MeV the related uncertainties are not negligible and different libraries (i.e. ENDF/B-VII.1 and JEFF-3.2) show discrepancies of up to 7% in the 1.5 MeV $< E_n <$ 3 MeV range. In the case of ^{237}Np recent experiments [2] measured the cross section to be up to 5% higher in the fast energy region.

A study of the ^{238}U(n,f) and ^{237}Np(n,f) cross sections was performed at the Van de Graaff accelerator of the UK National Physical Laboratory (NPL) in collaboration with the Joint Research Centre under the EC-FP7 CHANDA project. A twin-Frisch grid ionization chamber (TFGIC) was used as a fission fragment detector and a well-characterized long counter was employed to measure the absolute fluence. The ^{235}U(n,f) cross section was also measured.

2. Experimental setup

A twin Frisch-grid ionization chamber (TFGIC) was used as a fission fragment (FF) detector. The TFGIC consisted of 5 electrodes: 2 anodes, 2 grids and a cathode; and was filled with CH_4 and operated at a pressure of 1052 mbar. The samples were positioned in a sandwich with two plates acting as a cathode. The current from each electrode was fed into an independent charge sensitive preamplifier. The output signal of each preamplifier was then input into a 250MHz 14 bit waveform digitizer and saved for offline analysis. All channels were synchronized through a time card and had a common trigger, derived from the cathode signal [3].

All three samples used during this experiment were produced at JRC-Geel either by vacuum deposition (^{235}U) or by molecular plating (^{238}U and ^{237}Np) (Table 1) on solid backings. Two samples were positioned back-to-back between the cathode plates; each of them facing one side of the TFGIC and, thus, dividing the volume into two. This allowed us to perform two measurements of the (n,f) cross section simultaneously: an absolute and a relative one.

3. Measurements

The experimental work was supported by the EC-FP7 CHANDA project. A two week experimental campaign was carried out in January 2016 at the 3.5 MV Van de Graaff accelerator of NPL. The facility is well-known for being the national standard for absolute neutron measurements from thermal neutron energies up to 16.5 MeV, among other capabilities [4]. The beam lines reach the centre of a low-scatter experimental area (18 m × 18 m × 26 m) making this facility suitable for experiments that are sensitive to scattered neutrons. Two neutron producing targets were employed to cover neutron energies from 0.5 MeV up to 2.4 MeV (see Table 2).

For neutron energies in the fast region (0.1 MeV $<$ E_n $<$ 16.5 MeV) the facility uses a well-characterized long counter to measure the absolute neutron fluence [5]. The standard procedure is to measure the neutron fluence at the start of each day or every time that the neutron producing target or the proton energy is changed. For that two measurements are performed: one with the long counter at ca. 2 m from the neutron producing target and another with a shadow cone in between to subtract the scattered

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Table 1. Properties of the samples used during the experiment.

Isotope	Mass (μg)	Purity
^{235}U	562±2.0%	99.83%
^{238}U	681±2.6%	>99.99%
^{237}Np	489.5±0.5%	>99.99%

Table 2. Neutron producing reactions used, energies and FFs detected from each isotope.

		FFs detected		
Reaction	E_n (MeV)	^{235}U	^{238}U	^{237}Np
^{7}Li(p,n)^{7}Be	0.567	5687	–	2587
^{3}H(p,n)^{3}He	1.2	3169	–	3122
^{3}H(p,n)^{3}He	1.8	3950	–	4274
		8228	3271	–
		–	4876	12583
^{3}H(p,n)^{3}He	2.0	3597	–	3746
		8242	3672	–
		–	5988	14324
^{3}H(p,n)^{3}He	2.4	9770	4615	–
		–	5891	13614

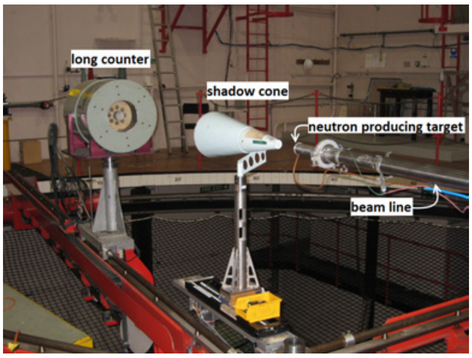

Figure 1. NPL low-scatter area with the long counter and shadow cone aligned with the beam line.

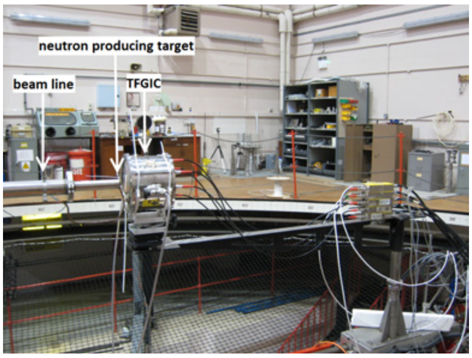

Figure 2. TFGIC aligned with the beam line and neutron producing target.

neutron contribution (see Fig. 1). This absolute neutron fluence is related to a current integrator monitor placed on the neutron producing target can. Then, the TFGIC is placed in front of the neutron producing target, ideally at ca. 1.5 m, but in this occasion it was placed as close as possible in order to maximize the neutron flux impinging on the samples (see Fig. 2). During the measurements with the TFGIC the current integrator monitor was used to derive the absolute neutron fluence on the target deposits.

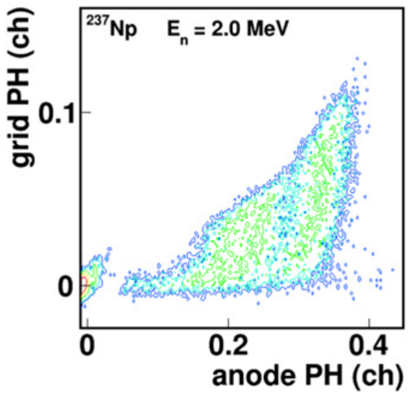

Figure 3. 2D pulse height spectrum obtained for the ^{237}Np at an incident neutron energy of 2 MeV.

4. Data analysis

As mentioned in Sect. 2, two measurements were performed simultaneously: an absolute and a relative one. The analysis consisted of two steps: treatment and quantification of the fission fragment (FF) signals (common for both measurements) and the absolute neutron fluence determination (only relevant for the absolute one).

4.1. Fission fragment determination

The signals stored using the waveform digitizer were analysed offline using digital signal processing software developed at JRC-Geel. The signals were treated with a baseline correction and a RC^4 filter to obtain their pulse height (PH). The analysis of both the grid and the anode signals allowed the quality of the samples being used to be verified as well as to trace back signals with a PH which was not well understood [6]. A typical 2D grid versus anode PH plot is presented in Fig. 3 for ^{237}Np at an incident neutron energy of 2 MeV. In the figure the separation at low PH values between α-particles, recorded when the trigger corresponded to the other sample, and the low energy FFs is clear.

The amount of FFs detected are corrected for the fraction of FFs with kinetic energy below the electronic threshold (1–3%) and for the fraction of FFs emitted at grazing angles and stopped within the sample deposit ($< 2\%$).

4.2. Neutron fluence at the sample positions

The determination of the absolute neutron fluence impinging on the samples is usually done with a code developed specifically for the needs of the facility. The program assumes that the detector is at a sufficient distance from the neutron producing target to apply a point-to-point calculation. However, in the case of this experiment the samples within the TFGIC were at about 8 cm from the neutron producing target. Therefore, some corrections need to be applied to account for the different neutron fluence rates and energies emitted from the neutron target into the different solid angles formed by the samples. For this reason an MCNP model of the experimental setup has been constructed and simulations were carried out. The source definition of the simulations have been obtained with the Neusdesc software [7].

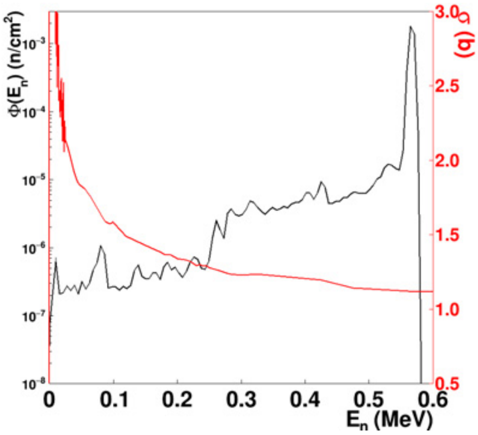

Figure 4. Neutron fluence at the sample position (black line) and (n,f) cross section of ^{235}U (red line) (see text for further information).

Additionally, the scattered neutrons in the experimental area, although expected to be only a small percentage of the total, are expected to have a non-negligible influence when impinging on the samples. This is especially true in the case of ^{235}U because of its higher (n,f) cross section at low energies. An example of the MCNP simulated neutron spectrum at the sample positions for an incoming neutron energy of 0.567 MeV, due to direct and scattered neutrons, is shown in Fig. 4 together with the ^{235}U(n,f) cross section.

The scattering of the neutron producing target can and the attenuation due to the front face of the TFGIC as well as the anode and grid facing the beam have also been calculated using MCNP.

4.3. Absolute measurements

The cross section calculation when considering the absolute measurement is as follows:

$$\sigma(E_n)=\frac{C_{\mathrm{corr}}\cdot k_{\mathrm{FF,low}}}{\epsilon}\frac{A}{m\cdot N_A}\frac{1}{\Phi_n(E_n)\cdot k_{\mathrm{PP-DD}}\cdot k_{\mathrm{TS}}\cdot k_{\mathrm{AttFC}}}, \quad (1)$$

where C_{corr} are the corrected counts for the electronic threshold, ϵ accounts for the FF loss within the sample, A is the atomic number of the sample, m its mass, N_A is Avogadro's number and $\Phi_n(E_n)$ is the absolute fluence. The correction factor $k_{\mathrm{FF,low}}$ accounts for the fission fragments produced by neutrons of lower energy than the main peak, $k_{\mathrm{PP-DD}}$ corrects for the point-to-point to disk-to-disk difference when extracting the absolute fluence, k_{TS} corrects for the neutrons scattered in the target can and k_{AttFC} corrects for the attenuation of neutrons in the front face of the TFGIC as well as the anode and grid facing the neutron beam. Results presented in Sect. 5 are preliminary and a full uncertainty budget still needs to be calculated.

4.4. Relative measurements

The calculation of the cross section when considering a reference sample is done by:

$$\sigma_i(E_n)=\frac{C_{i,\mathrm{corr}}\cdot k_{\mathrm{FF,low}}\cdot\epsilon_{ref}\cdot A_i\cdot m_{ref}}{C_{ref,\mathrm{corr}}\cdot\epsilon_i\cdot A_{ref}\cdot m_i}\sigma_{ref}(E_n), \quad (2)$$

where the subscript i refers to either the ^{235,238}U or ^{237}Np, the subscript ref to the reference sample considered and $\sigma_{ref}(E_n)$ the (n,f) cross section at the neutron energy evaluated for the reference sample according to the ENDF/B-VII.1 evaluation.

5. Preliminary results and discussion

The preliminary results are presented in Fig. 5 for the absolute measurement (top row) and the relative one (bottom row). For all measurements the uncertainties are only related to statistics, mass, efficiency and counts below electronic threshold. A full uncertainty budget will be presented in a future publication.

The derived ^{235}U(n,f) cross section is around 5–7% higher than the evaluations when considering both measurements. There could be several reasons for this discrepancy: an inaccurate description of the room scatter in the MCNP calculation, a possible problem with the ^{235}U sample, etc. This discrepancy highlights the difficulty of making both absolute and relative measurements with ^{235}U in such a facility. Further investigations of this issue will be carried out.

In the case of the cross section of ^{237}Np the agreement between experimental data and the evaluations is within 2–3% in the plateau region. The value on the fission threshold is slightly lower than the evaluated files, although within uncertainties. Therefore, these preliminary results are in agreement with the present evaluations and do not confirm the larger cross section seen by Ref. [2].

The results for the neutron-induced cross section of ^{238}U are in very good agreement with the ENDF/B-VII.1 evaluation, although there are limited statistics in this experiment. Thus the JEFF 3.2 evaluation for this isotope between 2.0 MeV and 3 MeV might need to be revised.

The lower values seen in the measurements of ^{237}Np and ^{238}U relative to ^{235}U are again an indication of a possible issue either with the corrections applied to the ^{235}U data or with the sample itself.

6. Conclusions

The neutron-induced cross sections of two isotopes with a fission threshold, ^{238}U and ^{237}Np, have been studied in order to solve discrepancies between some evaluated data files and recent experimental data. The experiment shows that in addition to ^{235}U(n,f) other standards are required when performing experiments at Van de Graaff accelerators without time-of-flight possibilities, for accurate cross section results.

The preliminary results shown here for the ^{238}U(n,f) and ^{237}Np(n,f) are in rather good agreement with present evaluations, especially ENDF/B-VII.1, and do not confirm the increase between 2.0 and 2.5 MeV of the ^{238}U(n,f) cross section derived by the JEFF 3.2 evaluation, or the 5% increase seen by Paradela et al. [2] of the ^{237}Np(n,f) cross section. The results for ^{235}U(n,f) need further studies and highlight the difficulties of measuring this isotope in this kind of facility. A full uncertainty budget will be described in a future publication.

The experimental techniques presented in this paper are very well-known by the community, however, to be able to reduce uncertainties to below the 2–3% level further developments need to be made.

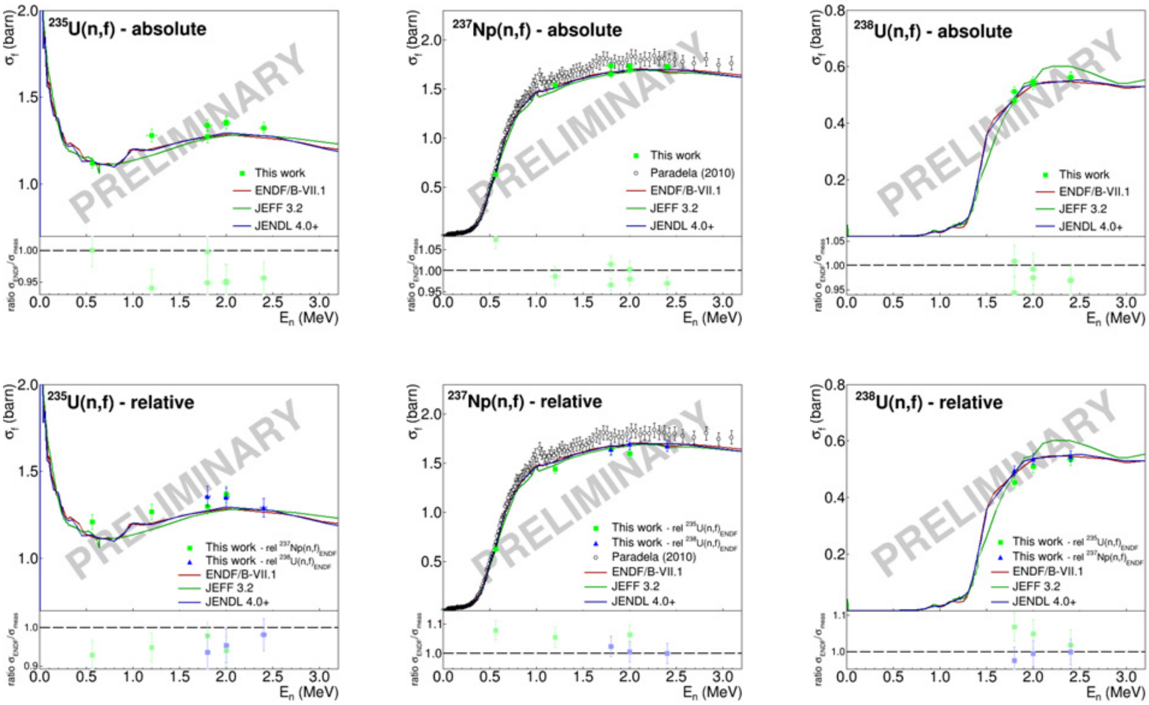

Figure 5. Preliminary results obtained via the absolute measurement (upper row) and the relative one (lower row) for the, from left to right, ^{235}U, ^{237}Np and ^{238}U. The data is compared to the ENDF/B-VII.1, the JEFF 3.2 and the JENDL 4.0+ evaluation. In addition, the ^{237}Np is compared to the experimental data from Paradela et al. [2]. For each case, the ratio between the measured data and the ENDF/B-VII.1 evaluation is also shown.

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