Final results of the ^{239}Pu neutron capture and fission cross-section measurements at n_TOF

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B. and the n_TOF collaboration

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Accelerator and Research reactor Infrastructures for Education and Learning ARIEL

Contents

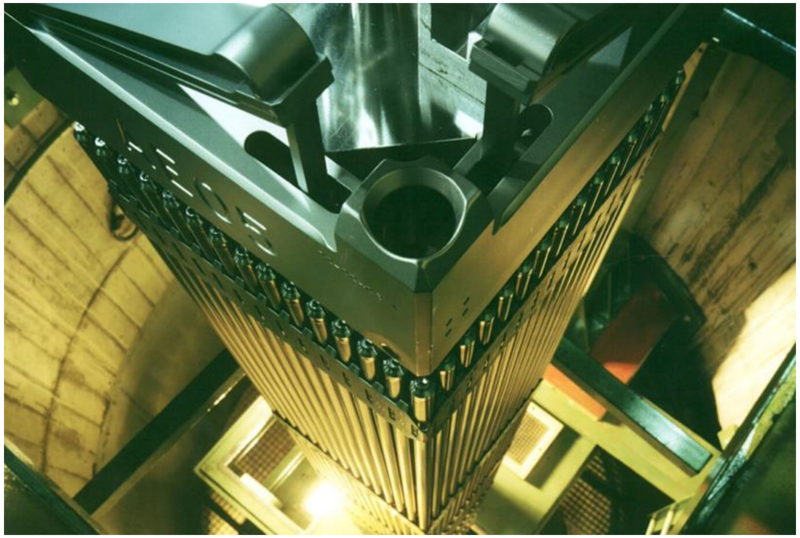
1

Context and Motivation

1. Context and Motivation

More precise ^{239}Pu capture and **fission** cross-section data are required for:

- Design of advanced nuclear devices (Gen IV reactors).
- Optimization of nuclear waste management strategies of current reactors.
- Operation of fast and thermal reactors that use MOX fuels.



Nuclear data evaluations for ^{239}Pu(n,g) and (n,f)

- Main evaluations for capture cross-sections show **significant discrepancies**.
- **Only two high-resolution measurements** for ^{239}Pu(n,g) cross-section exist, due to the intrinsic challenge of measuring a fissile sample.

^{239}Pu capture and fission cross-sections are included in the **NEA/OECD High Priority Request List.**

- **Objective**: to **reduce** the existing **uncertainties** of for **^{239}Pu(n,γ) and (n,f) cross-sections**.

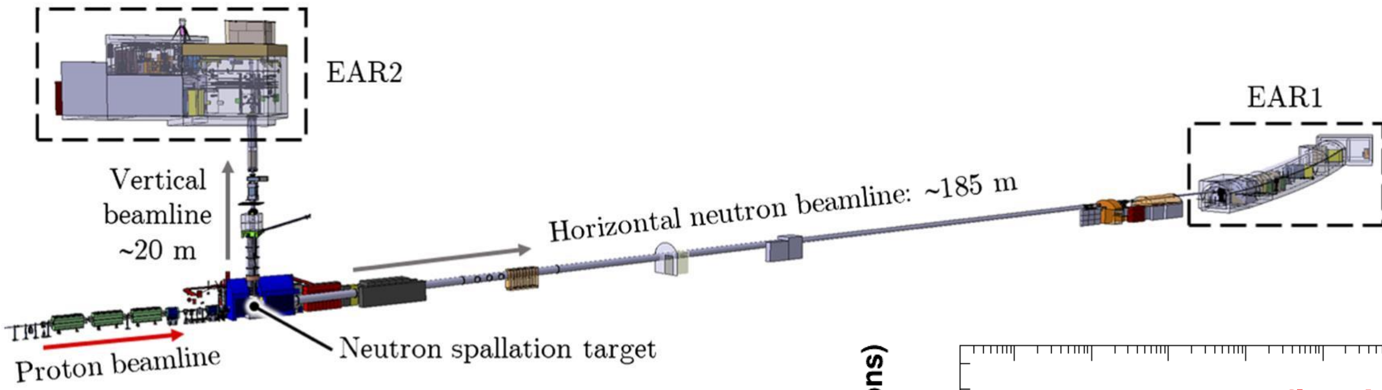
2

^{239}Pu measurement at n_TOF

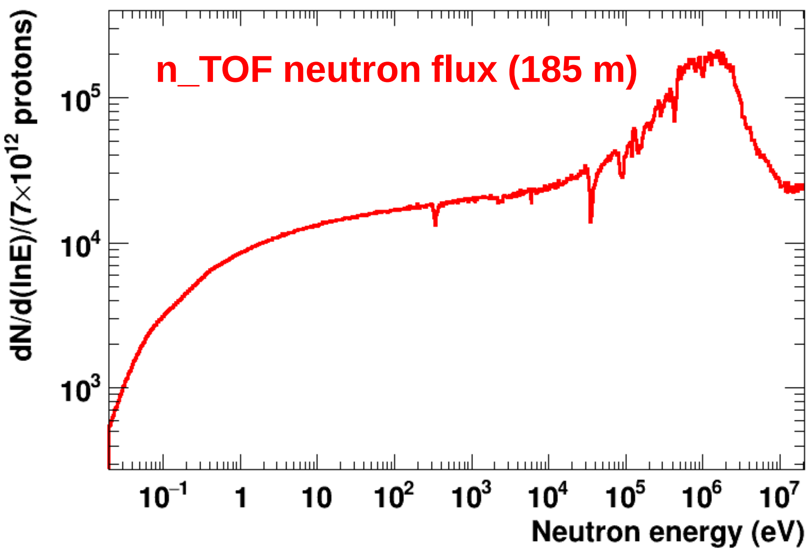
2. ^{239}Pu measurement at n_TOF

n_TOF: a neutron Time-Of-Flight facility

- A **185 m flight path** (**10 times larger** than in previous ^{239}Pu measurements) enables providing new data with **higher energy resolution**.



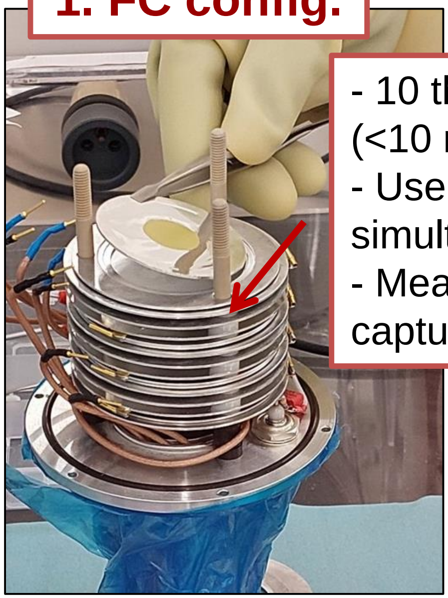
- **20 GeV/c protons** impinging on lead target, produce **neutron beam by spallation.**
- Low pulse repetition rate ~1 Hz; **no overlapping pulses**!
- **Broad neutron energy** spectrum **from meV to GeV**.
- **High intensity neutron flux**, ideal for radioactive samples.



2.1 Overview of the experiment and samples

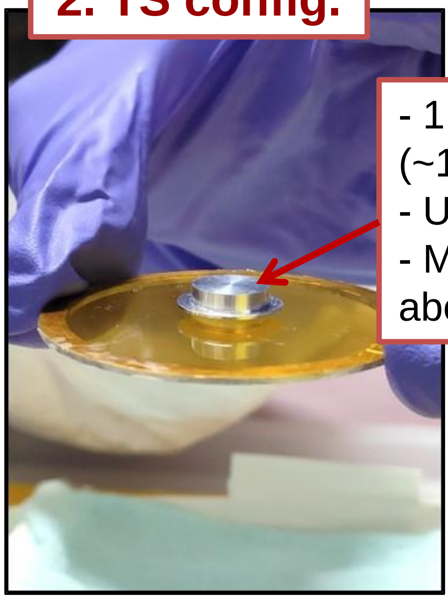
- The experimental campaign was conducted in the last quarter of 2022, with **2 months of beam time** (~$5 \cdot 10^{18}$ protons).
- The campaign was divided in **two different configurations**:

1. FC config.



- 10 thin Pu samples (<10 mg total mass).
- Use FICH + TAC simultaneously.
- Measure fission and capture (up to 1 keV).

2. TS config.



- 1 thick sample (~100 mg).
- Use only TAC.
- Measure capture above 1 keV.

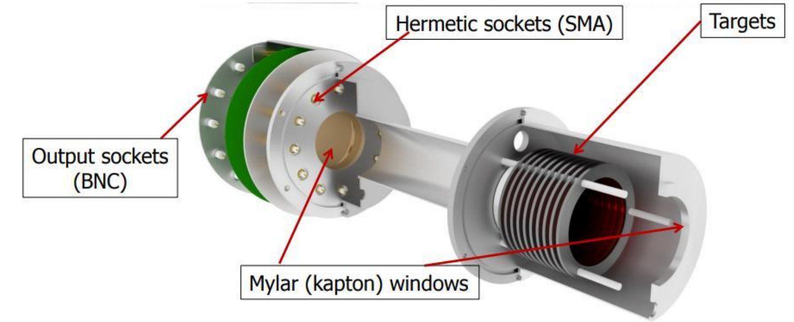
The ^{239}Pu targets

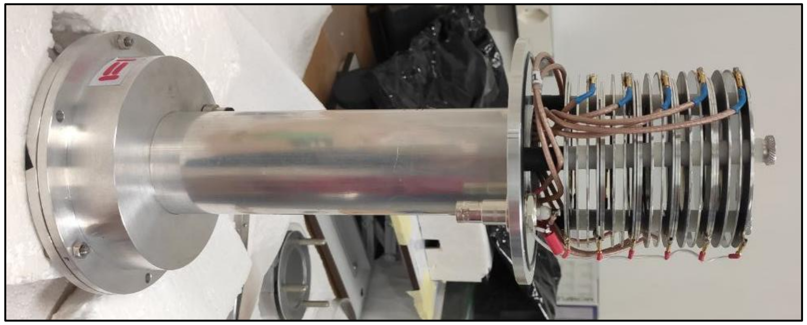
- The **PuO_2 (99.90% purity) 10 thin samples (~1 mg each)** and the **thick sample** (~**100 mg**) were produced, deposited and encapsulated by JRC-Geel+SCK·CEN.

2.2 Main detectors

Fission Fragment Detector (FFD)

- To perform **<u>fission</u> tagging** with the TAC and to **measure fission** cross-section.
- Housing of 10 parallel targets of PuO_2 deposited in 10 µm aluminum backing.
- Fast pre-amplifiers.
- Filled with Ar+CF_4 gas. Efficiency of ~90%.



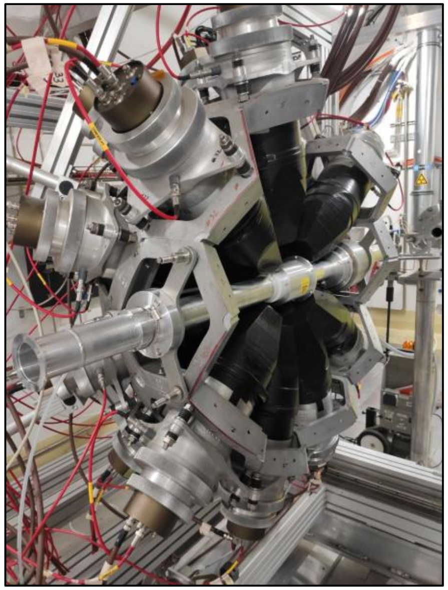


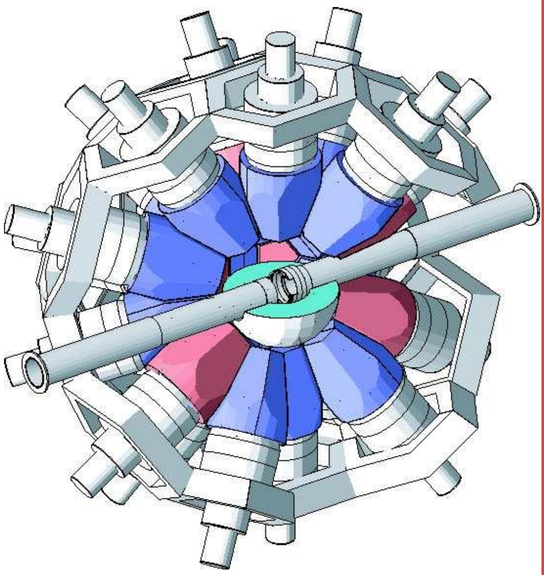
Silicon Flux Monitor (SiMon)

- Four silicon detectors + LiF foil.
- Measure **neutron flux** based on $^{6}Li(n,\alpha)^{3}H$ reaction.

Total Absorption Calorimeter (TAC)

- To **measure capture** by detecting ***γ*-rays**.
- Composed of 40 BaF_2 crystals.
- Fast response, high efficiency and low neutron sensitivity.

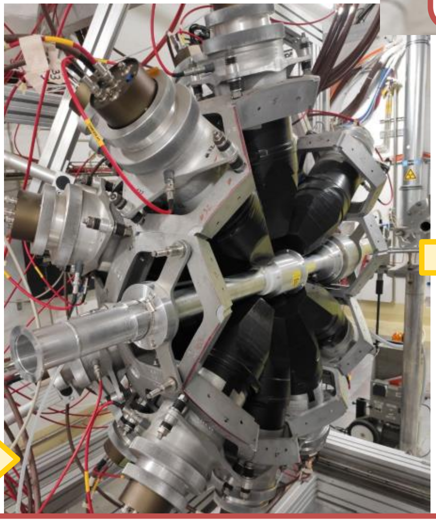




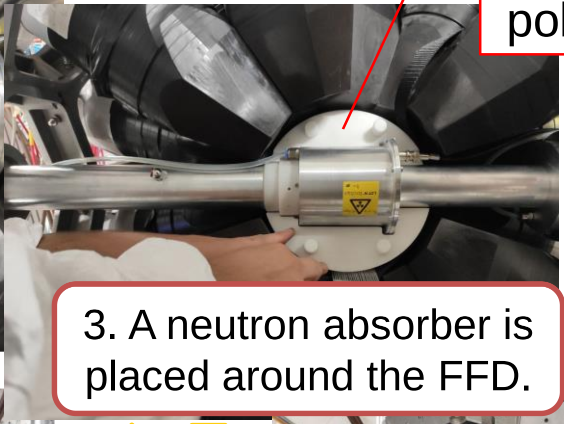
2.3 Experimental setup: mounting

The mounting procedure was similar in both experimental setups.

1. The chamber is placed within the TAC

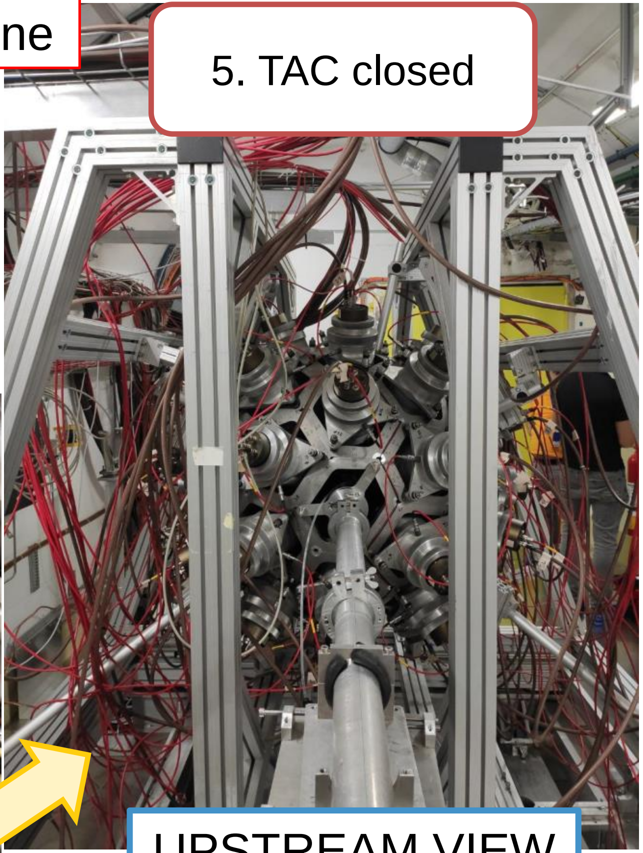


2. The TAC+FFD assembly is perfectly align with the neutron beam



3. A neutron absorber is placed around the FFD.

4. Neutron absorber closed.



5. TAC closed

3

Final results

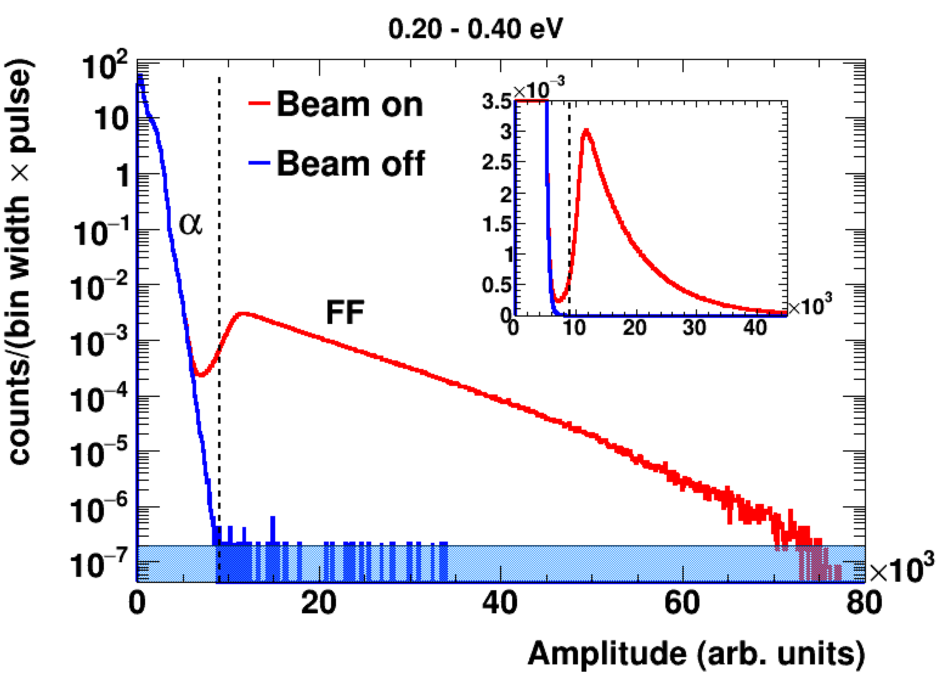
3.0 A glance at the Data Analysis

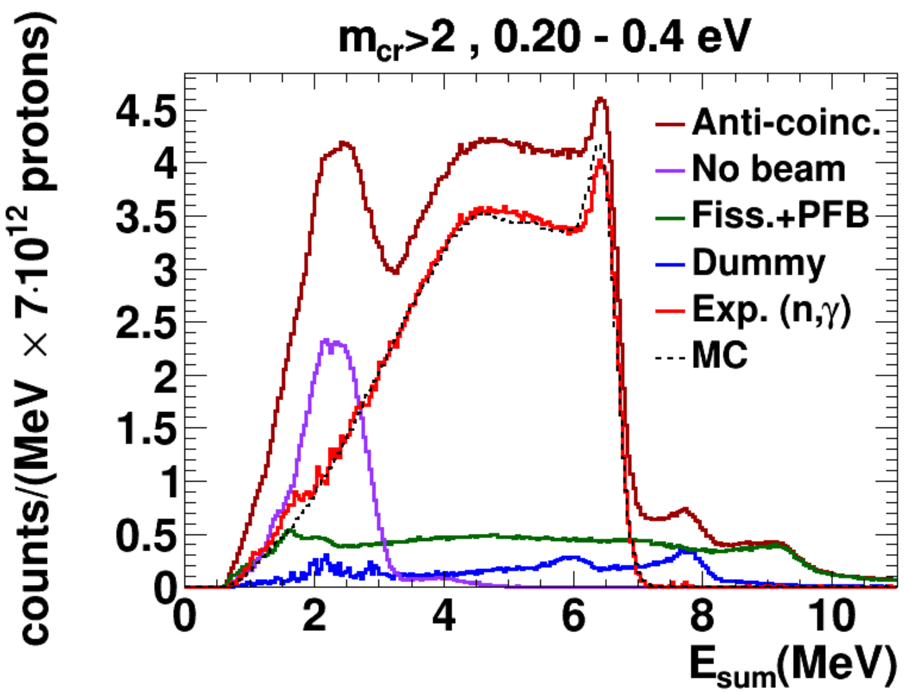
FFD

- Amplitude spectra in the fast fission detector.
- FF = Fission Fragment
- Vertical line: selected α-FF threshold.
- **1 FF per >2000 alphas.**

TAC

- Sum energy spectra with the different **background** components, for the first resonance at 0.3 eV.
- MC = Monte Carlo simulation of $^{239}Pu(n,\gamma)$.





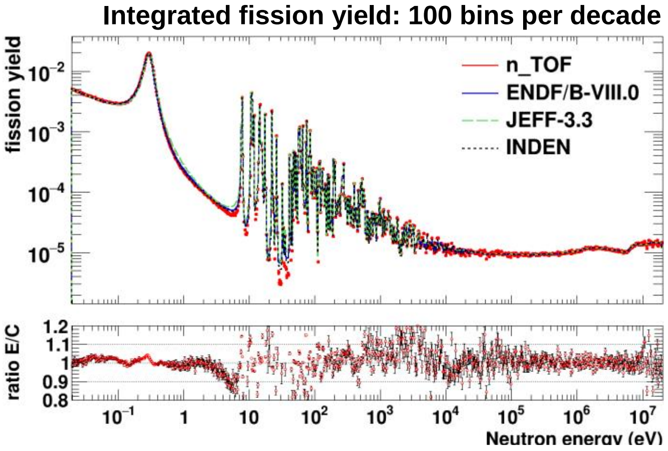
E. Mendoza et al. NuDEX: a new nuclear γ-ray cascade generator. EPJ Web of Conferences 239, 17006 (2020)
Access to code (github): **https://github.com/UIN-CIEMAT/NuDEX**

E. Mendoza et al. Study of photon strength functions of ^{241}Pu and ^{245}Cm from neutron capture measurements. EPJ Web of Conferences 239, 01015 (2020)

3.1 ^{239}Pu fission results

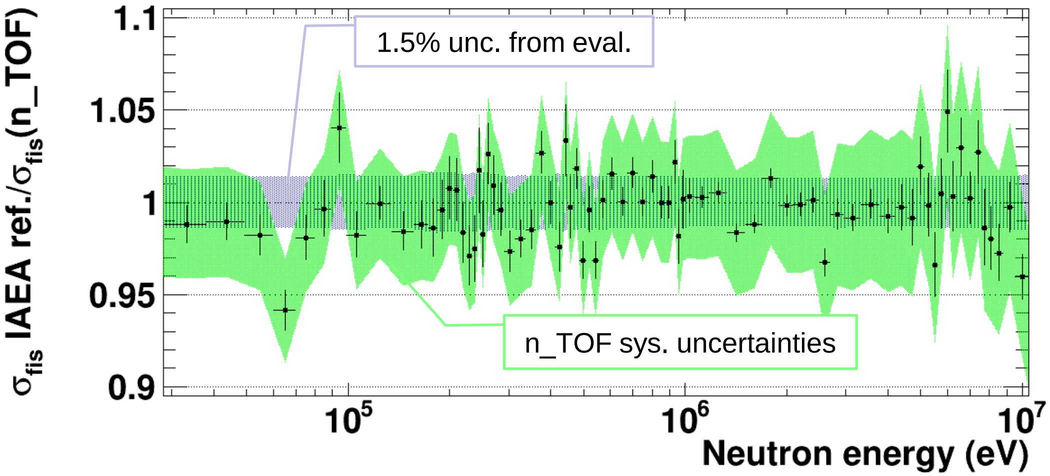
Fission yield was **normalized** to an evaluation of the XS integral in the 9-20 eV range.

An overall good agreement with evaluations for 9 orders of magnitude in one-single-measurement! (from thermal to high energy neutrons)



Ratios calculated relative to INDEN
https://www-nds.iaea.org/INDEN/

Our results are also compatible with the ^{239}Pu(n,f) cross section **reference (0.15-200 MeV) from the IAEA**'s Neutron Data Standards (NDS).



^{239}Pu(n,f) spectrum-averaged cross section (SACS) in the ^{252}Cf(sf) reference neutron field. Total SACS uncertainties are reported.

Data	SACS
Derived, this work	**1802±65 (3.6%)**
Derived, IAEA standard 2017[1]	1798±23 (1.3%)
Mannhart evaluation[2]	1812±25 (1.4%)
Capote et al. evaluation[3]	1826±19 (1.0%)

- Sys. uncertainties of 2-4.5% in the whole range (20 meV to 10 MeV).

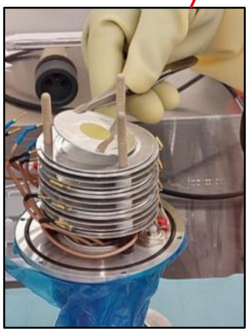
The new n_TOF ^{239}Pu(n,f) data is being considered for the next evaluation of the NDS.

3.2 ^{239}Pu capture results

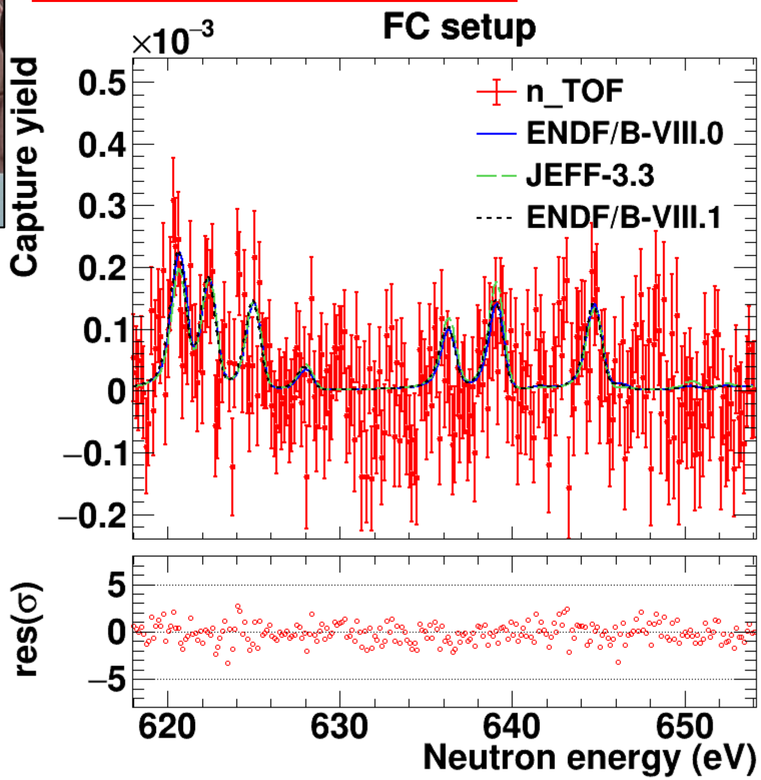
The final capture cross section **combines** the data from **both the FC and TS setup**.

- **TS data** extends the measurement to **higher neutron energies** and **improves precision in small resonances** with too low statistics in FC setup.

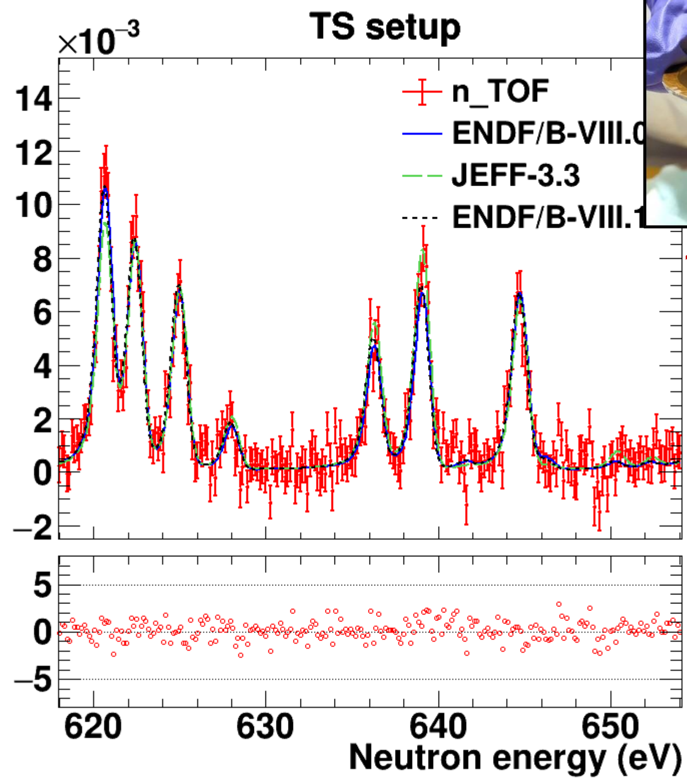
FC setup: limited statistics
A clear drop in statistics is observed above a few hundred eV.

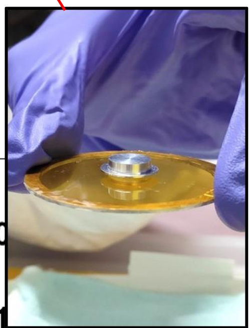


<10 mg



TS setup: improved statistics
The larger sample mass provides sufficient statistics to resolve the resonance shapes.

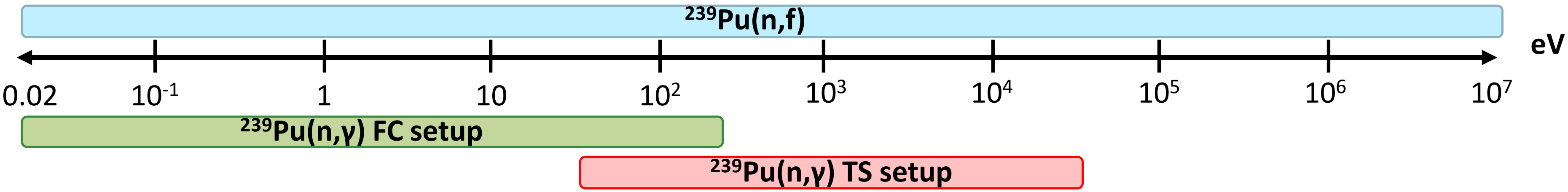




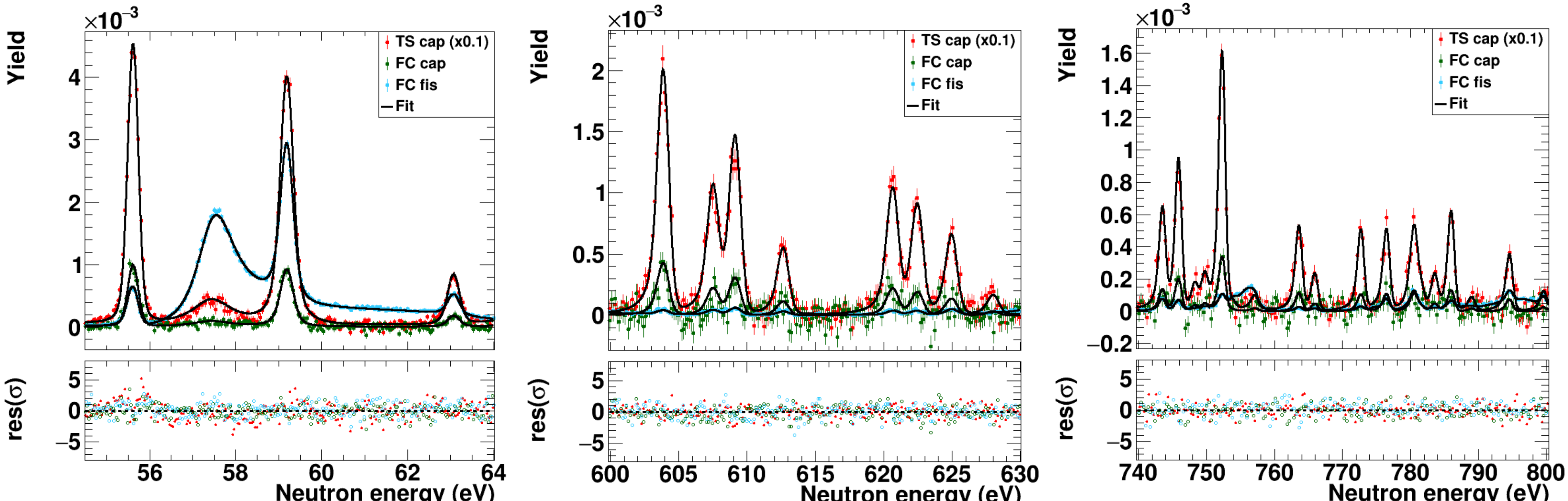
~100 mg

3.2 ^{239}Pu capture results

- The **results** from the **FC setup** was **used to subtract** the γ-ray **fission background** in the **TS setup**.
- **Resonance parameters (RPs)** were extracted with SAMMY by fitting FC fission, FC capture and TS capture data. The **final RPs set combines** the results from FC and TS.

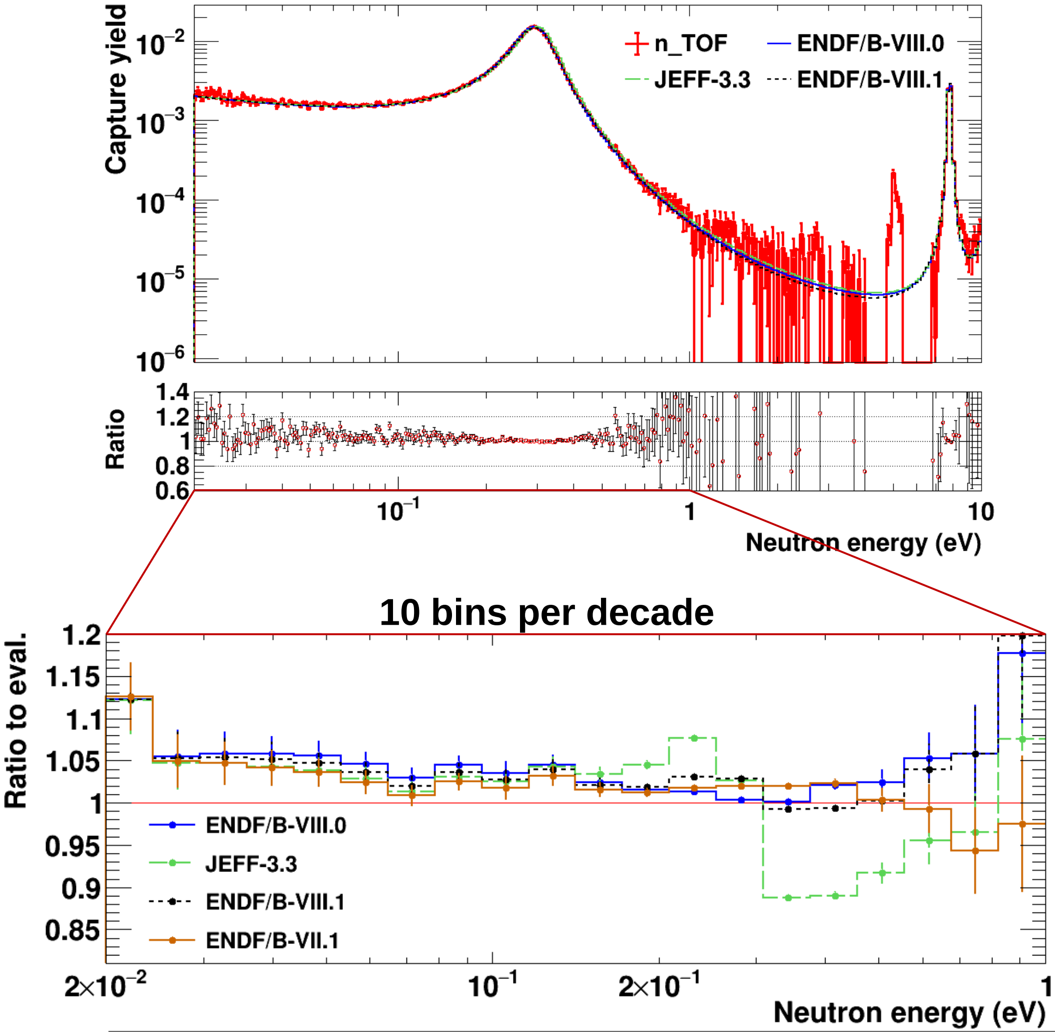


Examples of the SAMMY fit



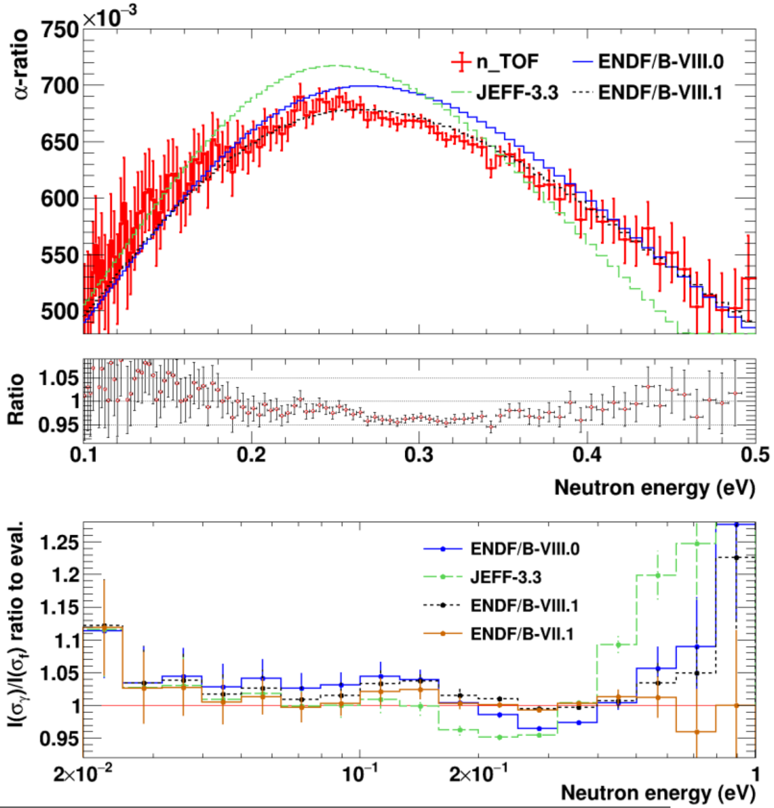
3.2 ^{239}Pu(n,γ) vs. evaluations

- Around the **first, large resonance (@0.3 eV)**, relevant for thermal reactor calculations, the n_TOF data exhibit **excellent agreement with ENDF/B-VIII.0**, and the **largest differences with JEFF-3.3**.



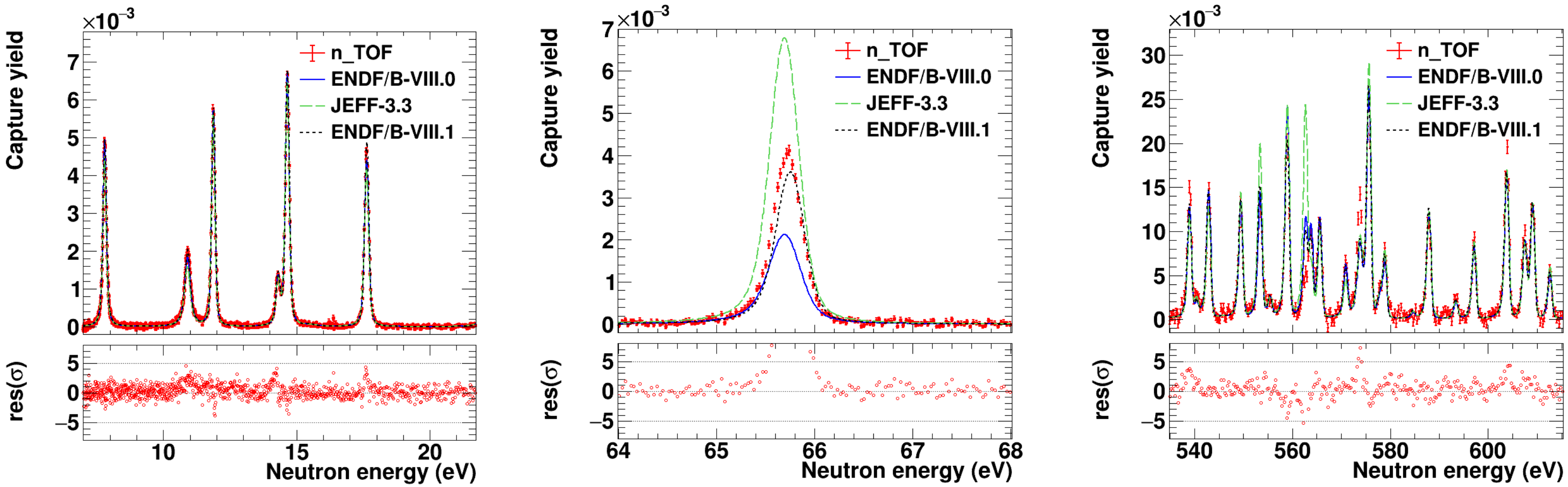
^{239}Pu α-ratio (capture-to-fission)

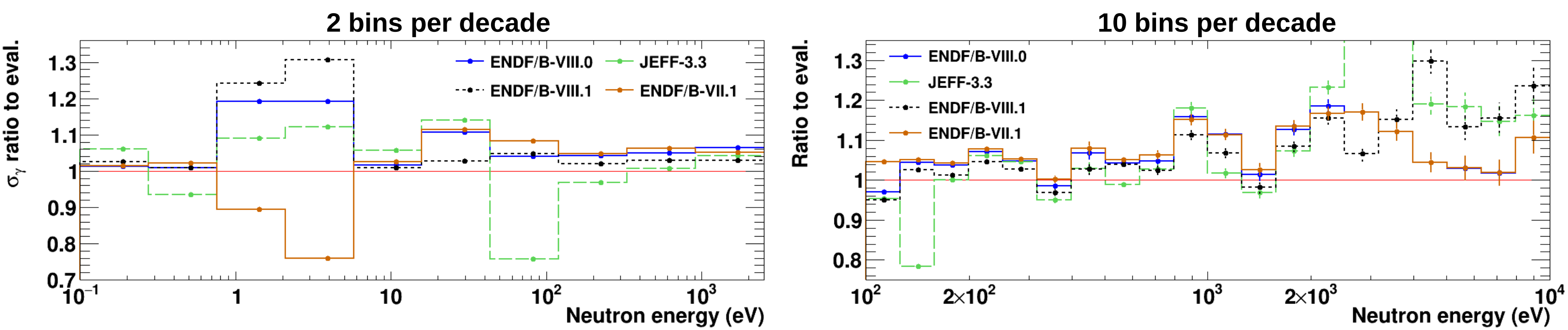
- Larger statistical uncertainties.
- **Better agreement with E/B-VIII.1**, and **3-4% below the other libraries**. This deviation is driven by the difference in the fission cross section.



3.2 $^{239}Pu(n,\gamma)$ vs. evaluations

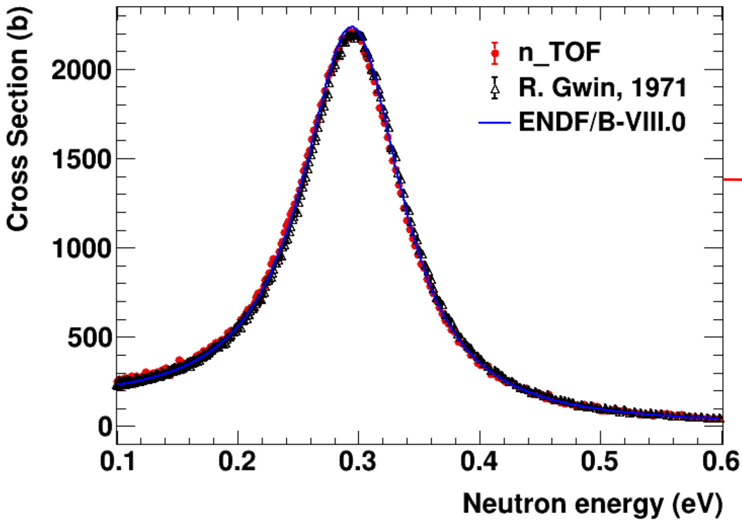
- **10-100 eV**, big discrepancies among evaluations. n_TOF closer to ENDF/B-VIII.1
- **E_n>100 eV**: n_TOF systematically ~5% above evaluations. Larger diffs. above 1~keV, closer to E/B-VII.1.





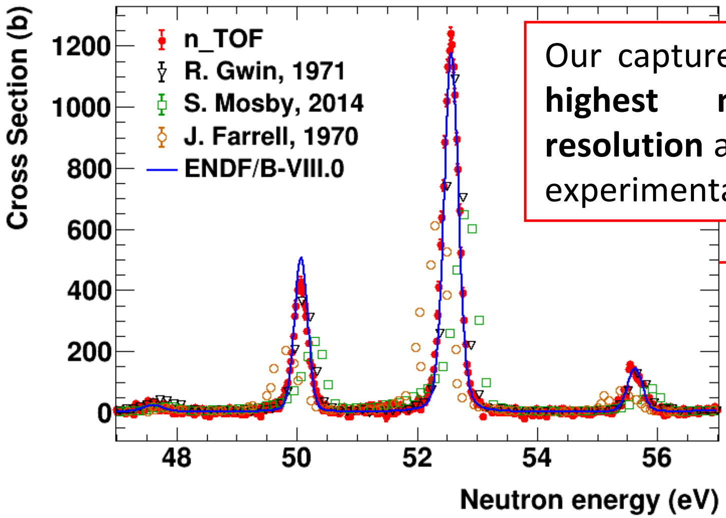
3.2 ^{239}Pu(n,γ) vs. previous data

- **Scarcity of high-resolution ^{239}Pu(n,γ)** cross-section measurements. Two datasets in EXFOR (Gwin 1971, Mosby 2014/2018) mainly used for all evaluations.

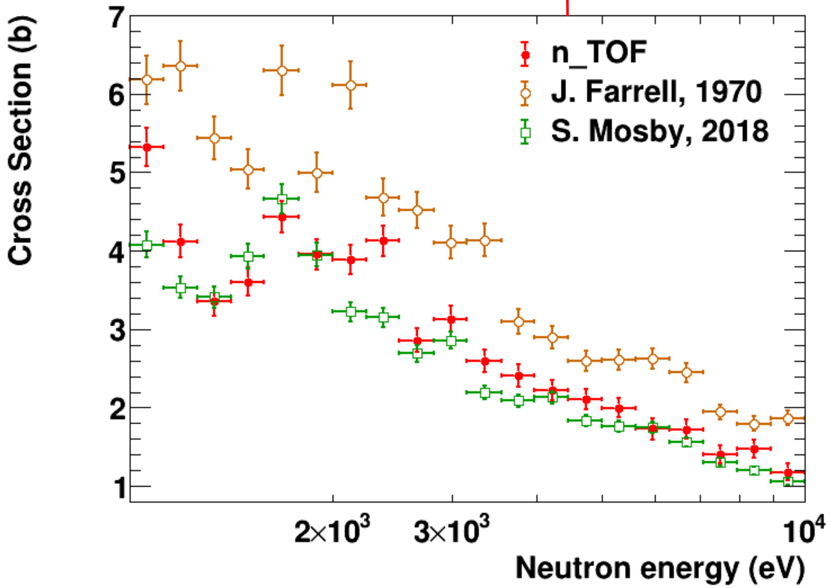


@0.3 eV resonance: good compatibility with Gwin 1971 data.

At higher neutron energies, **from 1 keV to 10 keV** (TS setup), the n_TOF cross section integrals generally stays **slightly above Mosby 2018** data.



Our capture data exhibit **the highest neutron energy resolution** among the existing experimental data.



4

Conclusions

4. Conclusions

- **The data analysis of the 2022 ^{239}Pu experimental campaign** at n_TOF has been completed, including:
  - Data reduction of fission and capture data and extraction of yields.
  - SAMMY fit of fission and capture yields, to obtain a final, combined set of resonance parameters.
- The measured **^{239}Pu(n,f) cross-section** show excellent results in terms of low uncertainties, high energy resolution, and an **unprecedented neutron energy coverage** from 20 meV to 10 MeV with a **single measurement**.
- **The ^{239}Pu capture cross-section measured from 0.1 eV to 10 keV,** improving energy **resolution** of previous datasets with **contained systematic uncertainties** (2-5% with 5 bins per decade). Significant differences observed with the latest nuclear data libraries, suggesting the revision of some resonances in future evaluations.
- Fission paper close to approval for publication.
- Capture paper on *writing* stage.

Acknowledgments

- This project has received funding from the **Euratom** research and training programme 2011-2018 under grant agreement No 847595 (**ARIEL**)

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- This activity is part of the scientific program approved by the European Commission *H2020* ***Supplying Accurate Nuclear Data for energy and non-energy Applications* – SANDA** project (WP2, Task 2).

SANDA

erc European Research Council Established by the European Commission

- **2021-1-RD EUFRAT-GELINA** project funding for the stay at JRC-Geel.

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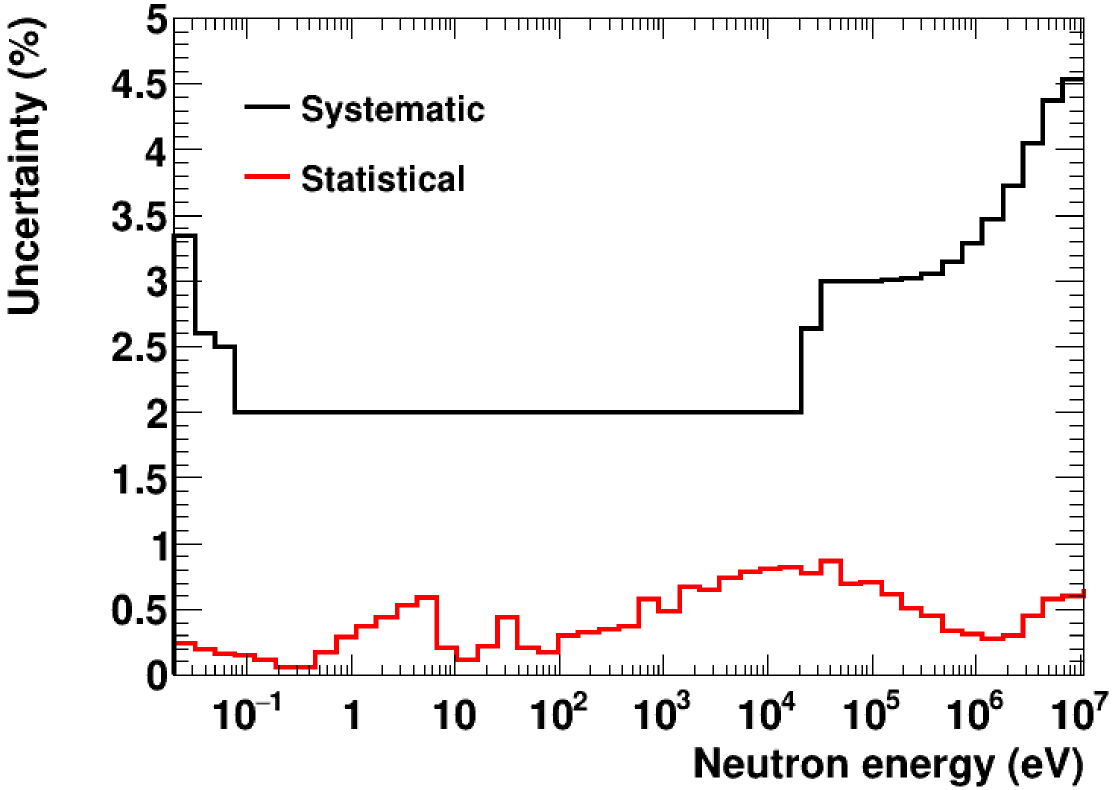
Standards for Nuclear Safety, Security and Safeguards

- **Spanish national projects** PGC2018-096717-B-C21, PID2021-123100NB-I00 and PDC2021-120828-I00.


THANK YOU!

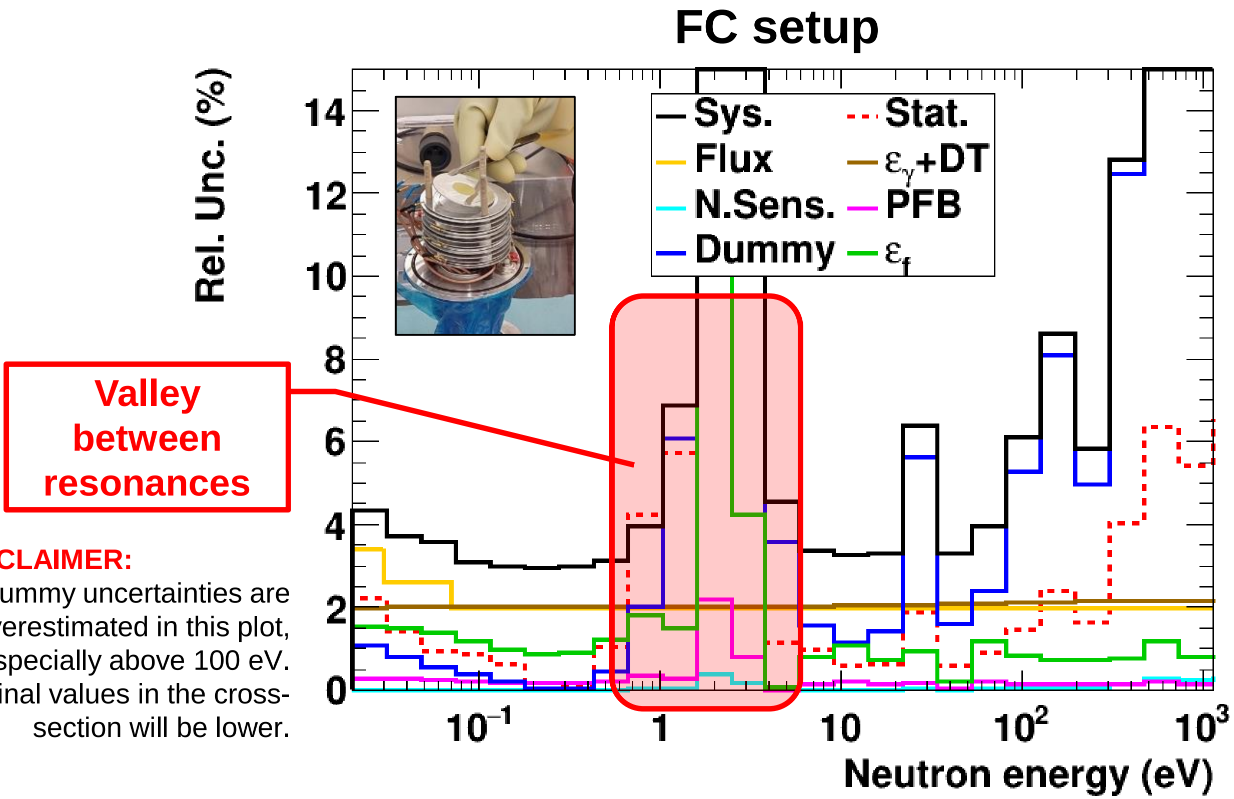
Extra slides

4.1 ^{239}Pu(n,f) uncertainties



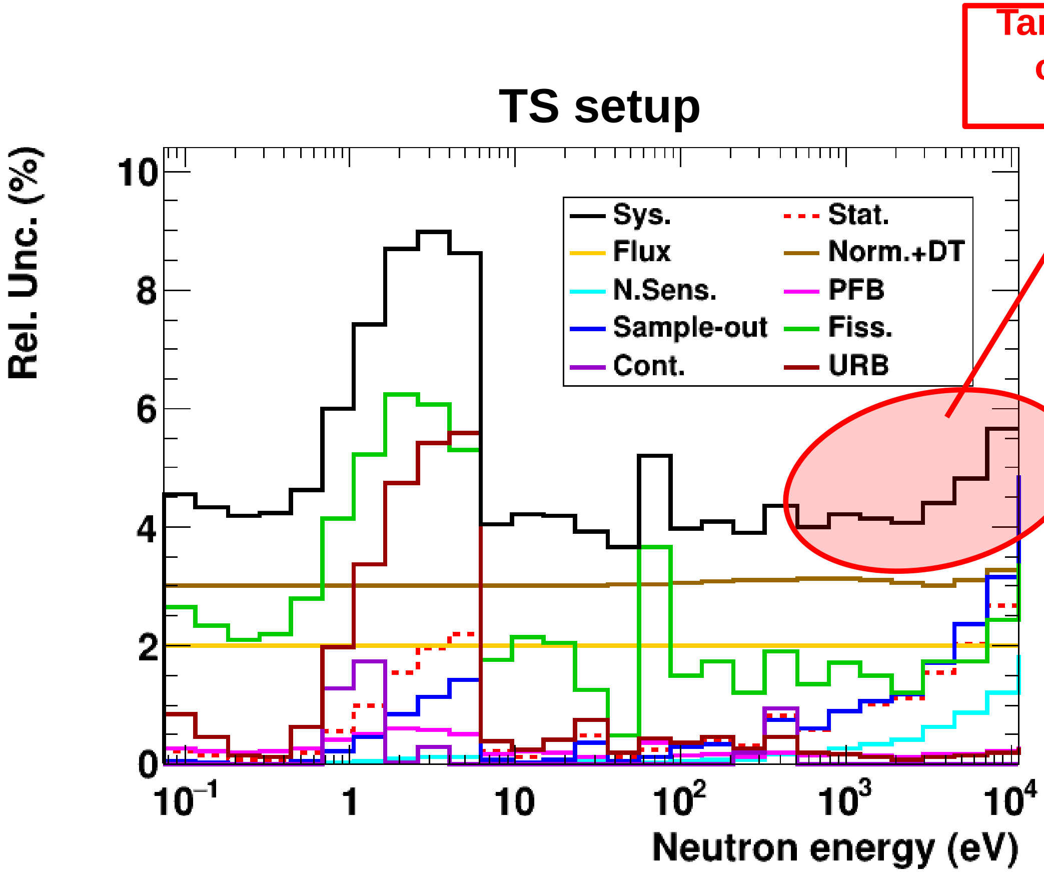
5 bins per decade

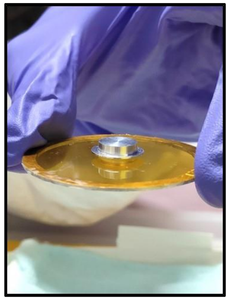
4.1 ^{239}Pu(n,g) uncertainties



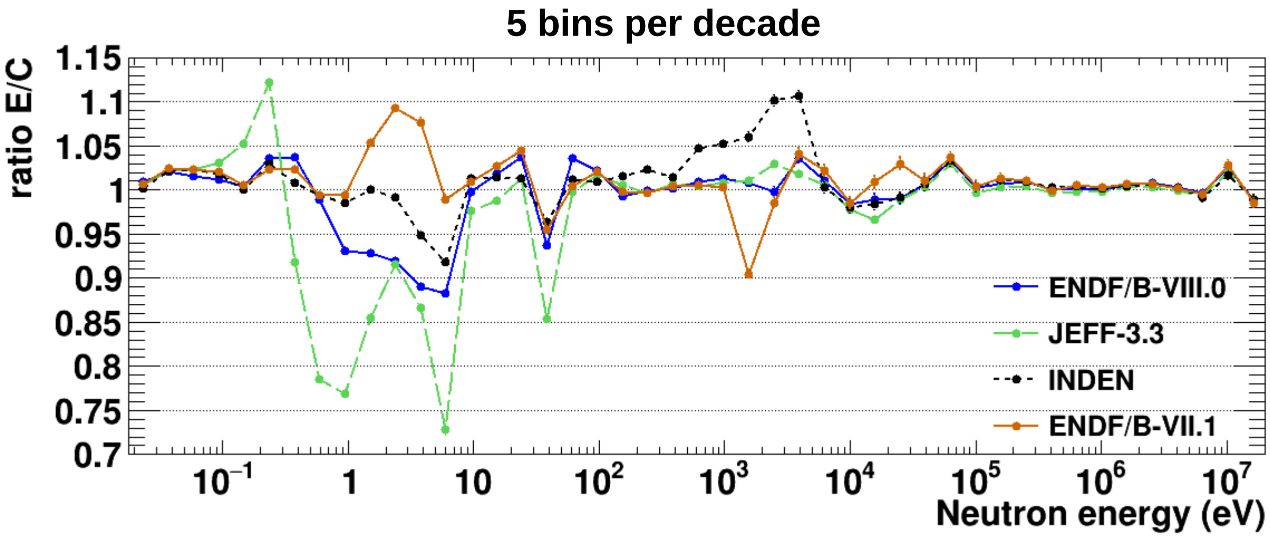
DISCLAIMER:
Dummy uncertainties are overestimated in this plot, especially above 100 eV. Final values in the cross-section will be lower.

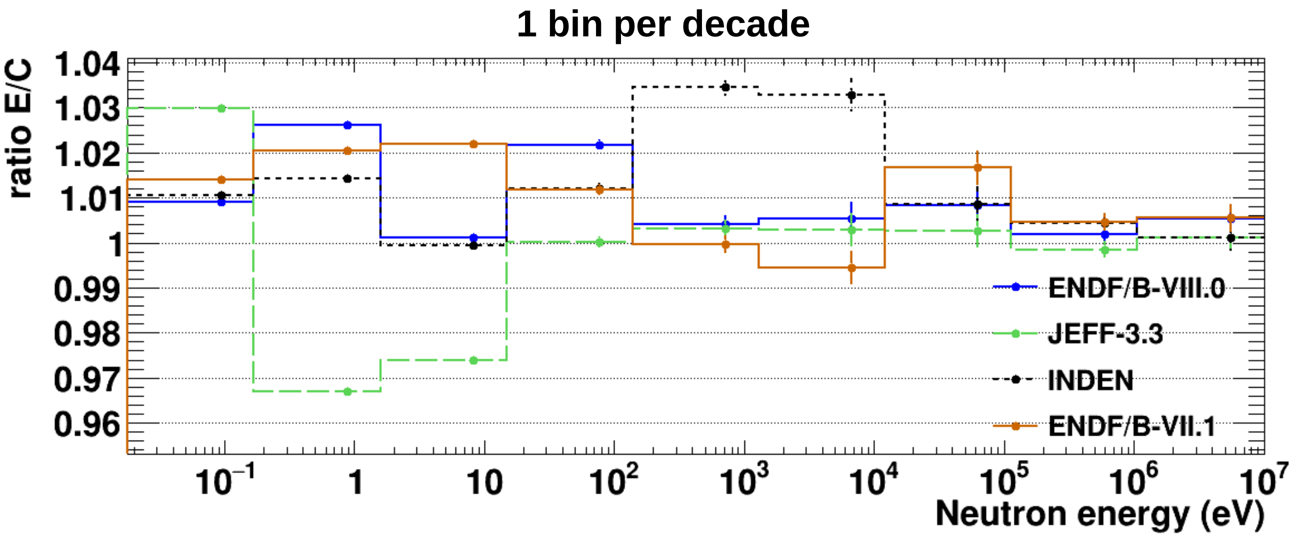
4.1 ^{239}Pu(n,g) uncertainties





4.1 ^{239}Pu(n,f) yield compared to evaluations





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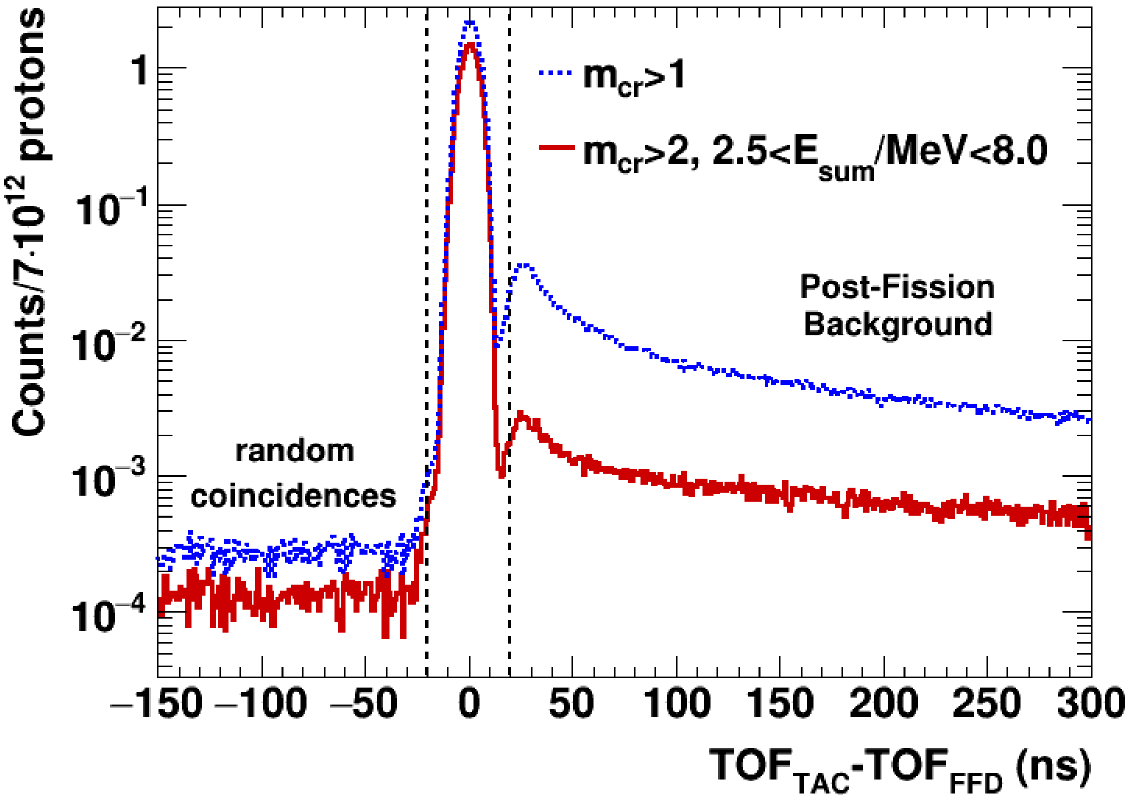
3.3 Coincidence analysis

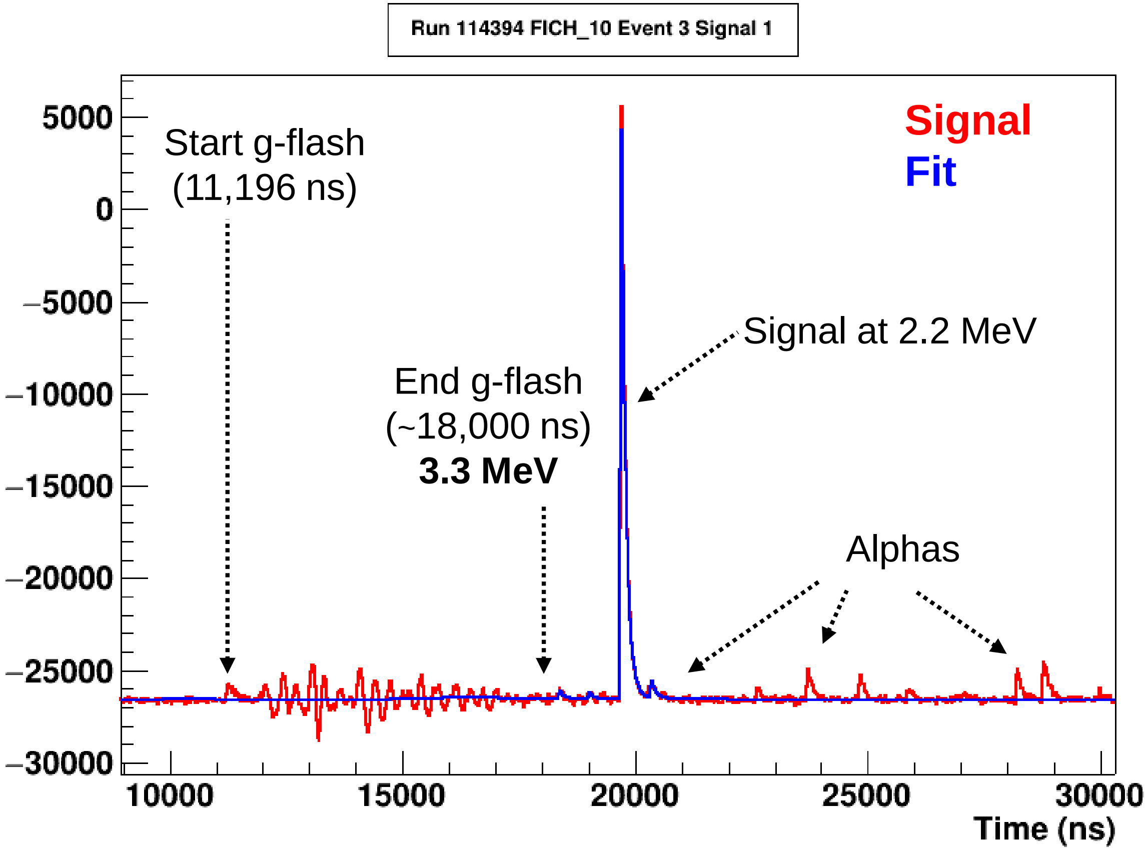
- **Time coincidences** between **TAC events** and **fission chamber (FICH) signals** in the energy region close to the 0.3 eV ^{239}Pu resonance.
- **Coincidence window (-20,+20) ns.**



Max. E_n in fission yield

Inspecting the data buffers, we can estimate the width of the gamma flash, thus obtaining the maximum valid neutron energy for the fission yield that we could potentially reach.

Plot taken from file run114394_0_s1.raw.finished. The Tflash has been obtained from Baf2 #18 from the same pulse. TOFD = 185.59 m.



According to this, we could measure fission without being affected by the gamma-flash **up to ~3 MeV**.

Targets description

Number of electronic output from preamplifiers	Target position in the FC chamber	Pu-239 samples			
		TP number	Activity [μg/cm^2]	Mass [μg]	Areal density [μg/cm^2]
6	1	2020-006-15	2.24E+06	975	310
1	2	2020-006-02	2.22E+06	965	307
7	3	2020-006-04	2.20E+06	959	305
2	4	2020-006-06	2.09E+06	911	290
8	5	2020-006-14	2.81E+05	122	39
3	6	2020-006-07	1.94E+06	844	268
9	7	2020-006-08	2.19E+06	953	303
4	8	2020-006-10	2.11E+06	920	293
10	9	2020-006-12	2.09E+06	912	290
5	10	2020-006-13	2.25E+06	982	312

Targets description

CLOSER TO NEUTRON SOURCES

Target position-10

Target position-9
Target position-8

Target position-7
Target position-6

Target position-5
Target position-4

Target position-3
Target position-2

Target position-1