EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurements of neutron induced capture and fission reactions on ^{233}U (EAR1)

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Abstract:

The ^{233}U plays the essential role of fissile nucleus in the Th-U fuel cycle, which has been proposed as a safer and cleaner alternative to the U-Pu fuel cycle. Considered the scarce data available to assess the capture cross section, a measurement was proposed and successfully performed at the n_TOF facility at CERN using the 4π Total Absorption Calorimeter (TAC). The measurement was extremely difficult due to the need to accurately distinguish between capture and fission γ-rays without any additional discrimination tool and the measured capture cross section showed a significant disagreement in magnitude when compared with the ENDF/B-VII.1 library despite the agreement in shape. We propose a new measurement that is aimed at providing a higher level of discrimination between competing nuclear reactions, to extend the neutron energy range and to obtain more precise and accurate data, thus fulfilling the demands of the "NEA High Priority Nuclear Data Request List". The setup is envisaged as a combination of the 4π Total Absorption Calorimeter (TAC) with MicroMegas (μMGAS) fission detectors. This setup configuration has been tested successfully in 2011 and proven to be adequate for performing measurements where both capture and fission channels are open. Additionally, the upgrade n_TOF Experimental Area 1 (EAR1) which is now a "Work Sector Type A", allow us to measure radioactive samples without the need to encapsulate them in a thick titanium canning reducing significantly the scattering background. The measurement will provide neutron-induced capture and fission cross sections as well very valuable information on the distribution of energies and multiplicities of the prompt γ-rays emitted after capture and fission reactions.

Requested protons: 4.3×10^{18} protons on target

1 Introduction

The present concern about a sustainable energy supply builds on several problems that must be addressed, namely the green house effect and foreseeable limits in fossil fuel resources, the concern about the environmental impact of nuclear fission energy and the long term fusion research. A variety of advanced strategies for the nuclear fuel cycle and related nuclear energy systems are being considered with emphasis on the extension of the life span of presently operating reactors, the increase of the fuel burn-up, the plutonium recycling, and in particular the incineration of actinides and long-lived fission products, the accelerator driven systems (ADS) concept and the possible use of the Thorium fuel cycle [1].

Thanks to the unique characteristics of the n_TOF facility at CERN, the n_TOF Collaboration has provided valuable capture and fission cross section data in the last few years [3]. Some examples of the measurements of neutron capture cross sections already performed at n_TOF are: ^{233,234,235}U, ^{237}Np, ^{240}Pu and ^{243}Am [4], ^{238}U and ^{241}Am [5]. All isotopes, except ^{233}U, show a characteristic threshold below which the fission cross section is very low and does not compete with the neutron scattering and the capture reactions. This is illustrated in Figure 1 for two particular cases measured already at n_TOF: the fissile ^{233}U and the non-fissile ^{237}Np (note the threshold just below 1 MeV).

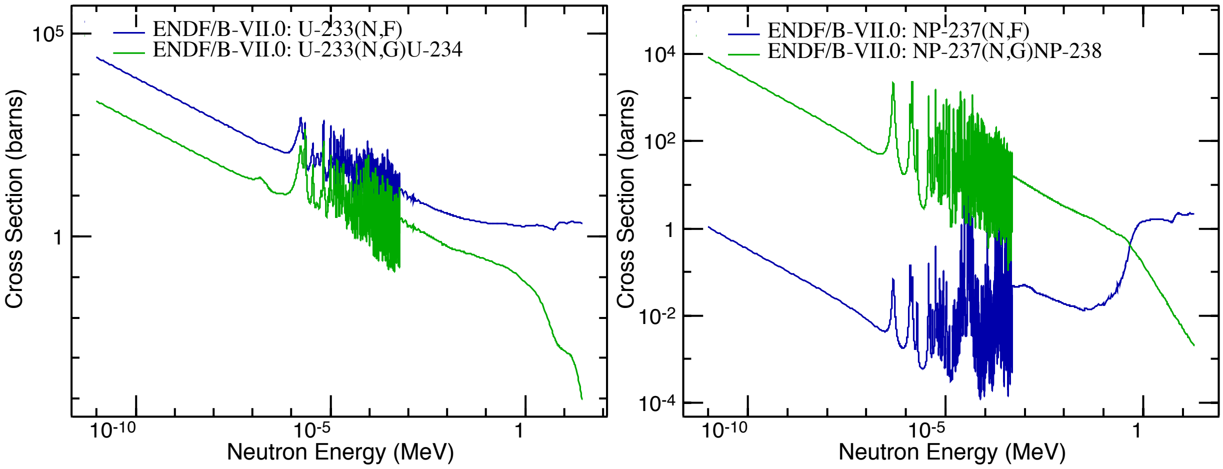


Figure 1: Neutron induced capture (green) and fission (blue) cross sections of the ^{233}U (left) and ^{237}Np (right). Data from the ENDF/B-VII.1 library [6].

The importance of the thorium fuel cycle is twofold: as an alternative fuel, it is a possibility for safe and sustainable energy generation, as well as the design and development of nuclear waste incinerators, based on the Th-U fuel. Indeed studies [1] for incinerating the radioactive waste from LWR's and for an alternative fuel with limited waste production allowing in addition a long-term sustainability have led to new interest in the accelerator assisted Th fuel cycle and more recent in Molten Salt Reactors (MSR) and Liquid Fluoride Thorium Reactor (LFTR) technologies.

This has been noted in the summary report of the INDC/IAEA consultant's meeting [8] that concluded:

1. There is a growing wide interest in advanced nuclear power technologies involving new types of nuclear fuel, providing inherent safety, resistance to nuclear proliferation, reduction of weapon grade plutonium and minimization of radioactive wastes.
2. The Th-U cycle is one of the most interesting solutions, promising to meet the above goals.
3. The nuclear data for nuclides of the Th-U fuel cycle have been evaluated in the early seventies and middle of the eighties and do not fulfill the current accuracy requirements. Similar efforts to the U-Pu cycle need to be undertaken for the Th-U cycle.

Important to note also that from the perspective of uranium resource conservation, many of the Generation IV systems investigated are fast neutron reactors that use plutonium and uranium recovered from spent fuel by reprocessing, and depleted uranium. Thorium was examined carefully by the Fuels Crosscutting Group during the original Roadmap and was not considered a first priority for Generation IV. However, this has changed and the Generation IV International Forum (GIF)[9] notes an interest in the use of thorium resources and are already seeing exploration of thorium-based fuels in some Generation IV systems to understand their potential benefits.

In order to simulate with the required accuracy the behavior of such alternative systems, and to advance towards their design, testing and eventual full deployment of a prototype based on the Th-U fuel cycle, basic nuclear data for the eight isotopes relevant in the Th-U fuel cycle, i.e. ^{230,232}Th, ^{231,233}Pa and ^{232,233,234,236}U, in the form of differential neutron cross sections, are needed. In the presently available compilation databases [10], nuclear data for the Th-U fuel cycle is in a less than adequate level, while the prospect of innovative concepts such as the Generation IV reactors, may broaden considerably the range of data required. The IAEA recommendations [8] included ^{232}Th, ^{231,233}Pa, and ^{232,233,234,236}U as first priority isotopes.

The target accuracies for ^{233}U together with other uranium isotopes are listed in Table 1.

Table 1: Information on fissile uranium isotopes listed in the NEA-HPRL [11]. Reaction channel and targeted accuracies are also given.

Isotope	Reaction	Quantity	Energy range	Targeted accuracy (%)
^{235}U	(n,f)	prompt γ-prod	Thermal-Fast	7.5
^{235}U	(n,γ)	σ, RP	100 eV-1 MeV	3
^{233}U	**(n,γ)**	σ	**10 keV-1.0 MeV**	**9**
^{233}U	**(n,γ)**	**nubar, σ**	**Thermal-10 keV**	**0.5, 5.0**

The ^{233}U has been measured at n_TOF (Figure 2) using the TAC and the Calorimetric Shape Decomposition (CSD) [12] technique and was considered successful despite all the difficulties of measuring the capture cross sections, taking in account it is a fissile isotope and the absence of additional discrimination setups [7]. The data analysis methodology performed allowed to assess simultaneously the capture and fission cross sections (12%

and 6.6% respective uncertainties) from 1 eV to 1 keV and the results showed a good agreement with the ENDF/B-VII.1 library in the case of the fission cross section.
Note that both cross sections were assessed simultaneously using the same methodology. Therefore, a systematic uncertainty in one cross section would reflect also in the other.

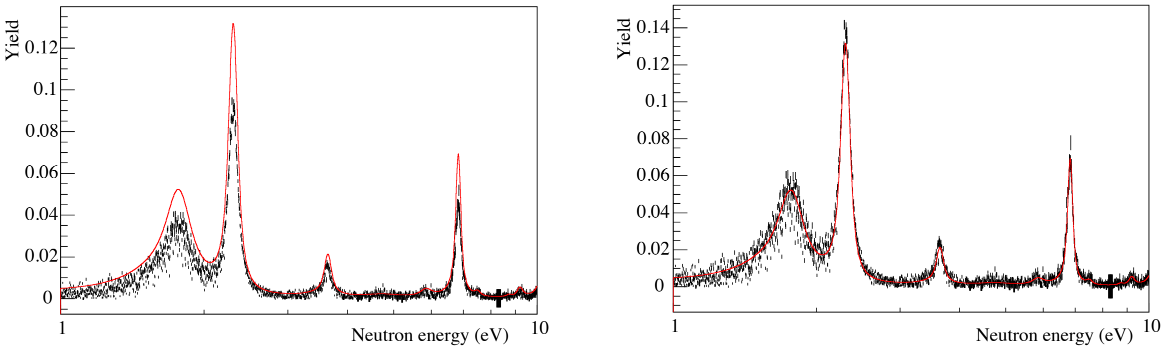


Figure 2: Comparison between the ^{233}U neutron capture yield measured at n_TOF (black) and the ENDF/B-VII.1 (red) library between 1 and 10 eV using the assessed normalization factor (left panel) and using and arbitrary normalization factor to match the ENDF/B-VII.1 library (right panel)

In the case of the capture cross section measured at n_TOF, a disagreement of ∼30% in normalization was found with respect to the data available in the ENDF/B-VII.1 library as shown in Figure 2. This disagreement cannot be explained by the 12% uncertainty of the measurement and we believe it points to a problem in current evaluations. However, it is important to confirm this result with a refined experimental system, which should also allow to improve the final accuracy in order to reach the NEA-HPRL request below 10 keV.
To this purpose, we propose a new measurement using a detection setup with better discrimination capabilities and a sample with reduced canning mass and complete absence of titanium, which prevented in the first measurement to collect data beyond few keV neutron energy. The absence of titanium and reduced canning mass is now possible due to the upgraded EAR1 at n_TOF which is now a "Work Sector Type A".

2 Experimental set-up

The proposed experimental set-up consists in the combination of the Total Absorption Calorimeter (TAC) [13] and the MicroMegas (μMGAS) [14] detectors (Figure 3 left panel) which have been used successfully in previous n_TOF measurements [15]. The TAC [13], is a segmented 4π array made of 40 BaF_2 crystals forming a spherical shell of 20 cm and 50 cm inner and outer diameters, respectively. The combination of its segmentation, high geometric and intrinsic efficiencies and energy resolution make the TAC an excellent device for measuring capture reactions of small mass and radioactive samples. The TAC has been successfully used in the past for measuring capture cross sections of actinides and has been also proven to be a powerful tool for the study of γ-ray strength functions [16].

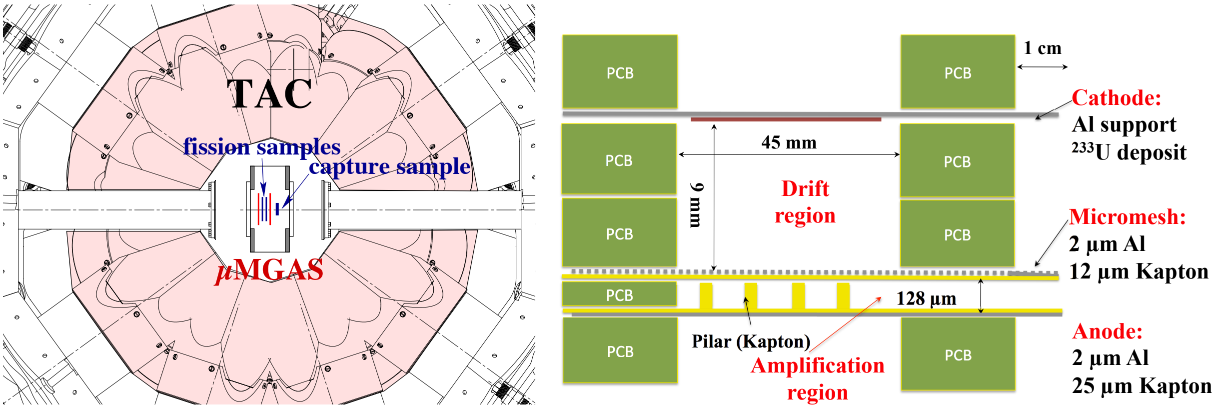


Figure 3: TAC and MicroMegas setup (left) and MicroMegas configuration (right).

The MicroMegas detector (μMGAS) [14], based in the microbulk technology, is a particular ionization chamber with a volume divided into two regions by a thin micromesh (see Figure 3 right panel). As the ionization takes place in the first region (drift region), the produced electrons are multiplied and amplified by an avalanche process in the second one (the amplification region). In our case the electrons will be generated by ionization of the gas by fission fragments from the ^{233}U samples. The advantage of this type of detectors is that the assembly has a good transparency to neutrons and γ-rays, thus minimizing the background associated with neutron reactions in the structural material while preserving the γ-ray detection efficiency. Indeed, several μMGAS detectors are being used regularly at n_TOF for monitoring the neutron beam (μMGAS loaded with ^{10}B and ^{235}U samples) and for measuring fission cross sections (^{240,242}Pu) and (n,α) reactions (^{33}S). The proposed set-up consists of a detector chamber containing 2 μMGAS detectors plus a thick capture sample placed in the inner hole of the TAC. The set-up is sketched in the left panel of Figure 3. Each detector will be loaded with a ^{233}U sample 4 cm in diameter with 300 μg/cm^2 ($\sim$ 4 mg/sample). The thickness has been chosen for reaching a detection probability larger than 90%. In this way, the detector assembly maybe operated in veto mode [15].

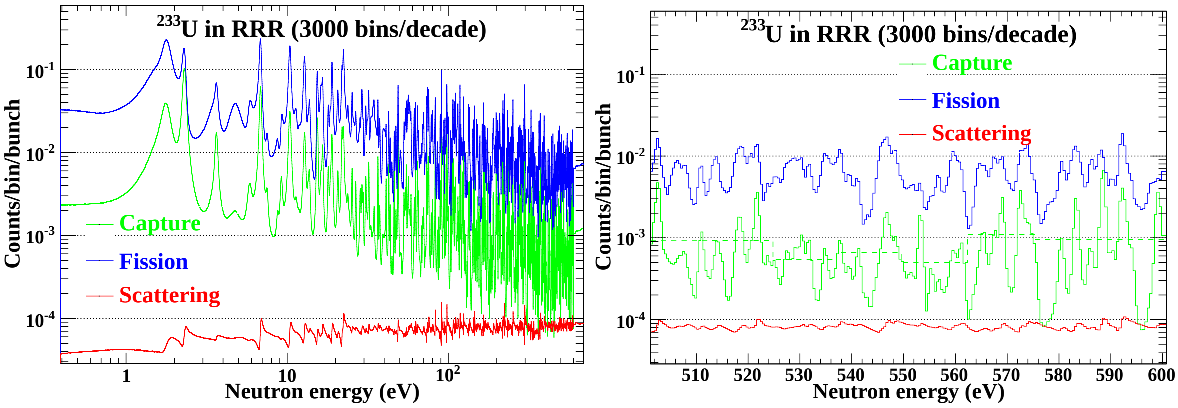
In the proposed detector, the electrodes are made of aluminum in order to reduce the large background produced in the TAC by the previously used copper electrodes above 600 eV. The active diameter of the detector is 4.5 cm and the drift region can be varied from 2 mm up to 1 cm. The amplification region is fixed at 128 μm. A vertical cut of the prototype detector is shown in Figure 3 and the different components and regions are identified.

The μMGAS consist of three electrodes made by Al deposition on a kapton ($C_{22}H_{10}N_{2}0_{5}$) layer. Because of technical difficulties in the chemical process, a layer of less than 100 nm of Cr is added in between. It is not expected that such a thin layer of Cr contributes significantly to the TAC background. This set-up will allow to measure up to few keV.

In order to extend the measured neutron energy range up to 1 MeV, and then fulfill the second part of the NEA request, gated photomultipliers [17, 18] should be used in order to strongly reduce the "γ-flash" effect in the TAC.

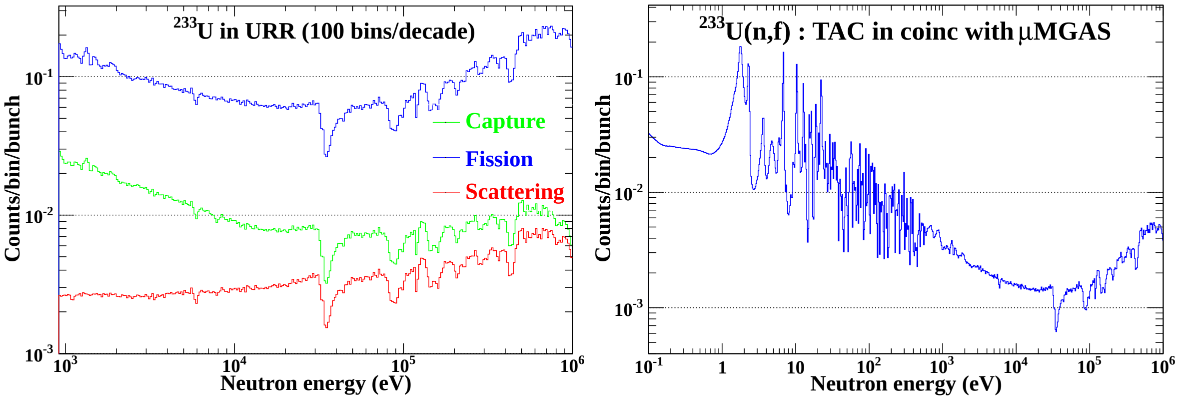
The samples foreseen to be measured consist of the thick high-purity ^{233}U sample (91 mg of ^{233}U) used in the previous measurement at n_TOF [7, 12] and two additional thin fission samples in the Al-μMGAS detectors. The target used in the first measurement was encapsulated in Al and Ti, to comply with the safety rules at CERN, prior to the modification of the experimental area to "Work Sector Type A". In the new measurement, the Ti canning will be removed, thus allowing to reach higher neutron energies. The thin fission samples can be obtained from ILL Grenoble. We are in contact with various Institutes (in particular with PSI) for the removal of the Ti canning from the thick sample.

3 Beam time request



(a) Counts in the resolved resonance region

(b) Zoom around 500 eV



(c) Counts in the unresolved resonance region

(d) Counts in TAC in coinc with μMGAS

Figure 4: Count rate estimates based on ^{233}U(n,*) cross sections from ENDF/B-VII.1 database

In order to perform these measurements several configurations will have to be used:

- Capture sample in TAC to extract the ^{233}U capture cross section
- Coincidence measurements between TAC and fission fragment detection to assess the TAC calorimetric response to fission (with capture sample in the TAC in order to investigate also any possible count rate effect in the TAC)

- TAC response to neutron scattering by using a carbon sample
- TAC response to sample canning
- TAC response to gold for normalization
- Measurements with capture sample without beam to assess the TAC response to sample radioactivity.

Panels a,b,c in Figure 4 show the expected counts in the n_TOF TAC [13] per bin and per proton pulse, deduced from the ENDF/B-VII.1 [6] evaluation for neutron induced capture (green), fission (blue) and scattering (red). The following assumptions were considered:

- The sample is the same as in 2003 (after removal of the Ti canning)
- The fraction of the beam intercepted by the sample (the so-called Beam Interception Factor) is 20% (1 cm sample diameter, 3.5 cm beam diameter)
- Events with γ multiplicity higher than 2 are only considered in the TAC. With this event selection criteria the following efficiencies are expected:
  - about 60% for capture events
  - about 70% for fission events
  - neutron sensitivity (neutron scattering) of the TAC is around 1%.
- 7×10^{12} proton per pulse.

The NEA HPRL [11] (see table 1) requests the capture cross section to 5.0% accuracy in the resolved resonance region (RRR). For measurement up to few keV the count estimate for the ^{233}U neutron capture reaction will be driven by the RRR. As can be seen in Figure 4b approximatively 5×10^{-4} counts/bin/bunch (green line) are associated with capture at a resolution of 3000 bins/decade. In this region and with that resolution a resonance is described by about 5 bins, resulting in about 2.5×10^{-3} counts/bunch per resonance. Therefore, a 3% statistical uncertainty per resonance requires a total of 4×10^5 bunches, corresponding to about $\mathbf{2.8 \times 10^{18}}$ protons.
If gated photomultiplier will be operational, the measurement could be extended to 1 MeV neutron energy without requesting additional protons, since the counting rate will be still driven by the RRR due to the less demanding accuracy (9% instead of 5%) in the URR. Nevertheless one should note that the capture signal in the TAC will be strongly overlapped by the fission signal that will be about 10 times stronger in the whole energy range as can be seen in Figure 4. The Calorimetric Shape Decomposition [12] will be used to overcome this difficulty, but the capture uncertainty in the high energy part of the RRR will be still limited by the statistical uncertainty of the fission shape. In order to obtain an accurate shape associated with fission events in the TAC a specific measurement will be performed. Figure 4d shows the expected number of counts in the TAC associated with neutron induced fission detected in coincidence with the set of MicroMegas detectors. By integrating this number of counts over the full energy range, one can expect about 4 fission events per bunch with this experimental set-up. In order to obtain an accurate

energy deposition spectrum in the TAC associated with fission, one needs about 2×10^5 fission events in the deposited energy spectra, corresponding to $\mathbf{4 \times 10^{17}}$ protons.
As can be seen in Figure 4a,c the neutron scattering signal in the TAC is at most 10% of the capture events in the RRR and in the URR. Accordingly, a statistical uncertainty of 10% in the RRR (100 bins/decade should be sufficient for the scattering contribution) and 5% in the URR are sufficient to determine the scattering contribution. To fulfill this request $\mathbf{5 \times 10^{17}}$ protons are necessary.
The canning induced background is expected to be similar to the counts obtained from ^{233}U, so an additional $\mathbf{5 \times 10^{17}}$ protons should be devoted to the canning measurement.
Finally $\mathbf{1 \times 10^{17}}$ protons should be devoted to measure the standard gold cross section.
A summary of the necessary protons is given in Table 2.

Table 2: Summary of the protons needed to perform the ^{233}U capture and fission cross sections measurement and auxiliary measurements

Measurement	Number of protons
^{233}U capture and fission	$2.8\cdot10^{18}$
Coincidence measurement between TAC and fission fragment detection	$4.0\cdot10^{17}$
TAC response to neutron scattering using a carbon sample	$5.0\cdot10^{17}$
TAC response to the sample's canning for background subtraction	$5.0\cdot10^{17}$
Gold	$1.0\cdot10^{17}$
Total	$\mathbf{4.3\cdot10^{18}}$

4 Conclusion

We propose to measure simultaneously the neutron capture and fission reactions on ^{233}U down to 0.5 eV. The data will provide the neutron capture cross section and the capture to fission ratio as a function of neutron energy up to at least 10 keV. The goal of the measurement is to reach a statistical uncertainty better than 3% and a total uncertainty in the capture cross section of about 5% in order to fulfill the NEA HPRL request in the low energy range. The energy range in the analysis of the data can be extended up to about 1 MeV if gated photomultipliers will be available for the n_TOF TAC, in this way fulfilling in addition the high energy request. Gated electronics suited for the TAC photomultipliers are currently under development.
The measurements will be carried out with a new set-up that combines the 4π BaF_2 Total Absorption Calorimeter with several MicroMegas detectors made of aluminum instead of copper. We propose to use the capture sample of the 2003 measurement after removal of the titanium capsule, and two additional fission samples of about 4 mg each.
Considering the available mass and targeted uncertainty a total of 4.3×10^{18} protons are requested to perform the measurements.

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