

# Absolute cross section measurements of $^{238}\text{U}(n,f)$ and $^{237}\text{Np}(n,f)$ in the neutron energy range 1-2.4 MeV

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**Abstract.** New standard (n,f) cross sections other than  $^{235}\text{U}$  are important to study the relevant cross sections for Generation-IV power plants. A specific need for such standards is for performing new experiments with quasi-monoenergetic neutron beams, such as those produced by Van de Graaf accelerators. Neutrons down-scattered to low energies in the experimental environment, so called room-return, become relevant for this type of measurements. Hence, a standard (n,f) cross section with a fission threshold is of great interest, in order to suppress the contribution from room-return background. For this reason we have performed two experiments at the VDG of the National Physical Laboratory to measure absolutely the (n,f) cross sections of  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{237}\text{Np}$  in the fast neutron energy region. Our preliminary results are in agreement with the most up-to-date evaluations.

## 1 Introduction

Modelling of Generation-IV nuclear power plants requires highly accurate values of cross sections in the fast energy region. Cross section measurements are usually performed relative to the primary standard  $^{235}\text{U}(n,f)$ , but in environments where the thermal and epi-thermal neutron background is non-negligible, such as the target hall of a Van de Graaff accelerator, other isotopes with a fission threshold should be preferred. Up to now, none of the isotopes with a fission threshold has been considered a primary standard. Two isotopes, for which the experimental database is sufficient, could potentially fill this role:  $^{238}\text{U}(n,f)$  and  $^{237}\text{Np}(n,f)$ .  $^{238}\text{U}(n,f)$  has a fission threshold at around  $E_n = 1.6$  MeV and is a secondary standard from  $E_n = 2$  MeV. Although the JEFF 3.2 evaluation showed discrepancies up to 7% in the range  $1.5 \text{ MeV} < E_n < 5 \text{ MeV}$  with respect to ENDF/B-VIII.0, the new JEFF 3.3 evaluation is in agreement with ENDF/B-VIII.0.  $^{237}\text{Np}(n,f)$  would be more suitable as a standard because of its lower fission threshold ( $E_n = 0.5$  MeV) and its higher cross section above  $E_n = 1.0$  MeV, but some discrepancies between measurements have been found in the last years [1].

To address these issues, two experimental campaigns have been performed at the Van de Graaff accelerator of the National Physical Laboratory (NPL,UK) under the European CHANDA project. The Nuclear Metrology group at NPL is known worldwide for its capability to provide very accurate and precise neutron flux measurements by using a well characterized long counter. Within their facilities there is also a large low-scatter target hall ideal for

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

Isotope	Sample ID	Mass ( $\mu\text{g}$ )	Purity
$^{235}\text{U}$	$^{235}\text{U}_{2016}$	562 $\pm 2.0\%$	99.83%
$^{235}\text{U}$	$^{235}\text{U}_{2017}$	701 $\pm 0.6\%$	99.93%
$^{238}\text{U}$	$^{238}\text{U}$	681 $\pm 2.6\%$	>99.99%
$^{237}\text{Np}$	$^{237}\text{Np}$	489.5 $\pm 0.5\%$	>99.99%
$^{242}\text{Pu}$	$^{242}\text{Pu}$	671 $\pm 0.9\%$	>99.97%

cross section measurements. A twin Frisch-grid ionization chamber was used as fission fragment detector. Measurements were done absolutely, by using the long counter, and relatively, by placing two samples in a back-to-back configuration in the fission chamber. The first campaign was performed in January 2016 and the isotopes measured were  $^{235}\text{U}(\text{n},\text{f})$ ,  $^{238}\text{U}(\text{n},\text{f})$  and  $^{237}\text{Np}(\text{n},\text{f})$ . For the second campaign, performed in January 2017, measurements were performed for  $^{235}\text{U}(\text{n},\text{f})$ ,  $^{237}\text{Np}(\text{n},\text{f})$  and  $^{242}\text{Pu}(\text{n},\text{f})$ .

Results from both campaigns are presented, although results on the  $^{242}\text{Pu}(\text{n},\text{f})$  cross section will be reported in a future publication as they are outside the scope of the present work. This paper presents a follow-up on the experiment reported on [2].

## 2 Experimental setup

The experimental setup consisted of a long counter (LC) and a twin Frisch-grid ionization chamber (TFGIC). The measurements were done in the following chronological order: first, a LC measurement was performed with the desired neutron energy to determine the neutron fluence per monitor count. The monitor is measuring the current on the neutron producing target can produced by the incident ion beam. Then, a shadow cone was used to subtract the contribution from scattered neutrons to the neutron fluence measured by the LC. Finally, the TFGIC was placed in front of the neutron producing target.

Two proton-induced reactions were used during the experiment:  $^7\text{Li}(\text{p},\text{n})^7\text{Be}$  for producing  $E_n = 0.565$  MeV and  $^3\text{H}(\text{p},\text{n})^3\text{He}$  for producing  $E_n = 0.9\text{-}2.4$  MeV.

### 2.1 Long counter

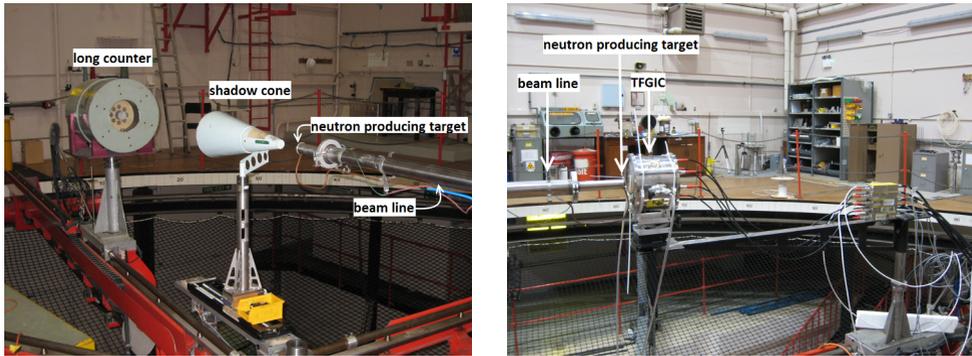
The LC consists of a  $\text{BF}_3$  tube surrounded by a moderating layer and an outer shield. It has a high sensitivity, a negligible gamma response and a nearly flat fluence response from 1 keV up to 7 MeV [3]. Figure 1 (left) shows the LC placed in front of the beam line and with the shadow cone in place.

### 2.2 Fission Fragment detector

Two different TFGICs were employed as fission fragment detector with very similar dimensions, one in 2016 and another one in 2017. For the measurements reported here the configuration was the same as in [2] (see Fig. 1 (right)).

### 2.3 Fissile samples

During both campaigns five different samples were used. Their properties are listed in table 1.



**Figure 1.** Left: NPL low-scatter area with the long counter and shadow cone aligned with the beam line. Right: TFGIC aligned with the beam line and neutron producing target.[2]

### 3 Measurements

Two measurement campaigns have been performed, one in 2016 and one in 2017. In 2016 the focus was on the ability to reproduce the  $^{235}\text{U}(n,f)$  cross section, thus proving our experimental setup and our data analysis. In addition, the  $^{238}\text{U}(n,f)$  and the  $^{237}\text{Np}(n,f)$  cross section were measured. As it will be shown in Sect. 5, the results of the 2016 measurement on the  $^{235}\text{U}(n,f)$  cross section were systematically higher than predicted by the evaluations. For this reason, the same cross section was measured in 2017 using two different samples, together with the  $^{237}\text{Np}(n,f)$  and  $^{242}\text{Pu}(n,f)$ .

### 4 Data analysis

#### 4.1 Neutron fluence at the sample positions

The neutron fluence determined by the LC needs to be corrected by several effects that are related with the fact that we could not perform a shadow cone measurement with the TFGIC. In addition, the code used to obtain the neutron fluence at the sample positions from the LC measurement treats the system as a point-to-point problem, it considers the neutron producing target as a point in the space and the fission samples as a point. However, this was no longer applicable when we measured with the TFGIC because the edge of the fission chamber was only around 3 cm away from the neutron producing target, therefore the total distance between the neutron producing target and the fission samples was only around 9 cm. For this reason MCNP6 simulations [4] were performed to allow for the neutron fluence emitted by the disk-like neutron producing target and transmitted through the disk-like samples. This correction is around 2-4% depending on the incident neutron energy.

##### 4.1.1 Target can scattering

The target can scattering was taken into account as this is subtracted in the shadow cone measurement performed with the long counter, but could not be taken into account in the TFGIC measurement due to the close distance between the TFGIC and the neutron producing target. The calculation was made using MCNP6 and the correction is of 2-3% depending on the incident neutron energy.

### 4.1.2 Neutron attenuation on the TFGIC

Finally, the attenuation of neutrons not only on the front face of the TFGIC, but on the first anode and grid, needs to be considered as these layers will be contributing to the neutron slowing down and, therefore, change the spectrum of the neutrons impinging on the samples in the cathode. The calculation was made using MCNP6 and the correction is of 1.5-2% depending on the incident neutron energy.

## 4.2 Fission fragment determination

The fission fragments were detected using the TFGIC. The signal used in our experiments corresponded to the anodes and grid only. A 2D grid vs anode pulse height was used to clean the anode pulse height (PH) distribution from any  $\alpha$ -particle background. In addition, the anode projected PH was extrapolated to lower PH values to account for FFs of lower energy than the level of the electronic threshold, this was done using a straight line. This correction is about 2-5%.

### 4.2.1 Reaction rate due to lower energy neutrons

An MCNP6 simulation was needed to simulate the target hall to account for all scattered neutrons that will be impinging on the samples with lower energy than expected. These neutrons will be inducing fissions in the samples as a function of their corresponding (n,f) cross section. Therefore, this correction will have more impact on the  $^{235}\text{U}$  samples as its cross section increases as the neutron energy decreases. The correction is of 4-10% depending on the sample and the incident neutron energy.

## 4.3 Cross section determination

The cross section has been determined as:

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

where  $C_{\text{corr}}$  are the corrected counts for the electronic threshold,  $\epsilon$  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  $k_{\text{FF,low}}$  accounts for the fission fragments produced by neutrons of lower energy than the main peak,  $k_{\text{PP-DD}}$  corrects for the point-to-point to disk-to-disk difference when extracting the absolute fluence,  $k_{\text{TS}}$  corrects for the neutrons scattered in the target can and  $k_{\text{AttFC}}$  corrects for the attenuation of neutrons in the front face of the TFGIC as well as the anode and grid facing the neutron beam.

## 5 Preliminary results and discussion

The preliminary results are presented in Fig. 2. All results presented include both statistical and systematic uncertainties. In all case, the most significant ones are the counting statistics and the uncertainty related to the neutron fluence determination at the position of the samples.

### 5.1 $^{235}\text{U}(n,f)$

The  $^{235}\text{U}(n,f)$  cross section was measured in both campaigns, 2016 and 2017. In 2016, only one sample was employed, namely  $^{235}\text{U}_{2016}$ , whilst in 2017 we measured with  $^{235}\text{U}_{2016}$  and with a new sample, namely  $^{235}\text{U}_{2017}$ . The main reason to measure with different samples was the increased cross section values found in 2016. In order to confirm that the issue was due to the sample, in 2017 we repeated the same measurement with the same sample and measured as well with another sample. The results we have obtained for the  $^{235}\text{U}(n,f)$  cross section are in very good agreement with the present evaluations when using the  $^{235}\text{U}_{2017}$  sample (red squares). However, there was clearly an issue with the  $^{235}\text{U}_{2016}$  sample (green and blue squares). This unidentified issue increased the measured cross section by around 5% in the investigated neutron energy range. Although additional experiments are planned to further study this effect, the most probable hypothesis is that the high-Z backing of the  $^{235}\text{U}_{2016}$  sample could have an impact on the amount of backscattered FFs detected.

### 5.2 $^{238}\text{U}(n,f)$

The  $^{238}\text{U}(n,f)$  absolute cross section measured in this experiment agrees within uncertainties with the present evaluations.

### 5.3 $^{237}\text{Np}(n,f)$

In the case of  $^{237}\text{Np}(n,f)$  measurements were performed in 2016 and 2017 with some neutron energies repeated, mainly due because in 2017 the  $^{242}\text{Pu}(n,f)$  cross section was measured at the same time. The  $^{237}\text{Np}(n,f)$  absolute cross section measured in this experiment agrees within uncertainties with the present evaluations and the newest data from Diakaki [5]. Therefore, does not confirm the 5% increase seen by Paradela [1]. (The ratio  $\sigma_{\text{ENDF}}/\sigma_{\text{meas}}$  is not shown as its value is out of scale.)

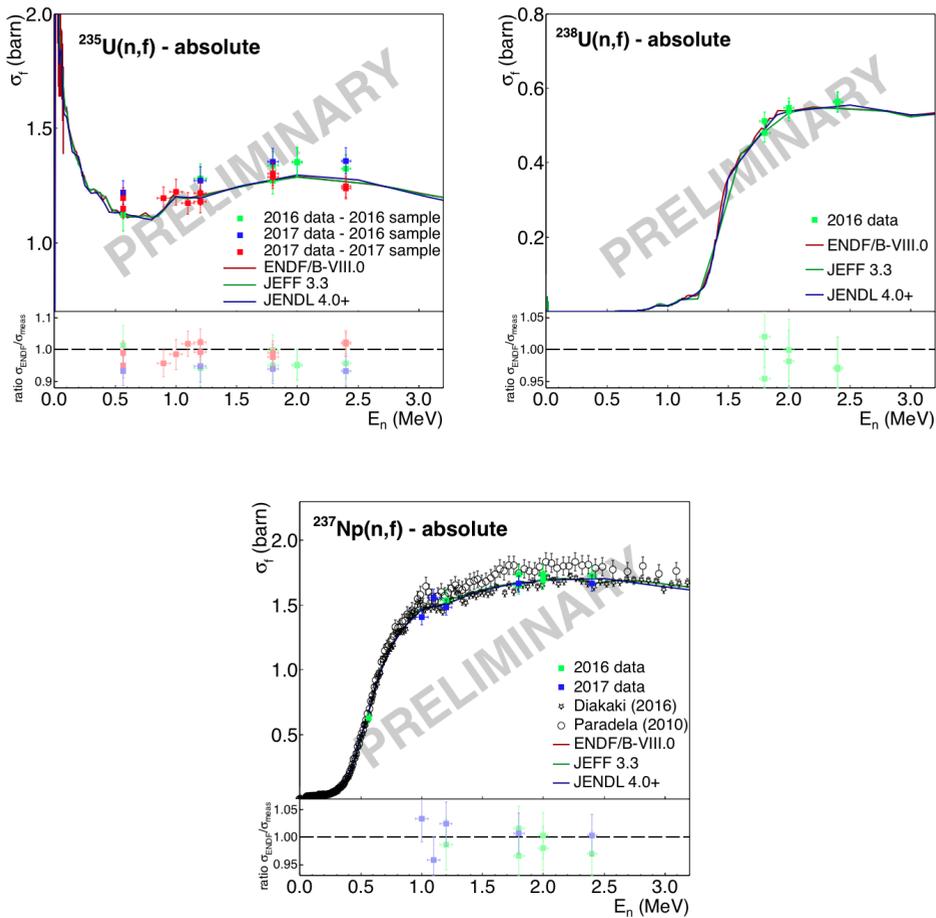
## 6 Conclusions

The neutron-induced fission cross section has been measured absolutely for  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{237}\text{Np}$ . The results show a very good agreement with the present evaluations for  $^{238}\text{U}(n,f)$  and  $^{237}\text{Np}(n,f)$ . In the case of  $^{235}\text{U}(n,f)$  our data agree with evaluation when using one of the samples ( $^{235}\text{U}_{2017}$ ), but we obtain a higher cross section when using the  $^{235}\text{U}_{2016}$  sample. However, we suspect that further investigations will be able to explain this mismatch. Our uncertainties are dominated by the statistical uncertainty due to the beam current limitation of the facility as well as the fluence uncertainty. A follow up experiment is planned to resolve the discrepancy obtained with the  $^{235}\text{U}_{2016}$  sample. Our preliminary results agree with the latest evaluations in the neutron energy ranges studied in this work and we believe that they could be used, when finalized, along with other high quality data in a possible evaluation of  $^{238}\text{U}(n,f)$  and  $^{237}\text{Np}(n,f)$  as standards in these energy ranges.

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## References

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**Figure 2.** Preliminary results obtained via the absolute measurement for the  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{237}\text{Np}$ . The data is compared to the ENDF/B-VIII.0, the JEFF 3.3 and the JENDL 4.0+ evaluation. In addition, the  $^{237}\text{Np}$  is compared to the experimental data from Paradela *et al.* [1] and Diakaki *et al.* [5]. For each case, the ratio between the measured data and the ENDF/B-VIII.0 evaluation is also shown. See text for further details.

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