EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of the neutron-induced fission cross section of ^{236}U at n_TOF

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Abstract: We propose to measure the neutron-induced fission cross section of ^{236}U in EAR-1 and EAR-2 of the n_TOF facility using Micromegas detectors. ^{236}U ($T_{1/2}$ =2.342x10^{7} years) is produced by neutron capture in ^{235}U fuel, therefore the accurate knowledge of neutron-induced reactions for this isotope is of high importance. As a result of its production path, the ^{236}U nucleus is directly linked to the neutron economy in all types of nuclear reactors which are based on uranium fuel. The aim of this experiment is to produce a single accurate data-set covering the energy range from thermal up to 0.5 GeV neutron energy. Due to the lack of experimental data in the thermal and resonance region, the available evaluated libraries show severe discrepancies of up to two orders of magnitude. In order to provide high-accuracy cross section data for such an extended energy region, the specific characteristics of both n_TOF experimental areas (EAR-1 and EAR-2) will be exploited and high-purity ^{236}U samples will be used. The EAR-1 measurement will cover the energy region from the fission threshold up to several hundreds of MeV, while in EAR-2, given the higher instantaneous neutron flux, useful data will become available for the first time in a unique data-set spanning from the thermal region and covering the resonances up to a few hundreds of keV.

Requested protons: 9x10^{18} (6x10^{18} in EAR-1, 3x10^{18} in EAR-2)
Experimental Areas: EAR-1 and EAR-2

1 Introduction

1.1 Motivation

High-accuracy cross section data for neutron-induced reactions are needed in a wide energy range for the design, feasibility and sensitivity studies on advanced nuclear systems [1, 2]. The ^{236}U isotope, with a half-live of 2.342x10^7 years, has the longest half-life compared to any other fission product or actinide produced in nuclear reactors. Due to the fact that its specific activity (2.4 MBq/g) is about 190 times higher than the one of ^{238}U, it significantly contributes to the radioactivity of reprocessed uranium. In current reactors based on U/Pu fuel, ^{236}U is produced by neutron capture on ^{235}U and it considerably affects the neutron balance in the reactor core, as well as the fuel composition. Moreover, ^{236}U builds up in the equilibrium state in the Th/U fuel cycle. As a consequence, knowledge of its fission cross section is required within 5% accuracy for the development of fast nuclear reactors and accelerator-driven systems (ADS) [3].

1.2 Present status of data

For the ^{236}U(n,f) reaction cross section in the thermal neutron energy region, evaluated libraries, in particular JENDL-5 [5], JEFF-3.3 [6], ENDF/B-VIII-0 [7] and TENDL-2021 [8], exhibit major discrepancies with each other of up to two orders of magnitude (Fig. 1). Among these evaluations, only JENDL-5 seems to fairly reproduce the only two measured data points in the thermal region (Wagemans et al. [9, 10]). Additionally, there are only two data-sets that indicate the existence of the first resonance of ^{236}U around 5.45 eV. Data from Sarmento et al. [11], with the highest resolution in this region, reveal that current evaluations, with the exception of JENDL-5 and TENDL-2021, need revision since they are overestimating the height of the first resonance by almost 150 times. Furthermore, in the region between 30 eV and 1 keV, data that point out resonance structures by Cramer and Bergen [12] have been adopted by some evaluations, nonetheless they are not corrected for the ^{235}U impurities present in the samples as well as for the γ-sensitivity of the detectors. On top of that, these resonance structures are not confirmed by latter measurements of Alekseev et al. [13] and Sarmento et al. [11] in the same energy region. As a general remark, it seems that for the energy region below the fission threshold the bottleneck for scarce and discrepant data is actually the quality of samples in terms of impurities, mainly ^{235}U and ^{233}U, that create an additional strong fission background. In combination with the admittedly low cross section of ^{236}U, this makes the measurement of its neutron-induced fission cross section challenging.

Above 500 keV, the situation of the experimental information is much better since there are a lot of measurements that have lead to improvement of the knowledge of the cross section in the high energy region (Fig. 2). Despite that, discrepancies of up to 15% are observed among different data-sets, especially for the region of interest for the fast nuclear reactors between 1 and 10 MeV (better visible in Fig. 3 where the cross section ratio of ^{236}U and ^{235}U of some measurements is shown). Additionally, above 40 MeV (which exceeds the upper limit of most evaluations), only three data-sets that reach higher neutron energies up to a few hundreds of MeV [14, 15, 16] are available. This region is particularly important to constrain theoretical models of the fission process.

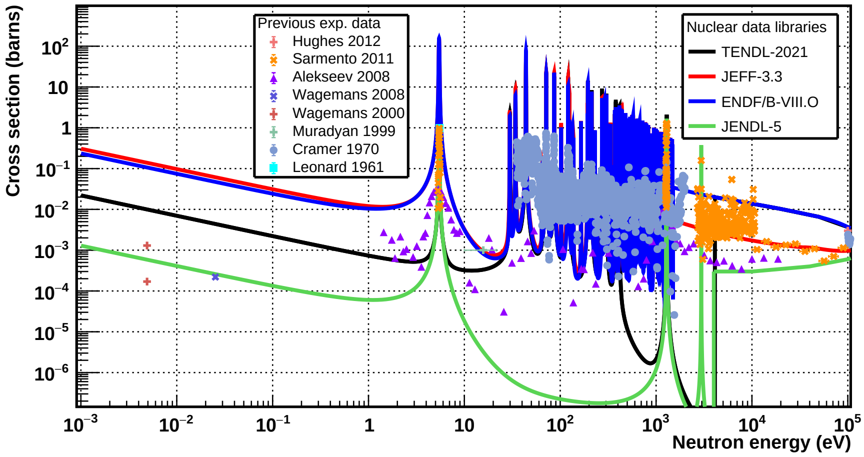


Figure 1: Comparison of available experimental fission cross section data retrieved from the EXFOR database and evaluated libraries for neutron energies between thermal and 100 keV. The error bars of the experimental data are omitted in this plot. It is evident that the experimental information for the ^{236}U(n,f) reaction in the thermal and resonance region is very poor.

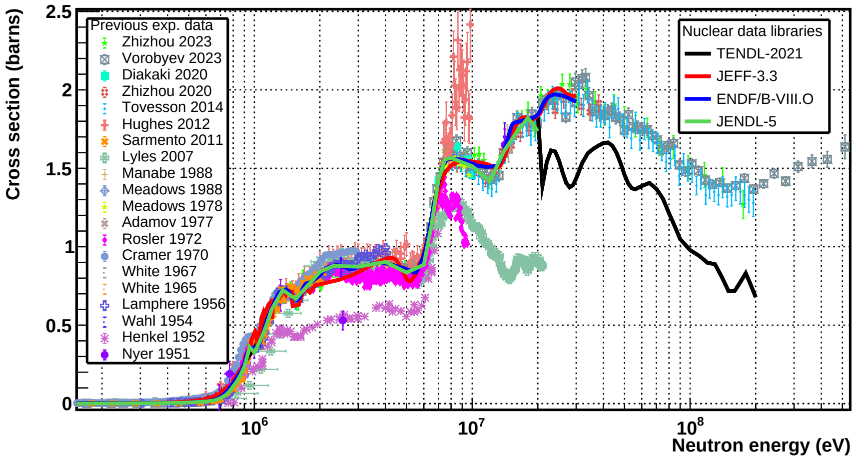


Figure 2: Comparison of neutron-induced fission cross section data of ^{236}U above 100 keV retrieved from the EXFOR database and nuclear data libraries.

1.3 Previous measurement at n_TOF and prospects for the new measurement

A measurement of the ^{236}U(n,f) cross-section was already performed at n_TOF in 2003 (EAR-1) during Phase I [11]. In this experiment, the fission yield of the ^{236}U(n,f) reaction was affected by the contribution of α-particles from the ^{236}U decay but most importantly from fission events originated from the 0.05% impurity of ^{235}U present in the samples. After the subtraction of the ^{235}U contamination, the analysis revealed resonance structures attributed to ^{236}U at 5.45 eV and at 1.25 keV (in the form of a triple resonance). Above the fission threshold, the data were

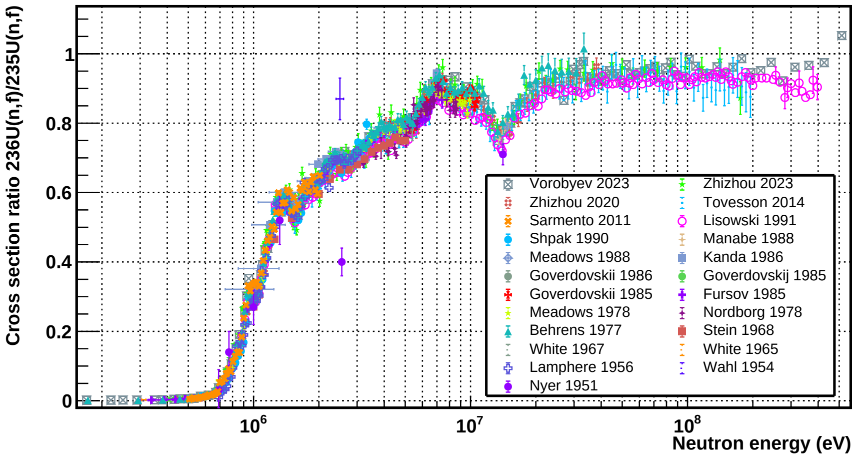


Figure 3: Fission cross section ratio of ^{236}U to ^{235}U based on the available experimental data above 100 keV retrieved from the EXFOR database. Discrepancies up to 15% are observed among different data-sets above 2 MeV.

limited only up to 2 MeV.

Accordingly, we propose a new measurement to address the discrepancies in the existing data and to expand the experimental information in the as yet unexplored thermal region, as well as to complement the data in the high energy region above the fission threshold. In order to achieve better data quality in the low energy region and measure the thermal cross section of the ^{236}U(n,f) reaction, we plan to use high purity ^{236}U samples with $\sim$ 12 times less ^{235}U impurity (^{235}U: 0.0043%). By performing the first part of the measurement in EAR-2, we aim in collecting useful data in the thermal and resonance region, taking advantage of the higher instantaneous neutron flux and the suppressed signal-to-background ratio due to the shorter flight path of 19 m from the lead spallation target. For energies higher than 700 keV in EAR-2 we expect to have limitations in the data analysis, mainly due to pile-up corrections and to the superposition of fission pulses on the γ-flash signal. In EAR-1, high-resolution and high-accuracy n_TOF data can be provided for neutron energies starting from threshold (around 300 keV) and up to $\sim$500 MeV.

2 Experimental setup

2.1 Samples

We plan to employ the same high-purity ^{236}U samples used by Wagemans et al. [9, 10] for the determination of the cross section in the thermal point. These two samples (^{236}U: 99.9732%, ^{234}U: <0.00001%, ^{235}U: 0.0043%, ^{238}U: 0.0225%), the characteristics of which are listed in Table 1, were produced by electrodeposition at JRC-Geel (Belgium). In addition, ^{235}U, ^{238}U and ^{10}B samples will be used as reference for the neutron flux determination. Due to the fact that the ^{236}U and the reference samples have different diameters, we plan to use aluminium "masks" with a diameter of 40 mm, matching the diameter of the smallest deposit. This step is instrumental in order to perform a relative fission cross section measurement since the same neutron beam interception factor (BIF) for all samples will lead to reduced systematic uncertainties in the analysis.

Table 1: Main characteristics of the ^{236}U samples. Effective quantities refer to the unmasked part of the target (i.e. to a 40 mm diameter).

Isotope	Areal density (μg/cm^2)	Diameter (mm)	Activity (kBq)	Effective activity (kBq)	Effective mass (mg)
^{236}U	210	50	9.9	6.3	2.64
^{236}U	132	40	4.0	4.0	1.66

2.2 Micromegas detectors

For neutron measurements it is of particular importance to minimise the amount of material present in the beam in order to reduce the background related to scattered neutrons as well as to avoid the perturbation of the neutron flux. For this reason, the microbulk design [17, 18] was developed based on the Micromegas principle (Fig. 4). This design has already been utilised at n_TOF for neutron-induced fission cross section measurements of ^{242}Pu [19], ^{240}Pu [20], ^{237}Np [21], ^{241}Am [22], ^{230}Th [23] and, most recently, ^{243}Am [24].

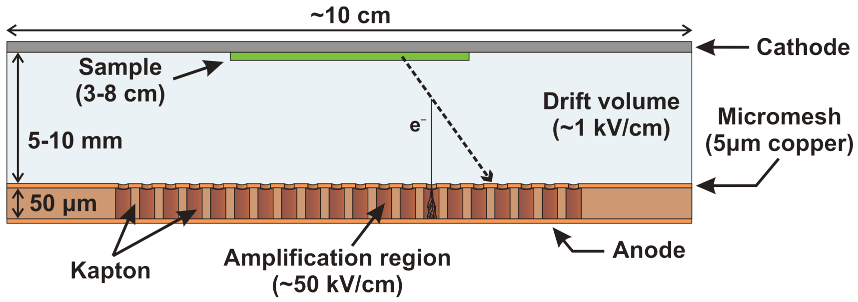


Figure 4: An illustration of the basic principle of operation of a Micromegas detector. An ionising particle emitted from a sample ionises the gas. The ionisation electrons drift towards the micromesh and are multiplied inside the high-field amplification region before being collected on the anode. Indicative values are given for the electrical field and dimensions of the two regions.

An aluminium chamber will be used to house the sample-detector modules. Within the chamber a continuous flow of a properly chosen gas mixture will be maintained, held at atmospheric pressure. More precisely, in EAR-1 it is planned to use an Ar:CF_4:isoC_4H_{10} (88:10:2) mixture which exhibits excellent timing characteristics due to its relatively high electron drift velocity, whereas in EAR-2, in order to minimise the elastic interactions with the hydrogen inside the drift region of the detector, it is proposed to use an Ar:CF_4 (90:10) mixture.

2.3 Electronics and data acquisition

A setup based on existing electronics from previous fission measurements, consisting of custom-made pre-amplifiers (INFN-Bari), will be used for fast signal shaping. Incremental improvements have been made over the past few years in the design of the pre-amplifiers, resulting in a significant reduction of post γ-flash baseline oscillations and in the enhancement of the signal-to-noise ratio. The output of the pre-amplifiers will be directed to the standard n_TOF Data Acquisition System based on flash-ADCs.

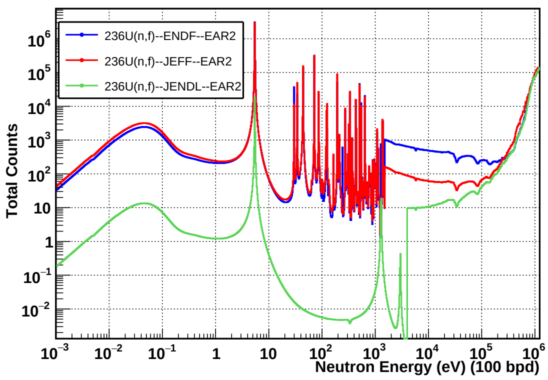
3 Beam request

By using the large collimator in both experimental areas, i.e. 6 cm diameter in EAR-2 and 8 cm diameter in EAR-1, we can profit from increased neutron flux. The upper and lower limit for the total reaction rate for EAR-2, employing different evaluations (JENDL-5, JEFF-3.3, ENDF/B-VIII-0) and assuming 3x10^{18} protons is shown in Fig. 5. In EAR-2 we can acquire experimental data in the neutron energy range from thermal up to 700 keV. Moreover, by adopting a coarser energy binning (e.g 10 bpd) in the thermal region we can further reduce the statistical uncertainty in this regime which was previously unexplored. At the same time, at around 5.45 eV we can provide a better mapping of the first resonance of ^{236}U which dominates the fission cross section below the threshold. It has to be mentioned that even if our measurement confirms the JENDL-5 evaluation limiting the precision of the measurement in the thermal part, the contribution of the first resonance of ^{236}U at 5.45 eV will account almost 90% of the total fission yield up to 100 keV and 99% up to 1 keV. Furthermore, by assuming 6x10^{18} protons for EAR-1 (Fig. 6), we can provide experimental fission cross section data from the threshold around 300 keV up to hundreds of MeV with a statistical uncertainty below 2% even at 100 bpd. In all the above mentioned reaction rate calculations, the n_TOF-Phase IV neutron flux was considered for both experimental areas.

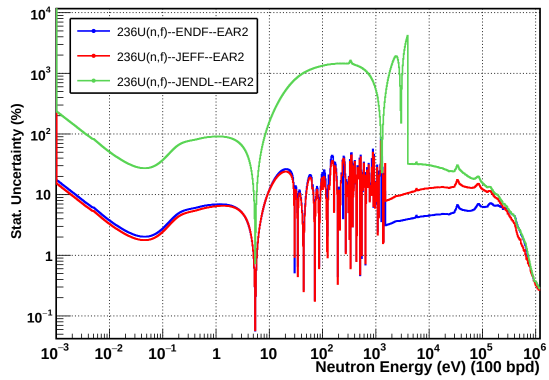
4 Summary

In conclusion, we propose the measurement of the neutron-induced fission cross section of ^{236}U for an extended neutron energy region, from thermal energies up to $\sim$ 0.5 GeV. The use of high-purity ^{236}U samples provided by JRC-Geel will allow for the collection of data with notably reduced ^{235}U impurities, compared to other TOF data-sets. The realisation of the experiment in EAR-2 will profit from an improved signal-to-background ratio, while the data collected from the EAR-1 measurement will allow for the extension of the cross section data in the high energy region. With this proposed measurement, the previous n_TOF data will be significantly extended in neutron energy. Discrepancies among previous measurements and evaluations will be addressed and a cross section measurement with a unified single data-set that covers 10 orders of magnitude in neutron energy from thermal up to $\sim$ 0.5 GeV will be produced.

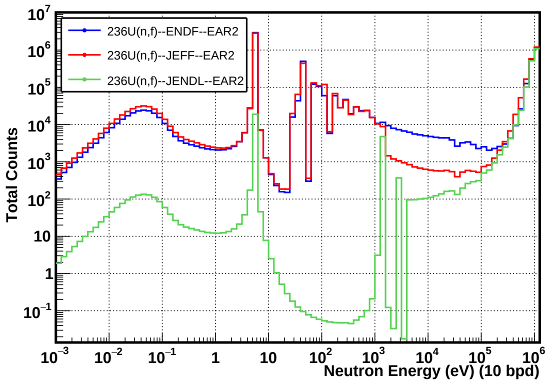
Summary of requested protons: 9x10^{18} protons on target
(6x10^{18} protons for EAR-1 and 3x10^{18} protons for EAR-2)
All tests and electronics optimisation needs are included in this request.



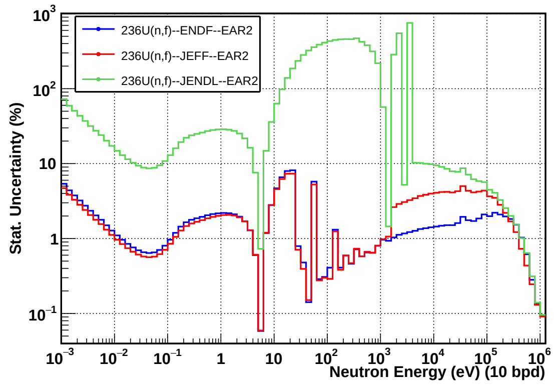
(a) Total counts at 100 bpd



(b) Statistical uncertainty at 100 bpd

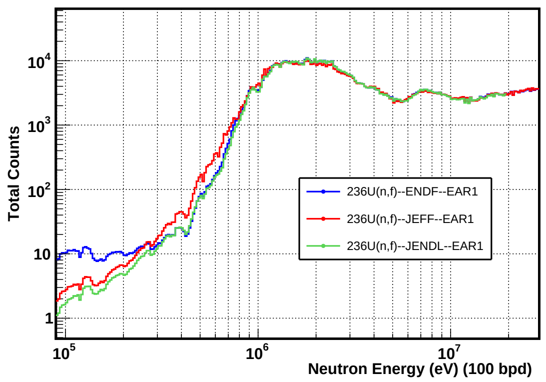


(c) Total counts at 10 bpd

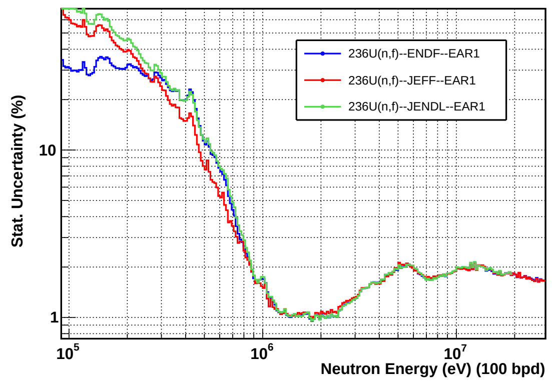


(d) Statistical uncertainty at 10 bpd

Figure 5: Reaction rate and statistical uncertainty estimates in EAR-2 from the thermal region up to 700 keV for 100 bpd (top panels) and 10 bpd (bottom panels) considering 3x10^{18} protons. Due to the considerable discrepancies among different evaluations both scenarios of data handling will be considered.



(a) Total counts at 100 bpd



(b) Statistical uncertainty at 100 bpd

Figure 6: Reaction rate and statistical uncertainty estimates in EAR-1 from the reaction threshold up to 30 MeV, which is the highest energy limit of the evaluations.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT
Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
If relevant, write here the name of the fixed installation you will be using [Name fixed/present n_TOF installation: uMegas/ EAR-1 and EAR-2	☒ To be used without any modification ☐ To be modified
If relevant, describe here the name of the flexible/transported equipment you will bring to CERN from your Institute [uMegas detectors]	☐ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing
[fission chamber]	☐ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing
[gas regulation system]	☒ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[preamplifiers]	☐ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing
[gas mixture of $Ar:CF_4:isoC_4H_{10}$ (88:10:2) and $Ar:CF_4$ (90:10)]	☒ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[samples of 236-U, 235-U, 238-U and 10-B]	☐ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing
[sample holders and masks]	☐ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT
Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Gas pressure inside the fission chamber	☒	[1.01325] [bar], [∼15][l]
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☐	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☐	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	☐	[laser], [class]
	UV light	☐	
	Magnetic field	☐	[magnetic field] [T]
Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	
	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			