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



<u>Z. Eleme</u>¹, <u>A. Tsinganis</u>², N. Patronis¹, J. Heyse², P. Schillebeeckx², M. Bacak^{3,4}, N. Colonna⁵, M. Diakaki⁶, S. Goula^{1,3}, M. Kokkoris⁶, N. Kyritsis⁶, V. Michalopoulou⁶, D. Papadimitriou¹, M. Peoviti¹, M.E. Stamati^{1,3}, R. Vlastou⁶ and the n_TOF Collaboration



¹ UOI, ² JRC, ³ CERN, ⁴ UMAN, ⁵ INFN-Bari, ⁶ NTUA



76th INTC Meeting, CERN 22 May 2024



Zinovia.Eleme@cern.ch

University of Ioannina

Motivation

- High- accuracy cross section data for neutron-induced reactions are needed over a wide energy range for the **design**, **feasibility and sensitivity studies on advanced nuclear systems**
- The ²³⁶U isotope ($T_{1/2}$ = 2.342x10⁷ years) has the **longest half-live** compared to any other fission product or actinide produced in nuclear reactors
- Due to its specific activity (2.4 MBq/gr) which is 190 times higher than the one of ²³⁸U, it significantly contributes to the radioactivity of reprocessed uranium
- In current reactors based on U/Pu fuel, ²³⁶U is produced by (n,y) on ²³⁵U \rightarrow considerably affects the neutron balance in the reactor core as well as the fuel composition
- Moreover, ²³⁶U builds up in the equilibrium state in the Th/U fuel cycle
- For the development of fast nuclear reactors and accelerator-driven systems (ADS) the knowledge of its fission cross section is required within 5% accuracy





Existing experimental data from EXFOR in the low energy region



Scarce and discrepant experimental data

*The error bars of the experimental data are omitted in this plot

- In the thermal region, evaluated libraries, in particular, JENDL-5, JEFF-3.3, ENDF/B-VIII-0 and TENDL-2021, exhibit major discrepancies of up to 2 orders of magnitude
- Only JENDL-5 seems to fairly reproduce the only 2 data points in the thermal region by Wagemans et al.
- There are only 2 data-sets covering the first resonance of ²³⁶U @5.45 eV
- Data from Sarmento et al. with the highest resolution in this region, reveal that current evaluations need revision since they overestimate the height of the resonance by ~150 times
- In the region between 30 eV and 1 keV, resonance structures reported by Cramer and Berger have been adopted by some evaluations but are not confirmed by later measurements



Existing experimental data from EXFOR in the high energy region



- Above 500 keV the situation of the experimental data is better since there are a lot of measurements that have lead to the improvement of the knowledge of the cross section in the high energy region
- Nonetheless, discrepancies of up to ~15% are observed among recent datasets especially for the region of interest for the fast nuclear reactors (0.5 - 10 MeV)
- Moreover, above 40 MeV, which exceeds the upper limit of most evaluations, only 3 data-sets are available
- The experimental information in this region is of particular importance to constrain the theoretical models of the fission process



Existing experimental data from EXFOR in the high energy region



- Systematic discrepancies among different data-sets above 100 keV are better demonstrated by the fission cross section ratio of ²³⁶U/²³⁵U
- Above 2 MeV, discrepancies of up to 15% are observed among EXFOR data-sets

Previous measurement at n_TOF

- A previous measurement was already performed at n_TOF in 2003 (EAR-1) during Phase I
- The fission yield of the ²³⁶U(n,f) reaction was affected by the contribution of α- particles from the ²³⁶U decay but most importantly from the 0.05% impurity of ²³⁵U present in the samples
- After the subtraction of the ²³⁵U contamination, the analysis revealed resonance structures attributed to ²³⁶U @5.45 eV and @1.25 keV in the form of a triple resonance
- Unfortunately, above the fission threshold, data were limited up to 2 MeV



FIG. 4. (Color online) Fission yield measured with samples (black histogram), ²³⁵U(*n*, *f*) yield normalized to the ²³⁵U impurity (green dashed histogram), and the residual α -particle background (orange circles). The contribution of the ²³⁵U contamination to the ²³⁶U signal is evident in the region above the ²³⁶U resonance at 5.45 eV.

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FIG. 6. (Color online) Resonance triplet at 1.25 keV was resolved at n_TOF with high resolution. The n_TOF data (black points, vertical error bars corresponding to statistical uncertainties) were fit with SAMMY (blue line).



FIG. 9. (Color online) Fission cross-section ratio of 236 U and 235 U above threshold. The n_TOF results are plotted together with experimental data available from the EXFOR database [9,14,15,21–24].

Plan of the measurement

- We aim at producing, for the first time, a single accurate data-set covering the energy range from thermal up to ~ 0.5 GeV neutron energy
- Fission collimators in both areas to profit from increased statistics (8 cm diameter in EAR-1 and 6 cm diameter in EAR-2)
- 6E18 protons in EAR-1 to cover the fission threshold up to ~0.5 GeV
- 3E18 protons in EAR-2 to collect data in the thermal and resonance region up to ~700 keV
- Use the same high-purity ²³⁶U samples used by Wagemans et al. for the determination of the cross section at the thermal points
- Use aluminum "masks" in all samples (236U and references), so as to achieve the same effective diameter of 4 cm

 → important step in order to perform a relative fission cross section measurement since the same
 "Beam Interception Factor" (BIF) for all samples will lead to reduced systematic uncertainties in the analysis





Micromegas detectors

- Micromegas (MICRO- Mesh Gaseous Structure) detectors offering ~100% efficiency
- Parallel plate avalanche gaseous detectors consisting of two regions: the conversion and the narrow amplification region
- Each detector is coupled with a sample, therefore creating a stack of sample-detector modules (2 x ²³⁶U samples, 6 x reference samples, 1 x empty)
- All the sample-detector modules, will be housed in an aluminum chamber
- The fission chamber will be filled with a constantly circulating gas mixture @ 1 atm EAR-1: Ar:CF₄:isoC₄H₁₀ (88:10:2) and for EAR-2: Ar:CF₄ (90:10) → minimize the elastic interactions with the hydrogen inside the drift region of the detector → better γ-flash tail
- Employ a gas regulation system \rightarrow to ensure stable gain conditions for the detectors







Anode

236U samples from JRC-Geel

- 2 x ²³⁶U samples of high purity
- Mass isotopic composition 234U: <0.00001% 235U: 0.0043% 236U samples 236U: 99.9732% Target ID: NS0010 NS99008 238U: 0.0225% Description: 236U oxide layer (lot number. 2329) Preparation method: Electrolysis Diameter of deposit (mm): 50 40 Backing: 20 um Al 0.5 mm Si Areal Density (ugr/cm2): 210.442 +/- 1.365 131.758 +/- 0.466 Total Activity of 236U (kBg): 9.9 4.0 * Refers to the unmasked part of *Effective Activity (kBq): 6.3 4.0 the target *Effective Mass (mgr): 2.64 1.66
 - For the neutron flux determination we will use the already existing reference samples (¹⁰B, ²³⁵U and ²³⁸U samples with 6 cm diameter) from the ²⁴³Am(n,f) campaign which are well optimized for both experimental areas



Proposed set-up inside the fission chamber



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EAR-2: Reaction Rate @100 bpd

- Upper and lower limit for the reaction rate and stat. uncertainty estimates in EAR-2 from the thermal region up to 1 MeV
- Beam request: 3E18 protons



EAR-2: Reaction Rate @10 bpd

- By adopting a coarser energy bin (e.g 10 bpd) in the thermal region we can further reduce the stat. uncertainties
- In the end, we can use a coarser energy binning for the thermal region and around 5.45 eV we can provide a better mapping of the resonance that dominates the fission cross section below the threshold



EAR-2: Cumulative fission yield

 Even if our experiment confirms the JENDL-5 evaluation, limiting in this way the measurement in the thermal part, the contribution of the first resonance of ²³⁶U @5.45 eV will be important because it accounts for almost 90% of the total fission yield up to 100 keV and 99% up to 1 keV



Cumulative Counts 236U-JENDL---EAR2---



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EAR-1: Reaction Rate @100 bpd

- Reaction rates and stat. uncertainty estimates in EAR-1 from the threshold of the reaction (~300 keV) up to 30 MeV
- Beam request: 6E18 protons

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• We can provide fission cross section data above 1 MeV with a stat. uncertainty below 2% even at 100 bpd



Summary

- We propose the measurement of the neutron-induced fission cross section of ²³⁶U for an extended energy region, from thermal energies up to ~0.5 GeV by exploiting the specific characteristics of both n_TOF areas
- The use of **high-purity** ²³⁶U samples provided by JRC-Geel will allow for the collection of data with notably reduced ²³⁵U impurities, compared to other TOF data-sets
- In EAR-2, given the higher instantaneous neutron flux and the improved signal-to-background ratio, 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
- Data collected from EAR-1 measurement will cover the energy region from the fission threshold up to several hundreds of MeV
- We aim to improve significantly the accuracy of current data and resolve long standing discrepancies of evaluations and different data-sets in the low energy region
- Summary of requested protons: 6E18 protons in EAR-1 and 3E18 protons in EAR-2



Thank you for your attention!!

Back up slides

Counting rate estimation in EAR-2

Counting rate estimation taking into account the thickest ²³⁶U sample (210 ugr/cm2) and using ENDF/B-VIII.0 library

 We can extend the measurement up to 700 keV where the CR< 1 MHz and therefore we will not suffer from pile-up in the analysis



Expected Counting Rate Total Uranium-ENDF---EAR2---



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Expected reaction rate according to mass composition

Scenario A: cross section of ²³⁶U close to **JENDL-5**

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Scenario B: cross section of ²³⁶U close to ENDF/B-VIII.0



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Reference samples from JRC-Geel

- For the neutron flux determination we will use the already existing reference samples from the ²⁴³Am(n,f) campaign which are well optimized for both experimental areas
- ${}^{10}B$, ${}^{235}U$ and ${}^{238}U$ samples with 6 cm diameter

	Sample ID	Areal density (ugr/cm²)
10B (EAR-2)	TP2020-011-02	0.3
235U (EAR-2)	TP2020-009-04	4.3
	TP2020-009-06	3.5
235U (EAR-1)	TP2020-009-02	74
	TP2020-010-04	76
238U (EAR-1)	TP2020-010-04	226



Many thanks to the Target Preparation Laboratory team of JRC-Geel for the sample production (A. Moens, G. Sibbens, D. Vanleeuw)

Expected FF reaction rate of 236U according to JENDL-5 in both EARs



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Sample holders with masks

- Not all samples have the same dimensions
- In order to reduce the systematic uncertainties in the analysis the same "Beam Interception Factor" (BIF) should be applied to all the samples
- To achieve that, we plan to use aluminum "masks" with a diameter of 40 mm, so that all the samples will end up with the same exposed "effective" surface to the neutron beam

