### **EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH**

#### Proposal to the ISOLDE and Neutron Time-of-Flight Committee

# Neutron-induced fission cross-section of <sup>237</sup>Np obtained with two different detection systems

January 13, 2014

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#### Abstract

We propose to measure the fission cross-section of the  $^{237}Np(n,f)$  with the PPAC detection setup used in previous experiments in EAR-1 and with Micromegas detectors in EAR-2. This experiment would take advantage of the high energy resolution in EAR-1 and the high neutron flux of EAR-2 of the n\_TOF facility to solve the discrepancies in the MeV region and beyond between the current evaluations and more recent measurements. Furthermore, the experiment with the PPAC setup will also provide the fission fragment angular distribution (FFAD), extending the current knowledge beyond 15 MeV.

**Requested protons** :  $4x10^{18}$  protons on target in total:  $2x10^{18}$  (EAR-1),  $2x10^{18}$  (EAR-2) **Experimental Area** : EAR-1 and EAR-2

# 1. Introduction and motivation

The <sup>237</sup>Np isotope is abundantly produced in present nuclear reactors and is one of the major long-lived components of nuclear waste which can be considered as a potential target of incineration in fast neutron reactors. This requires a good knowledge of its neutron interaction properties above the fission threshold. In particular, the fission cross-section received a special attention in the last decade and has been measured at different facilities. The most recent one performed in a ToF facility is the n\_TOF measurement [1] which appears higher by 6% by comparison to previous measurements and evaluated files (Figure 1).



**Figure 1:** Fission cross-section ratio of  $^{237}Np$  and  $^{235}U$  data in the energy region above the threshold step. The most recent n\_TOF results [1] are plotted together with the experimental data available from the EXFOR data base.

This singularity of the first n\_TOF data could shed some doubt on its validity. However, several features still speak in their favor: i) the cross-consistency of the previous measurements and data libraries often lies on arbitrary re-normalizations [2], ii) critical experiments are better reproduced when using the n\_TOF data [3], iii) measurements of fission rates involving <sup>237</sup>Np are also often re-normalized by calibration in a reference flux, so that they are not sensitive to a global factor on the cross-section.

Moreover, after the publication of the first n\_TOF data, a pair of experiments with monoenergetic beams have been not able to resolve this discrepancy. On one hand, the results obtained by Diakaki et al. [4] with Micromegas detectors in the energy range 4.5-5.3 MeV are scattered between the n\_TOF data and the evaluation. On the other hand, an experiment performed at the JRC-IRMM Van de Graaff to determine the cross-section of <sup>240</sup>Pu and <sup>242</sup>Pu using <sup>237</sup>Np as reference seems to show better agreement with the current plutonium evaluations when using n\_TOF <sup>237</sup>Np data [5].

From this status we can conclude that the absolute fission cross-section of <sup>237</sup>Np, even when proposed at certain points as a secondary reference, is not accurate enough and a new experiment is highly desired to give a final answer.

In the meanwhile, we have developed a new detection setup based on PPAC [6] which overcomes the drawbacks suffered by the original setup used in Ref. 1. The current setup, successfully tested between 2010 and 2012 at n\_TOF, providing an accurate  $^{235}U/^{238}U$  ratio reported in Ref. 7, and the  $^{232}$ Th(n,f) fission fragment angular distribution published in Ref. 6. Moreover, a new analysis technique based on this new setup's characteristics has been developed to directly obtain the detection setup efficiency from the registered data for each target and energy region [8].

Furthermore, a new fission detection system based on Micromegas detectors [9] has been successfully used during 2011 and 2012 at n\_TOF to study the  $^{242}$ Pu(n,f) cross-section in EAR-1, as well as for the recent measurement of the  $^{240}$ Pu(n,f) reaction in EAR-2. A preliminary analysis of the latter data shows that the detectors can operate reliably in the high neutron-rate environment of EAR-2.

The combined use of both detection systems in two experimental areas with different characteristics will allow to cross-check the obtained result, minimize the systematic uncertainties and give a definitive answer to the <sup>237</sup>Np puzzle by a measurement at 3% accuracy.

# 2. Experimental setup

The proposed experiment will be performed with the same PPAC setup that has been used for the experiment n\_TOF-14 [6,10] in EAR-1 and the Micromegas-based detection setup used for the experiment proposed in CERN-INTC-2014-051 / INTC-P-418 ("*Measurement of the* <sup>240</sup>*Pu*(*n*,*f*) reaction cross-section at the CERN n\_TOF facility EAR-2") [11] in EAR-2.

### 2.1 EAR-1 setup

The EAR-1 setup consists of 10 Parallel Plate Avalanche Counters (PPAC) and 9 targets, both tilted by  $45^{\circ}$  with respect to the neutron beam in order to cover the full angular range from  $0^{\circ}$  to  $90^{\circ}$ . The fission fragments are detected in coincidence by the two PPACs surrounding each target, so the trajectories can be reconstructed and the emission angle with respect to the beam can be obtained.

We propose to include 50 mg of  $^{237}$ Np, distributed in 4 new targets deposited on thin aluminium backings of 0.7 microns in thickness and one old Neptunium target used in the 2003 campaign for cross-checking. To obtain the cross-section, as reference we will use two  $^{235}$ U targets with the same backings as the  $^{237}$ Np targets, and two  $^{238}$ U targets of about 10 mg each. The targets will be provided by IPN Orsay and the target masses will be measured by  $\alpha$ -counting to an accuracy better than 1%.

### 2.2 EAR-2 setup

The second detection setup is based on Micromegas detectors which are developed with the micro-bulk technique [12,13]. This setup was successfully tested both for monitoring the neutron beam and for the measurement of fission cross-sections in both EAR-1 and EAR-2. Microbulk detectors are composed of very thin layers of material, thus minimising neutron scattering effects.

A chamber that can house up to 10 sample-detector modules was constructed for previous measurements and is available, as well as the needed number of micro-bulks.

The chamber will again be filled with an  $Ar:CF_4:isoC_4H_{10}$  gas mixture (88:10:2), which is commonly used at CERN and has excellent timing properties due to the relatively high electron drift velocity.

New electrodeposited <sup>237</sup>Np targets will be provided by IRMM, Geel. In total, 4 targets of 60 ug/cm<sup>2</sup> each, will be needed. Both <sup>235</sup>U (2 samples of 70 ug/cm<sup>2</sup> each) and <sup>238</sup>U (340 ug/cm<sup>2</sup> each) will be used as reference targets. All deposits will have be 3 cm in diameter on a 5 cm diameter aluminium backing. The sample holder design is ready and simple to manufacture.

Finally, if possible, an extra <sup>237</sup>Np target as the ones used in the PPAC detection setup will be measured with the Micromegas detection setup, which will further reduce the systematic uncertainties related to the target-detector assembly.

In each setup (PPAC and Micromegas) we will include targets made with <sup>237</sup>Np coming from 2 different sources: CEA (followed by chemical purification) and IRMM. This will reveal any sample-related problem as, for example, the presence of any long-lived actinide like <sup>238</sup>U which can contribute to fission but is difficult to detect by  $\alpha$ -counting.

# 3. Beam request

On the basis of previous measurements, we request a total of  $2x10^{18}$  protons in the target for the PPAC experiment in EAR-1 by using the fission collimator to maximise the counting rate with our 8 cm diameter samples. The same amount of neutrons is requested for the Micromegas setup in EAR-2.

Figure 2 (left panel) shows the number of counts expected for each target of the PPAC set up for 100 bins per energy decade. The fission cross-section for <sup>237</sup>Np is always higher than that of <sup>238</sup>U, and only in the threshold region is it below that of <sup>235</sup>U. Therefore, with the proposed target load, the statistical uncertainty will be dominated by the counting rate of the reference targets above 1 MeV and by the <sup>237</sup>Np counting rate around the fission threshold and in the resonance region. We intend to reach a 1% statistical uncertainty on each target to control the target-related systematic uncertainties.



**Figure 2:** Left: Number of counts expected for the <sup>235</sup>U and <sup>237</sup>Np targets of the PPAC detectors at 100 bins per energy decade up to 1 GeV. **Right:** Total expected counts from <sup>237</sup>Np in EAR-2 at 20 bins per energy decade along with the corresponding statistical uncertainty. Due to the absence of very high-energy neutrons in EAR-2 and the effects of the  $\gamma$ -flash, an upper energy limit of around 10-20 MeV is expected.

In EAR-2, the requested beam will allow to reach statistical uncertainties below 1% above 0.4 MeV (at 20 bins per energy decade), below 3% above 0.2 MeV and below a few percent for selected sub-threshold resonances. The estimated fission counts and the corresponding statistical uncertainty are shown in Figure 2 (right panel).

## 4. Summary

Significant discrepancies in the measured fission cross-section of <sup>237</sup>Np exist among n\_TOF datasets, as well as n\_TOF data and other measurements performed elsewhere and with different techniques, especially between the first- and second-chance fission thresholds. To clarify this situation, we propose to re-measure this cross-section with two completely different setups in EAR-1 (with an improved PPAC detector assembly) and EAR-2 (with Micromegas detectors), thus minimising the overall uncertainties and discerning eventual systematic ones. The detectors and electronics have all been previously used at n\_TOF and the existing analysis software can be used with minor modifications. In conclusion, this experiment, which aims to produce high-quality results on an important cross-section and resolve discrepancies in the MeV region, can be carried out with existing equipment and with a very reasonable expected duration of about 3 to 5 weeks (for 2x10<sup>18</sup> protons), depending on the beam delivery rate.

# Summary of requested protons:

4.0x10<sup>18</sup> protons on target in total: 2x10<sup>18</sup> (EAR-1), 2x10<sup>18</sup> (EAR-2)

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