

Proposal: measurement of $^{239}\text{Pu}(n,\gamma)$ and α -ratio at EAR1 with TAC + fission detectors

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WP2. New nuclear data measurements for energy and non-energy applications.

Subtask 2.2.1. Capture measurements of fissile isotopes. Collaboration between CIEMAT, University of Lodz and JRC-Geel

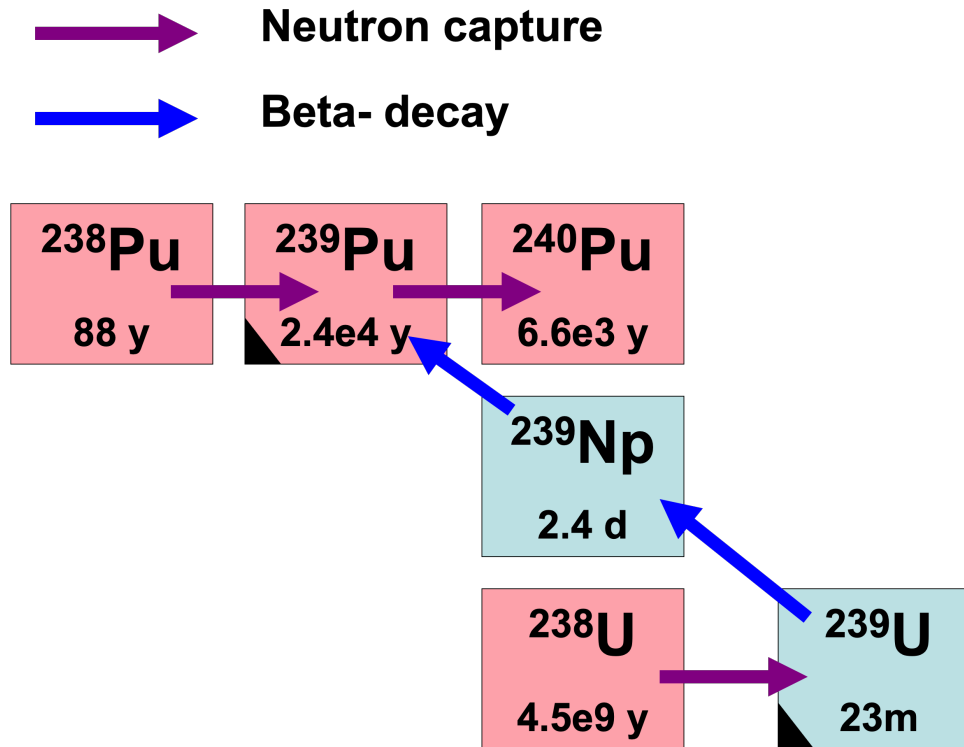


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y Tecnológicas



^{239}Pu production in LWR

Standard fresh nuclear fuel for thermal reactors has a 5% ^{235}U and a 95% ^{238}U . ^{239}Pu is produced during the reactor operation mainly by neutron captures in ^{238}U + decays of ^{239}U and ^{239}Np . The ^{239}Pu is fissile and thus its neutron induced fission contributes to the power.



Motivation

^{239}Pu is a very important isotope in actual Light Water Reactors (LWR), contributing up to **30% of the energy produced**.

After ^{238}U and ^{235}U , ^{239}Pu is the most abundant actinide in the spent nuclear fuel (SNF). The SNF contains ~1% mass contribution of ^{239}Pu (60 GWd/tU). On average, ~700 kg are produced every 4.5 years of LWR operation. The uncertainty in this mass is dominated by nuclear data (A. Villacorta, PhD thesis).

^{239}Pu is considered as a **major component of the nuclear waste** and also as a valuable resource. Pu-loaded fuel burning:

- MOX fuel is used in LWRs (France, ...)
- Use of MOX and highly enriched fuels in ^{239}Pu is planned for Fast Reactors (Na-cooled, Pb-cooled, Gas-cooled) and some Small Modular Reactor designs.

^{239}Pu cross section data **are necessary** for improving the simulation capabilities for actual reactors (higher burnups, life extension, burnup credit). For these reasons, more accurate ^{239}Pu capture and fission cross section data are needed and those measurements have been listed in the NEA/OCDE **High Priority Request List**.

Status of the $^{239}\text{Pu}(n,\gamma)$ cross section

Until recently, the only *high resolution* capture measurement in the RRR (with enough data points to perform a reasonable resonance analysis) available in EXFOR is:

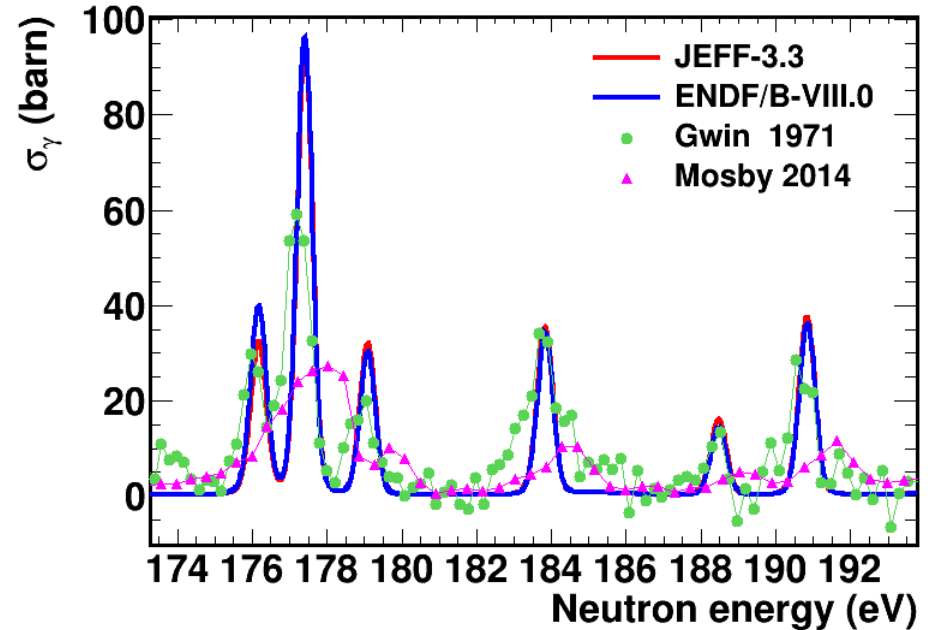
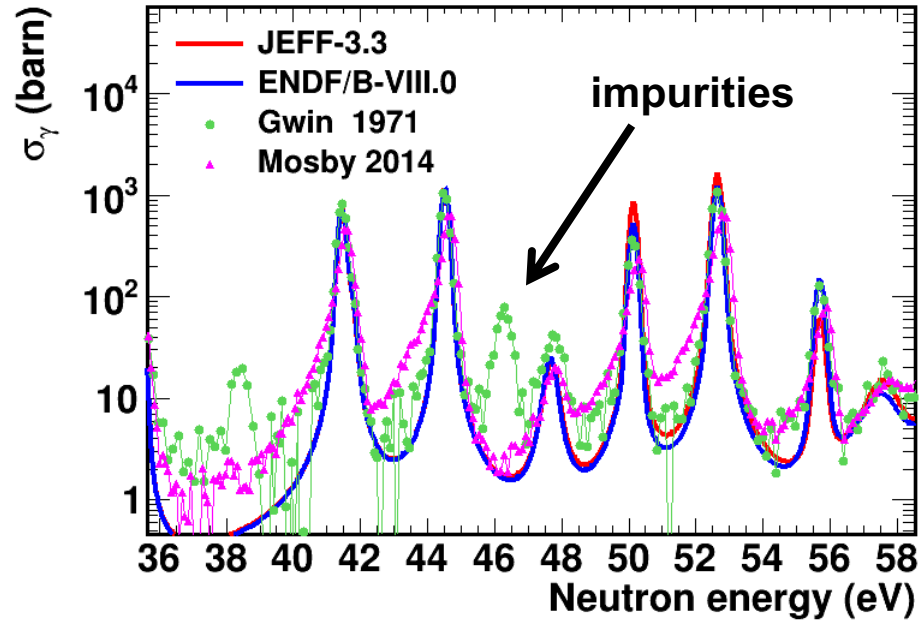
- [Gwin et al., Nucl. Sci. Eng. 45, 25 \(1971\)](#) → (0.02 eV to 30 keV)

There is a new measurement performed at LANSCE, in the energy range between 10 eV and 1.3 MeV (Note: one measurement → three papers).

- [S. Mosby et al., Phys. Rev. C 89, 034610 \(2014\)](#)
- [S. Mosby et al., Phys. Rev. C 97, 041601 \(2018\)](#)
- [S. Mosby et al., Nucl. Data Sheets 148, 312 \(2018\)](#)

The data from Mosby et al. have been normalized to ENDF/B-VII.1 between 17 and 18 eV.

Status of the $^{239}\text{Pu}(n,\gamma)$ cross section data



Goals of the proposal

We propose to measure the **α -ratio, the fission and the capture cross sections** at the n_TOF EAR1, using the **Total Absorption Calorimeter and a (multi target) ionisation chamber** developed at the University of Lodz.

This is a very challenging measurement due to the competing (~5 times larger) **fission γ -ray background** and to the **high α -activity of ^{239}Pu (2 MBq/mg)**.

We will apply the expertise and methodologies developed for the $^{235}\text{U}(n,\gamma)$ measurement and analysis (*Balibrea et al.*, PRC 102, 2020) and perform a measurement with an **improved experimental setup**.

Experimental technique

Fission tagging: γ -rays in coincidence (fission background) and anticoincidence (capture signal) with the fission detector. (*J. Balibrea et al.*, PRC 102, 2020)

$$Y_{\gamma} = \frac{C_{aco,\gamma} - \frac{1 - \epsilon_f^*(E_n)}{\epsilon_f^*(E_n)} C_{tag} - C_{oth,\gamma}}{\epsilon_{\gamma} \phi_N}$$

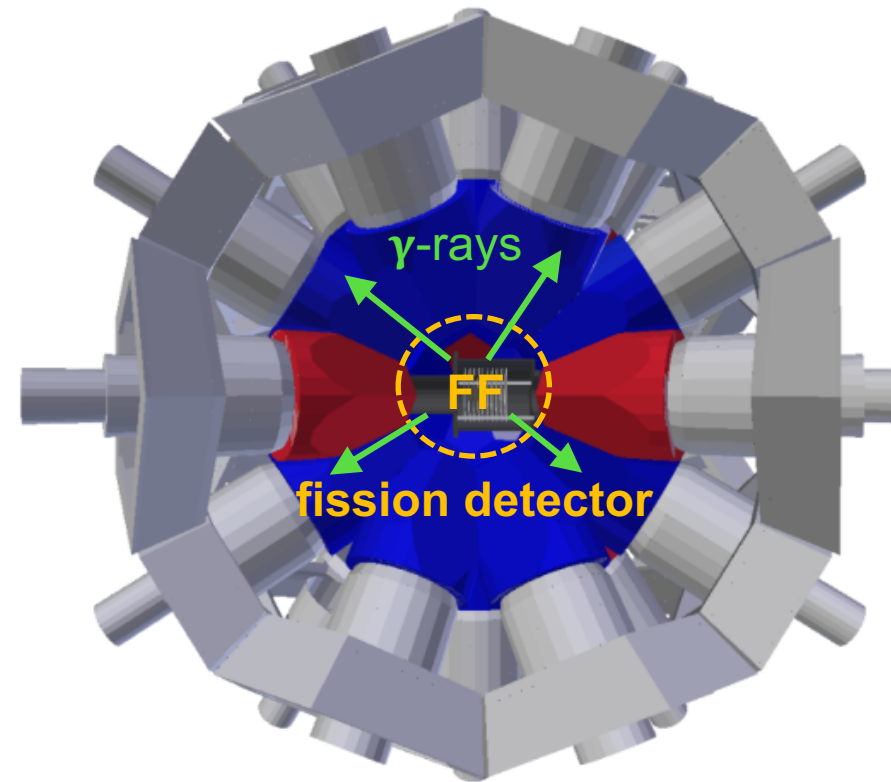
C_{tot} = counts in the TAC.

C_{tag} = counts in the TAC in coincidence with the ionisation chambers.

C_{oth} = background in the TAC.

ϵ_f^* = fission tagging efficiency.

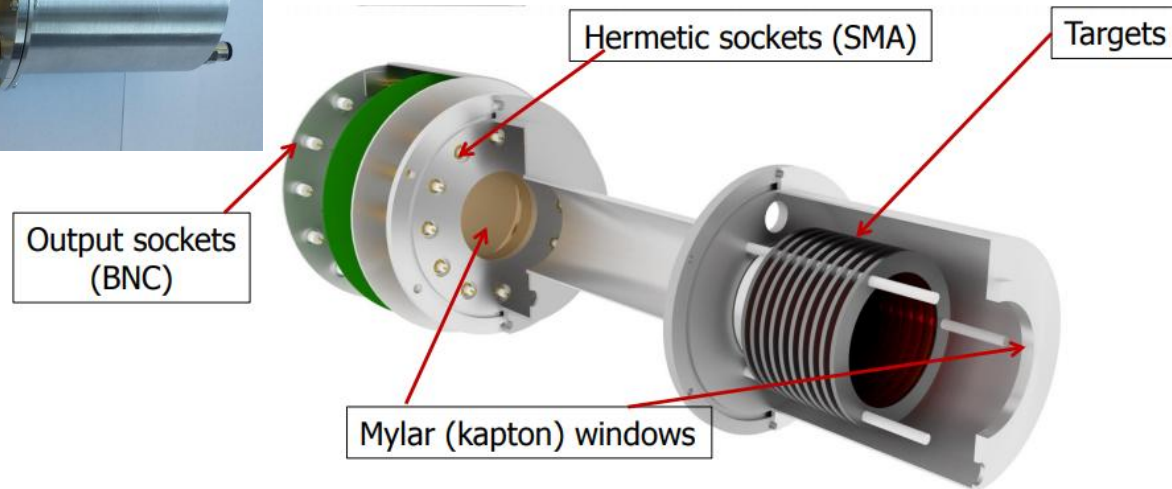
ϵ_{γ} = capture detection efficiency.



The fission chamber

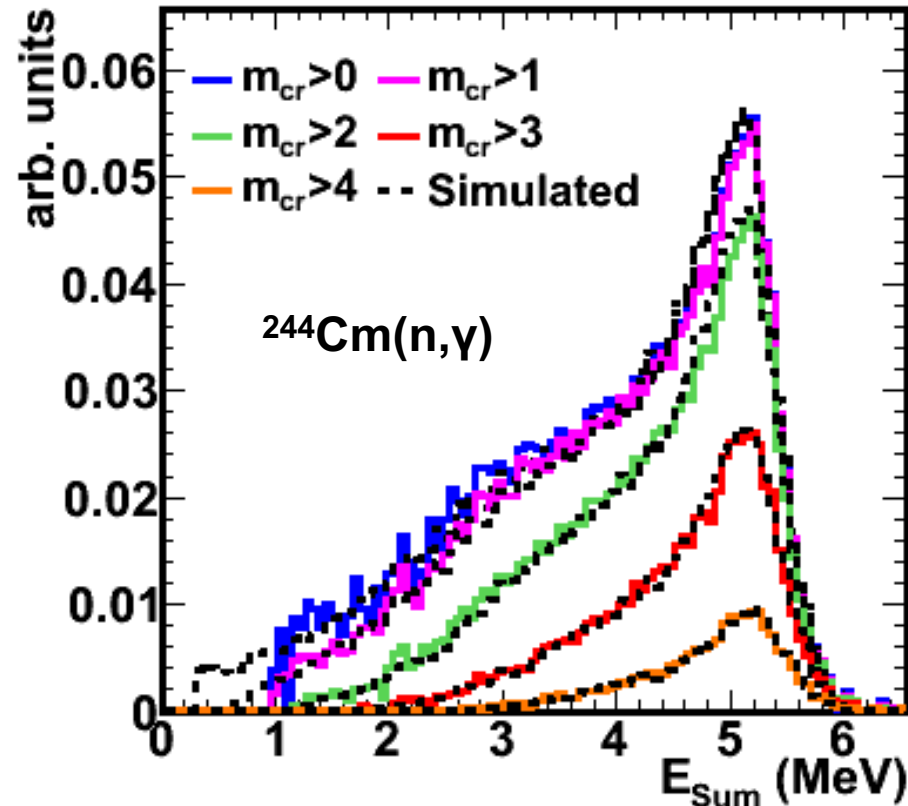
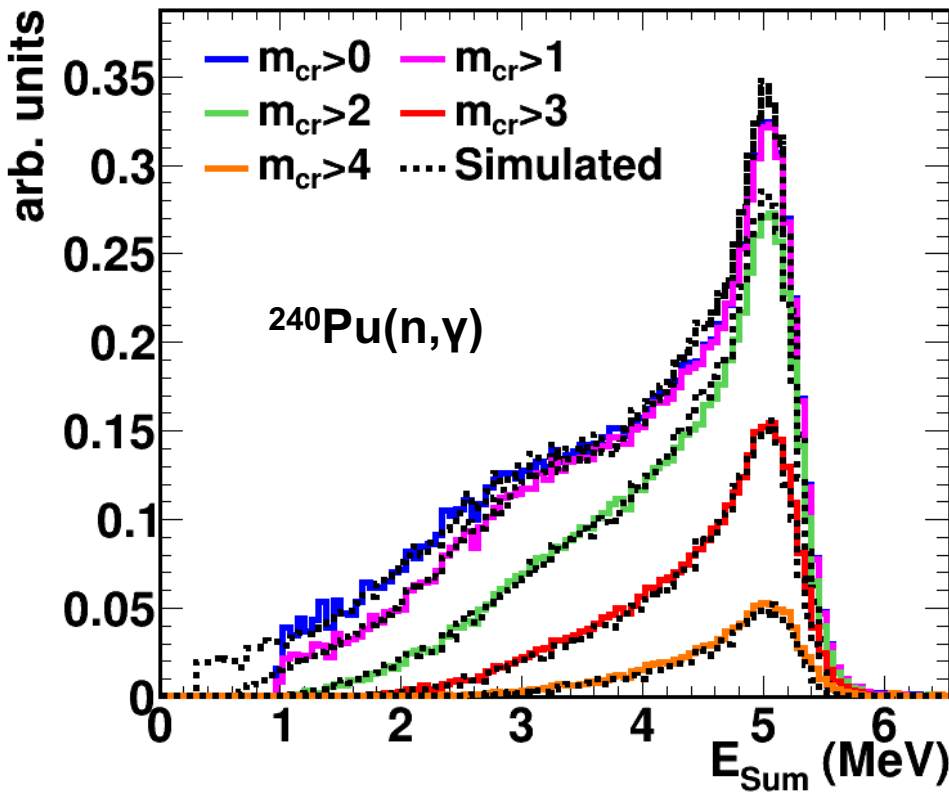
The multi target fission chamber has been designed taking into account the following important characteristics:

1. **Low mass** intercepting the neutron beam, to minimize the background in the TAC due to captures and elastically scattered neutrons.
2. Good **discrimination** between **alphas (2 MBq/mg)** and **fission fragments**.
3. Small **pile-up** effects.



The capture efficiency

The simulation of realistic capture cascades using **NuDEX** (*E. Mendoza et al., ND2019 EPJ Conf. Proceedings*) allows to determine the capture efficiency with a high accuracy ($\sim 1\%$).



Comparison between experimental and simulated deposited energy spectra in the TAC due to $^{240}\text{Pu}(n,\gamma)$ and $^{244}\text{Cm}(n,\gamma)$ cascades, measured in 2017.

The samples

- I. Ten thin ^{239}Pu **samples will be manufactured at JRC-Geel** with a total mass of **~10 mg (1 mg/sample)**.

The samples will be placed inside the fission detector, in the center of the TAC. The limitations come from the signal-to-background ratio.

Overall uncertainties: ~3% below 100 eV and 4-6% between 100 eV and 1keV.

- II. A thick sample, in order to extend the measurement to higher neutron energies.

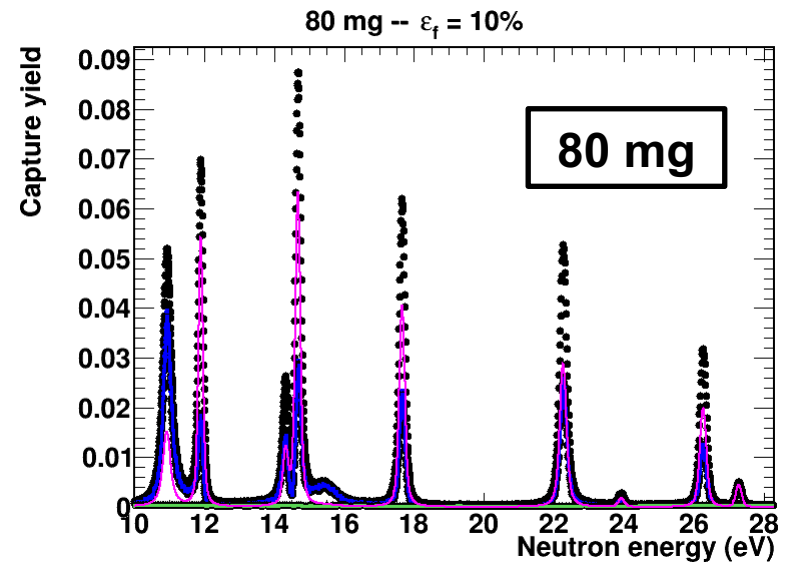
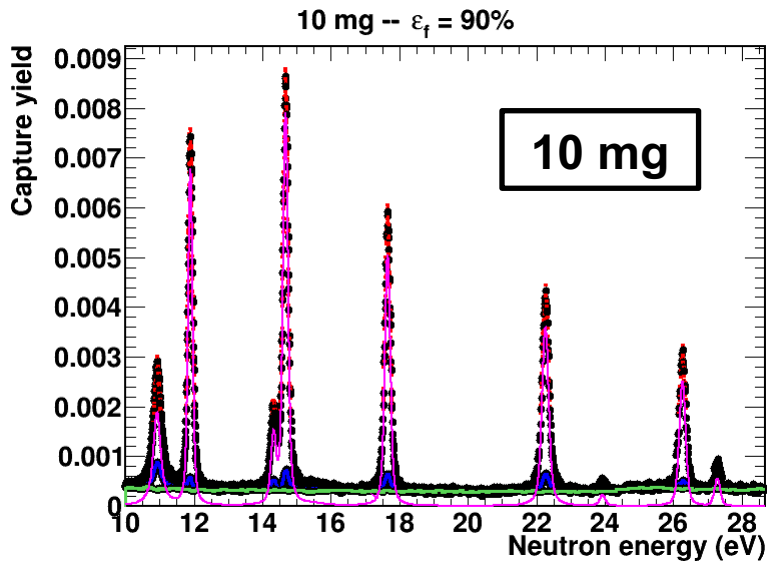
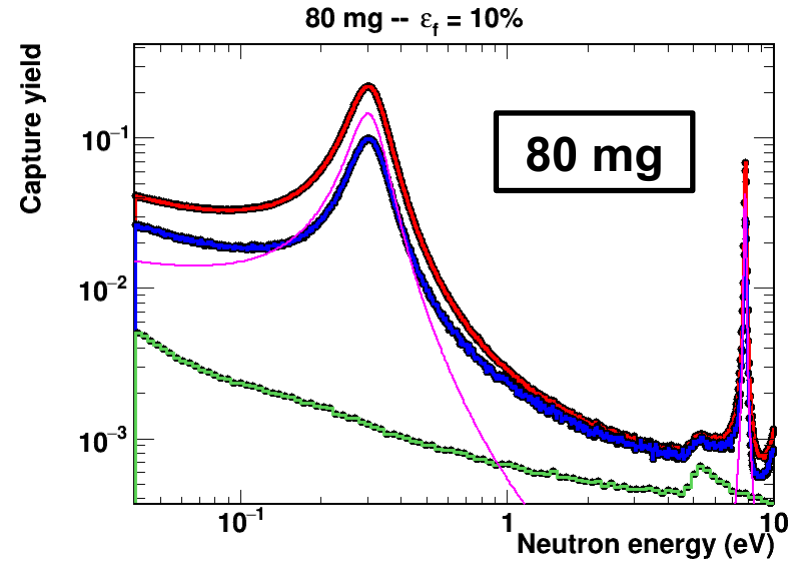
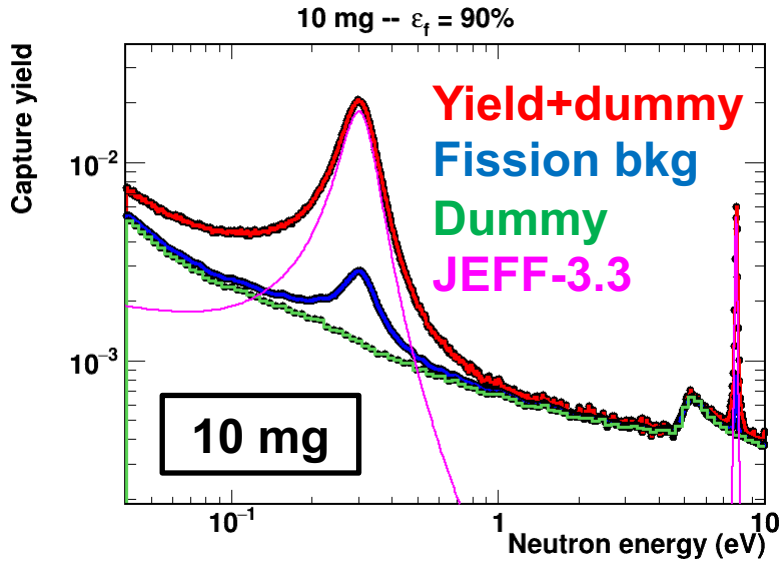
This sample will be measured **without fission detector**. With **~80 mg** we could extend the measurement **up to 10 keV**.

Overall uncertainties: 3-4% between 100 eV and 10 keV.

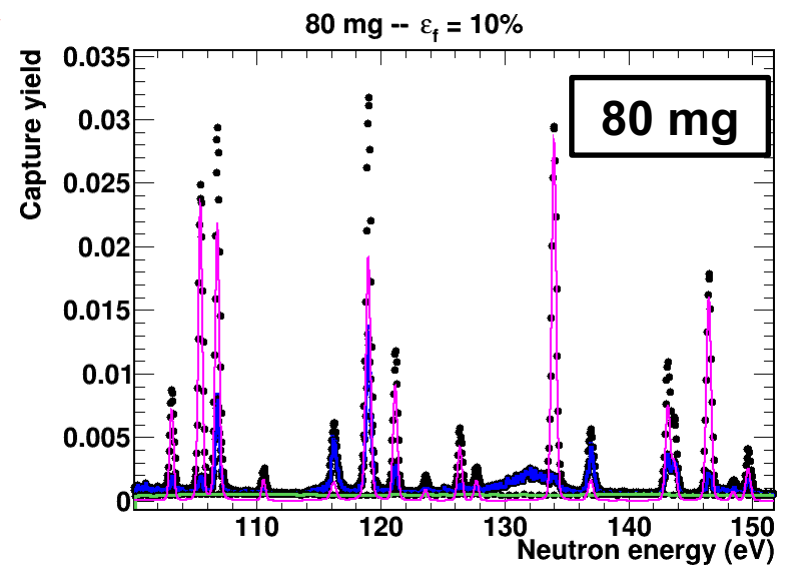
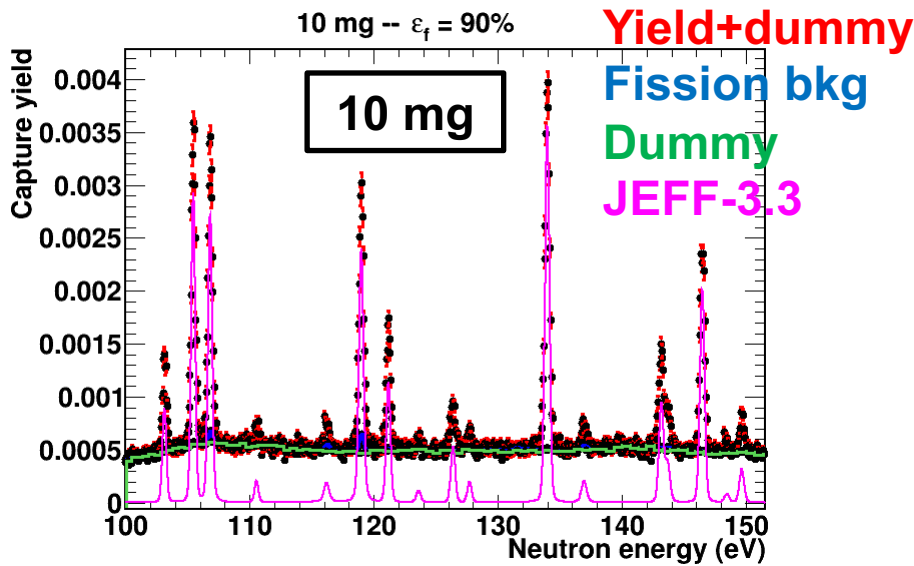
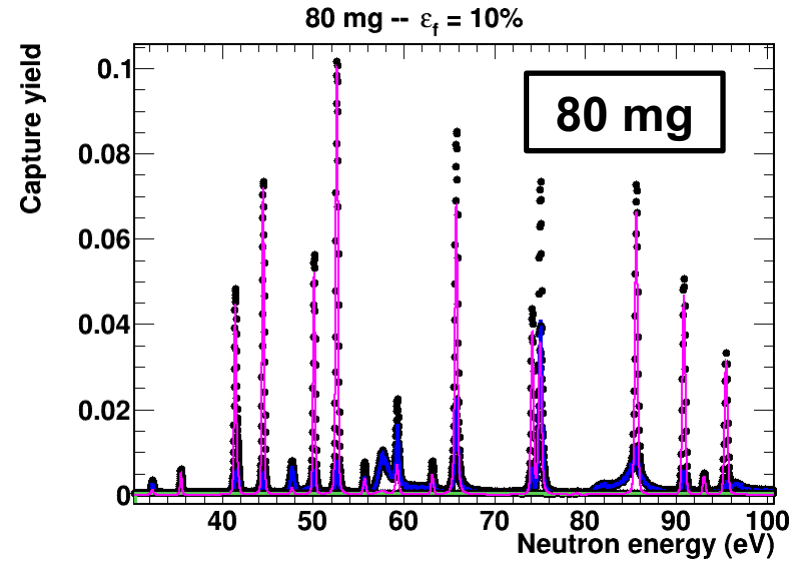
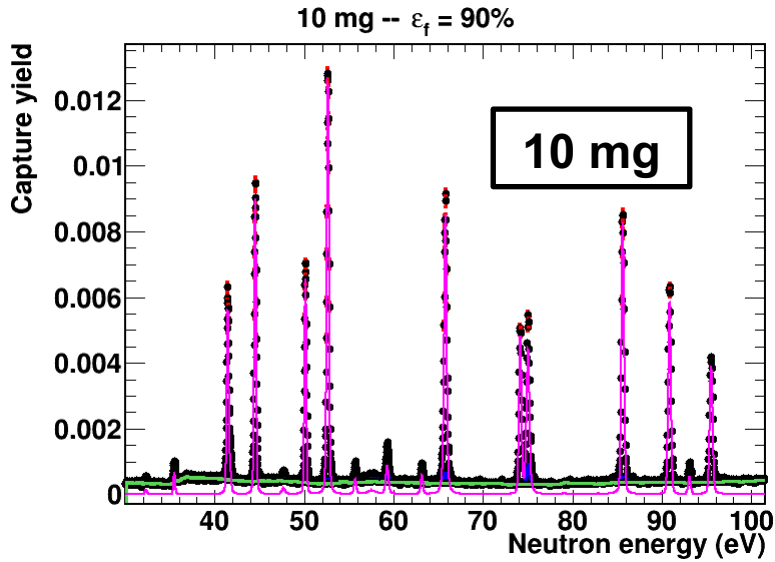
The n_TOF measurement will improve significantly the previously existing data by:

- providing a **capture yield** with an overall uncertainty **~3%** (0.025 eV – 10 keV). Normalisation to the better-known $^{239}\text{Pu}(n,f)$ cross section measured simultaneously.
- Providing an absolute **α -ratio**, thanks to the accurate determination of the fission and capture detection efficiencies. (*J. Balibrea et al.*, PRC 102, 2020)
- Providing data with significantly **better energy resolution**, measured with a flight path 10 times longer than in *Mosby et al.* and *Gwin et al.*, which will improve significantly the resonance analysis.
- **Solving the existing discrepancies** between the recent JEFF-3.3 and ENDF/B-VIII.0 evaluations.
- Providing very valuable information on the **distribution of the γ -rays** cascades emitted in $^{239}\text{Pu}(n,\gamma)$ and $^{239}\text{Pu}(n,f)$ reactions, as has been the case in previous experiments performed with the TAC (**information on photon strength functions**).

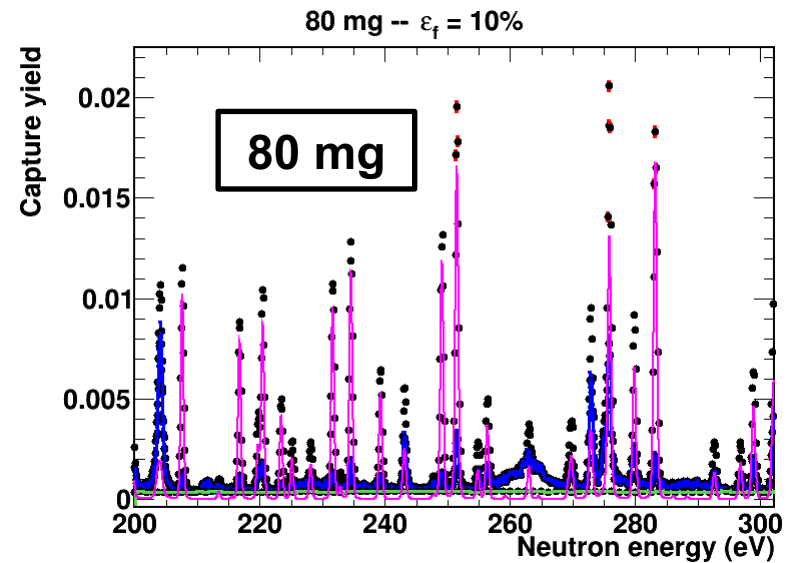
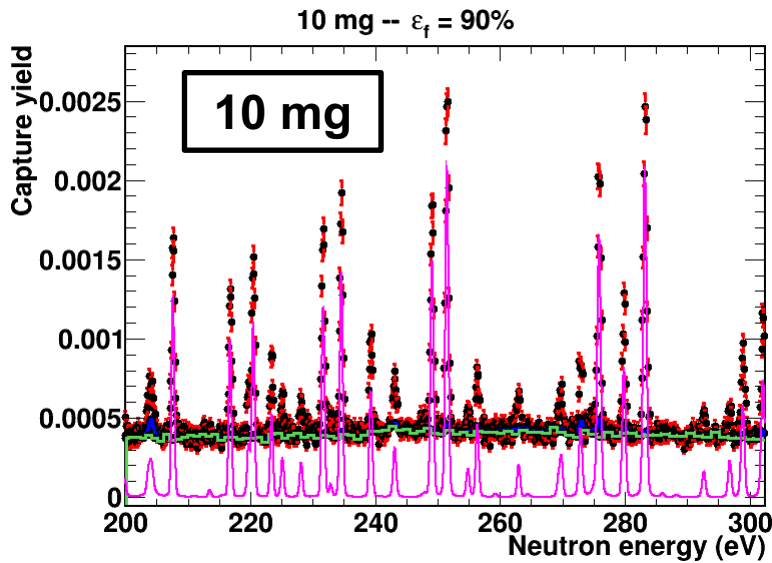
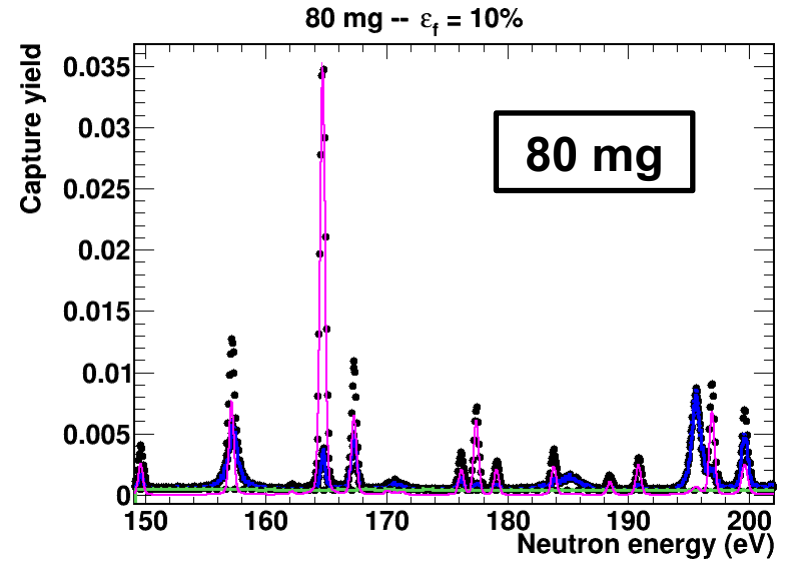
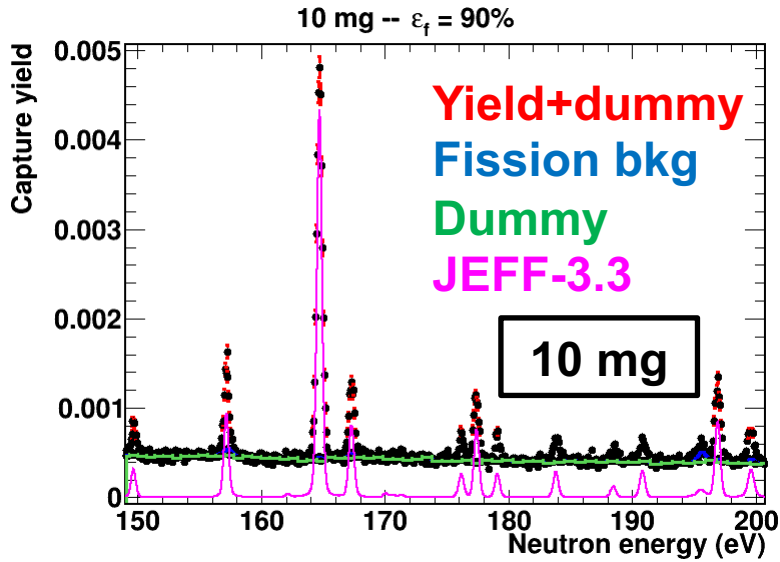
Expected capture yield



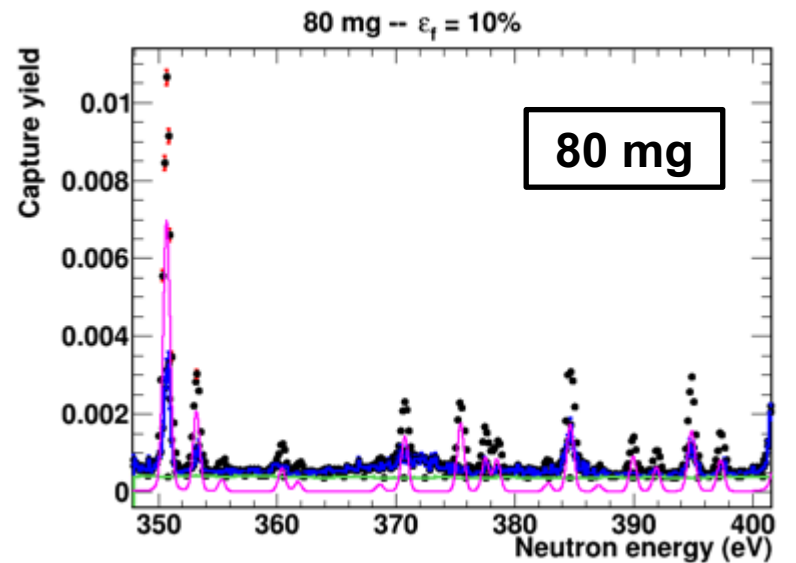
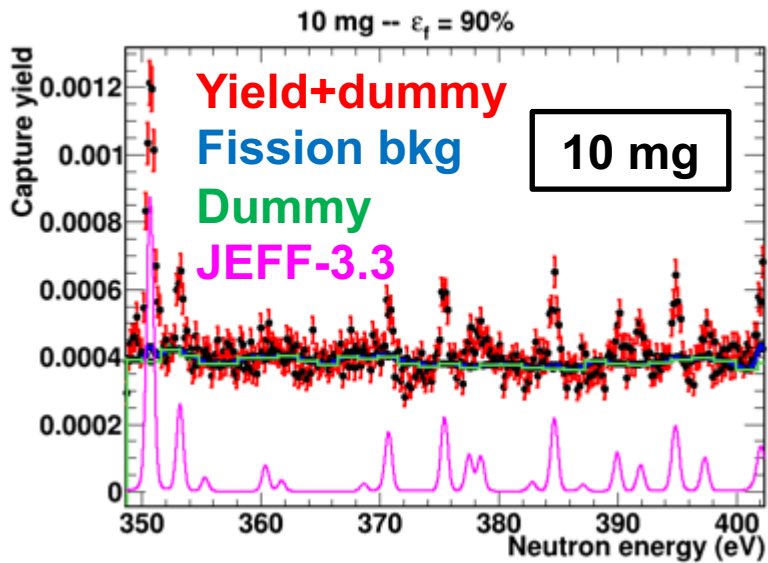
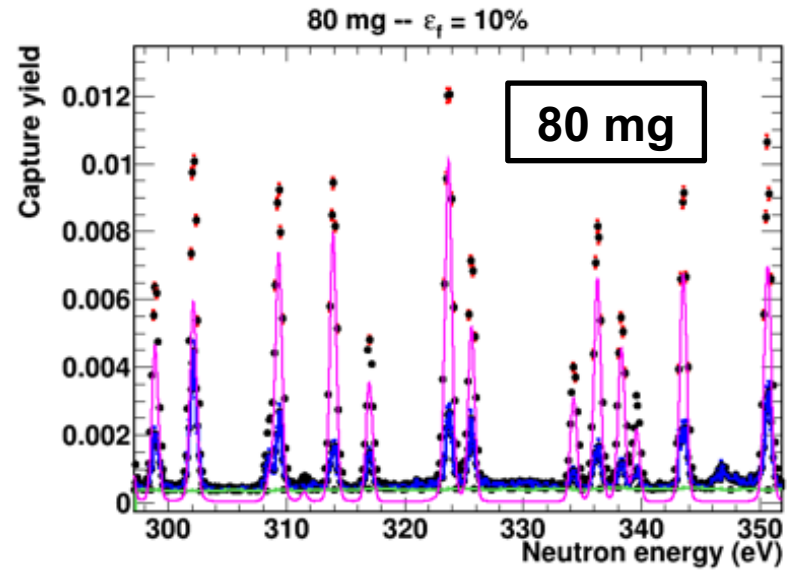
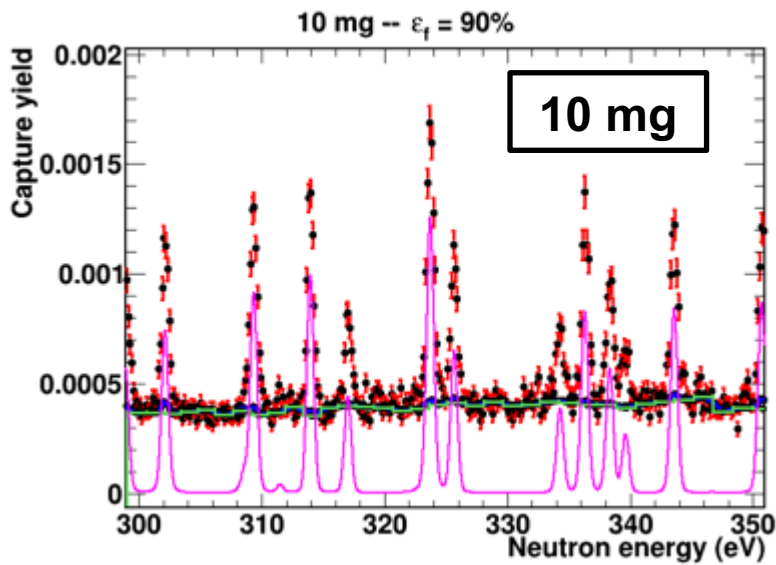
Expected capture yield



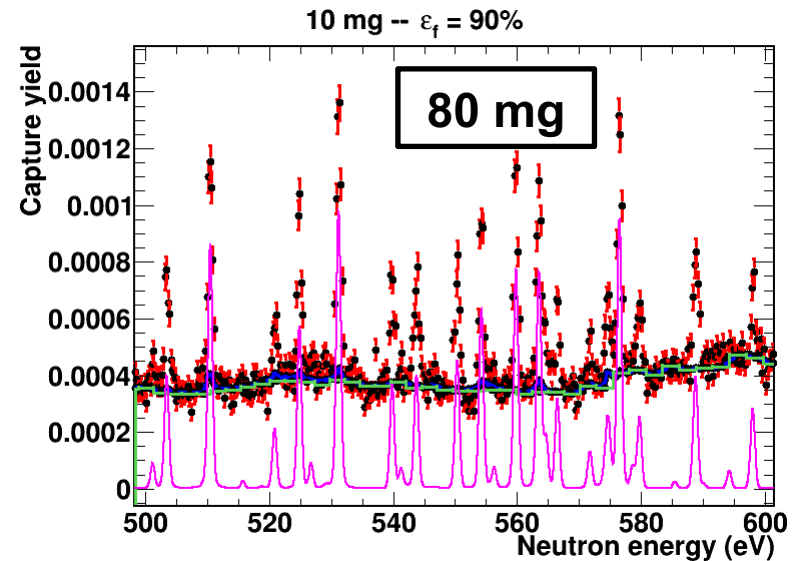
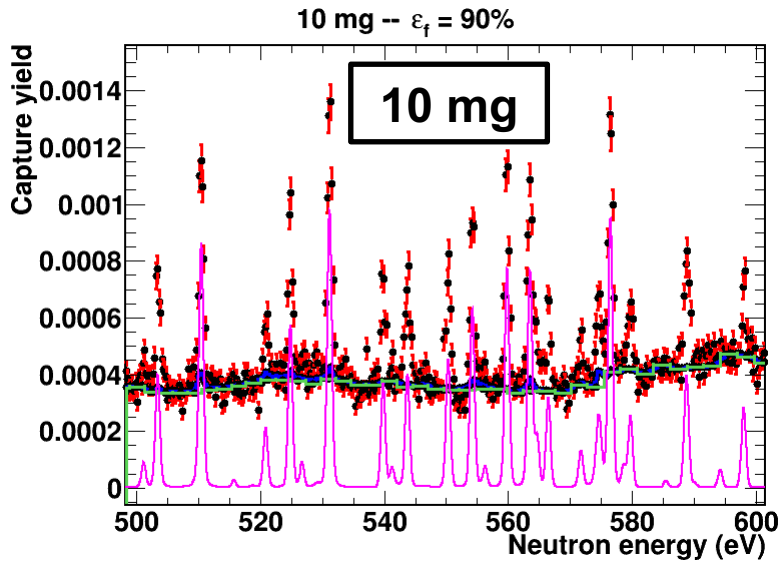
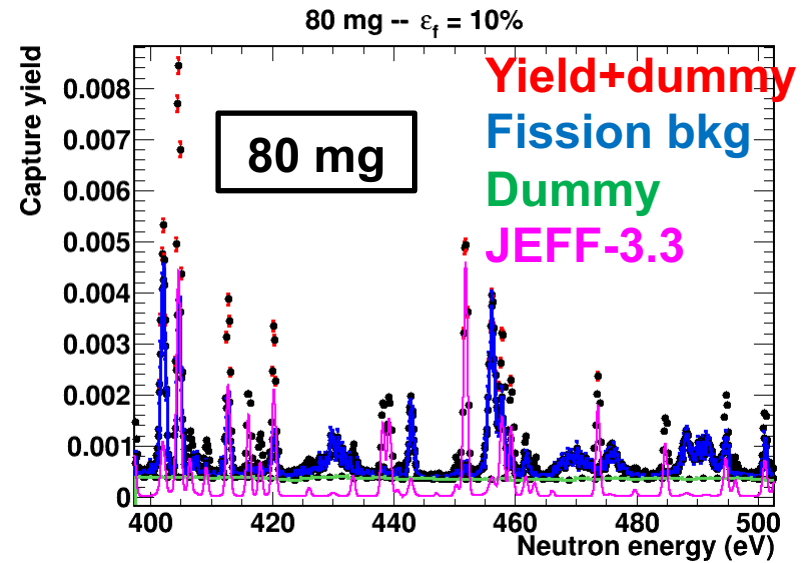
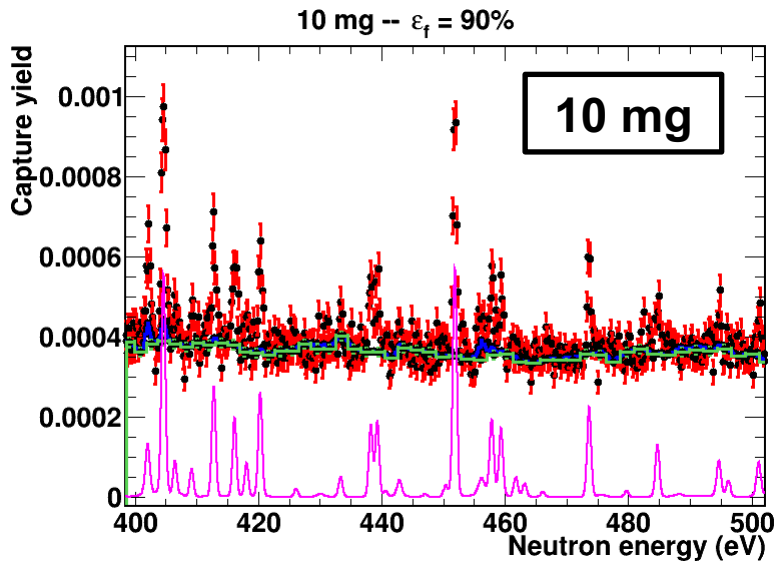
Expected capture yield



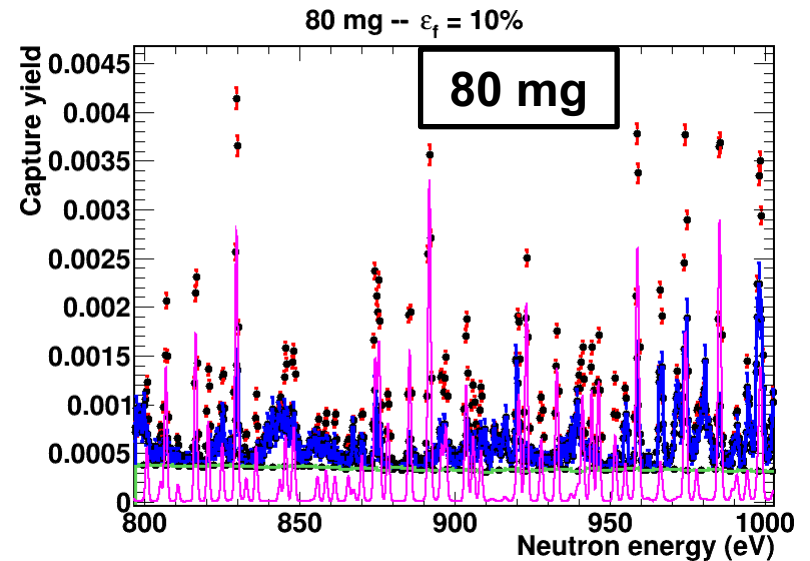
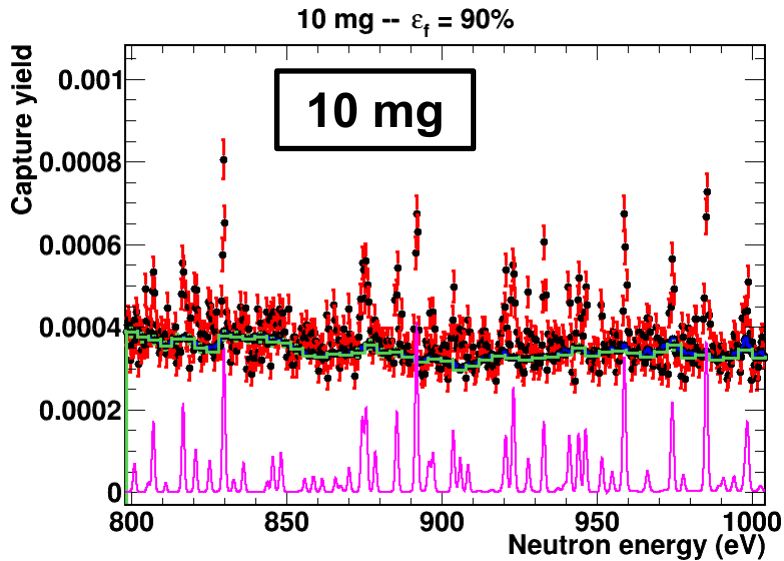
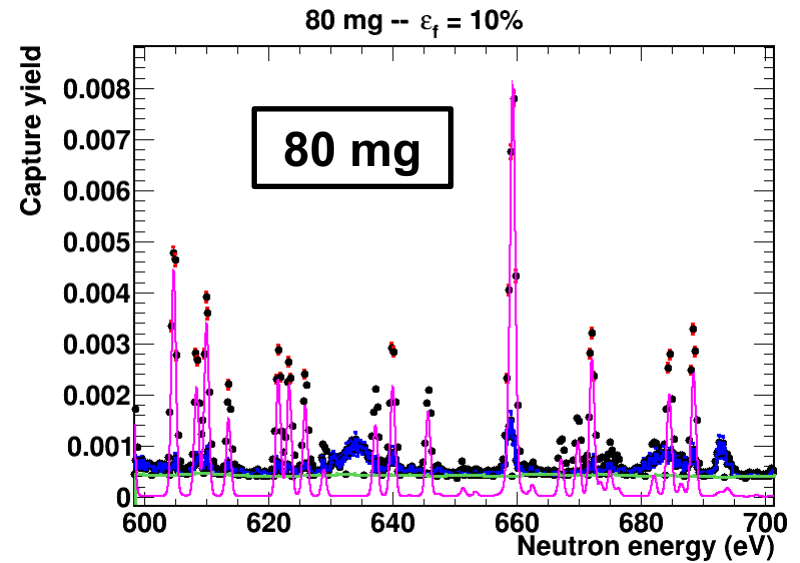
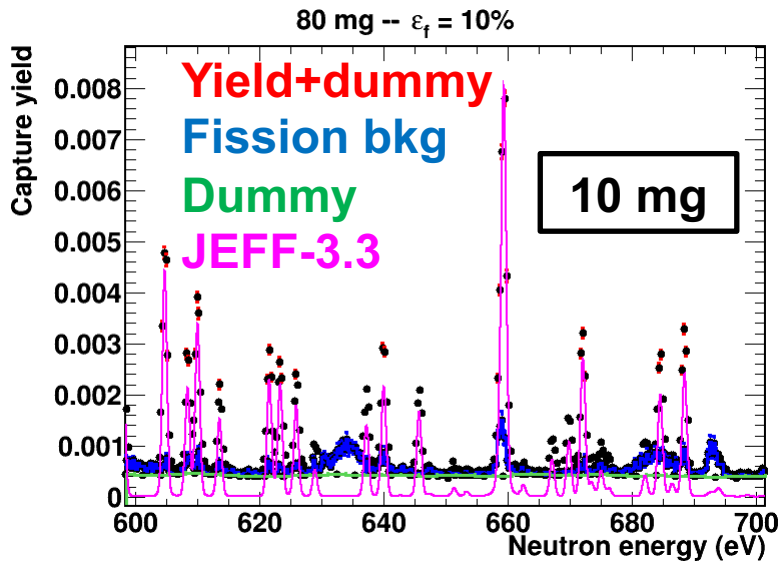
Expected capture yield



Expected capture yield



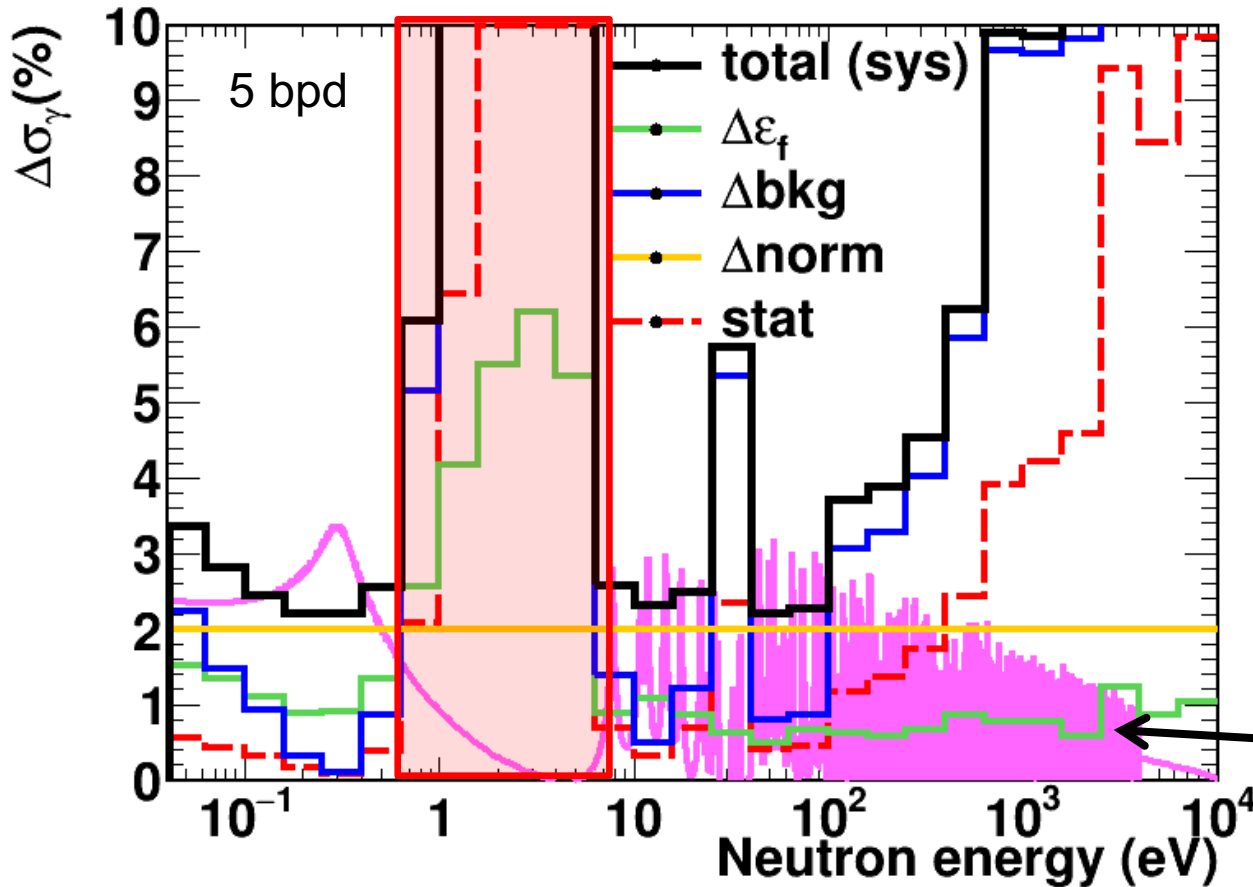
Expected capture yield



Estimation of uncertainties

Valley between resonances

$$\text{total} = \sqrt{(\Delta\varepsilon_f)^2 + (\Delta\text{bkg})^2 + (\Delta\text{norm})^2}$$



$\Delta\varepsilon_f$ → uncertainty in the fission efficiency.

Δbkg → uncertainty in the determination of the beam background.

Δnorm → normalization uncertainty (mainly due to the capture efficiency).

stat → due to counting statistics.

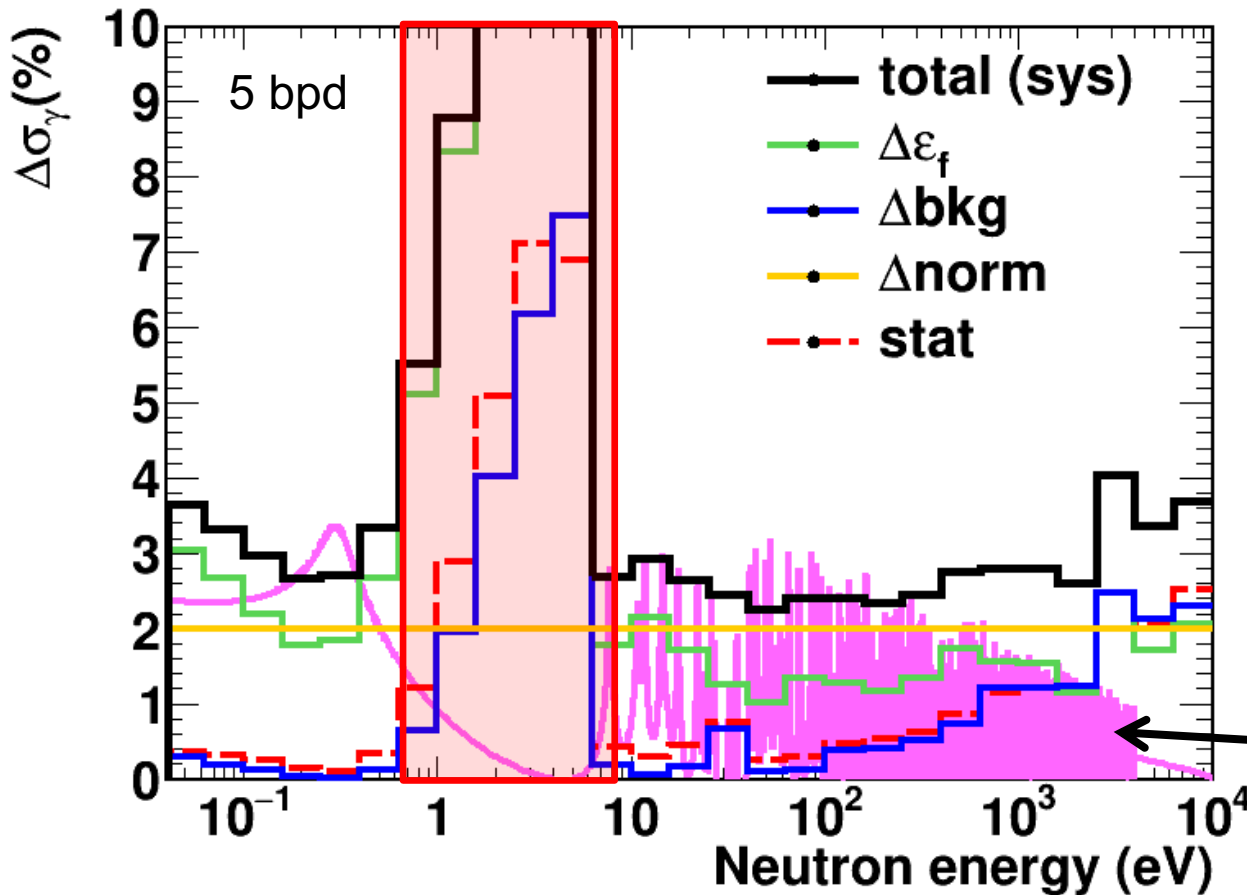
$\text{Log}_{10}(\sigma_\gamma/\text{barn})$
(JEFF-3.3)

Estimated uncertainties in the measured ^{239}Pu capture cross section, using a **10 mg** ^{239}Pu sample.

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(JEFF-3.3)

Estimated uncertainties in the measured ^{239}Pu capture cross section, using a **80 mg** ^{239}Pu sample.

Beam time request

Measurement	Purpose	Protons
^{239}Pu samples (10x1 mg)	(n, γ) cross section measurement (up to 0.5 keV)	$1.2 \cdot 10^{18}$
Dummy chamber (10x1 mg)	Canning related background	$0.8 \cdot 10^{18}$
^{239}Pu sample (50 – 100 mg)	(n, γ) cross section measurement (0.01–10 keV)	$0.8 \cdot 10^{18}$
Dummy (50 – 100 mg)	Canning related background	$0.5 \cdot 10^{18}$
Sample out	Beam related background	$0.5 \cdot 10^{18}$
^{197}Au	Validation of the measurement	$0.5 \cdot 10^{18}$
Graphite	Background due to elastic scattering	$0.3 \cdot 10^{18}$
^{239}Pu without neutron absorber	Determination of the $^{239}\text{Pu}(n,\gamma)$ cascades	$0.4 \cdot 10^{18}$
^{239}Pu (no beam)	Background without beam	0
Total		$5 \cdot 10^{18}$

Conclusions

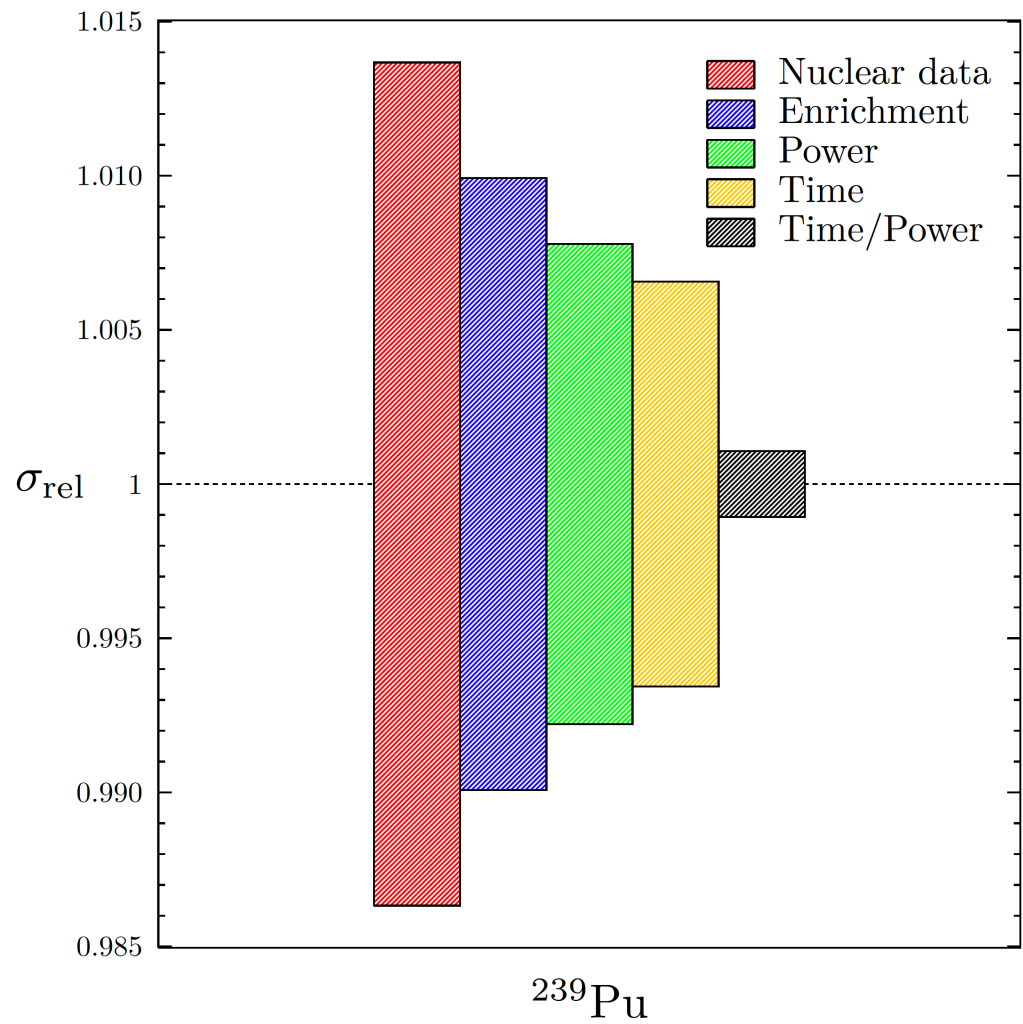
- More accurate ^{239}Pu capture and fission cross section data are needed, as they have been listed in the NEA/OCDE High Priority Request List (HPRL).
- We propose to measure the α -ratio, (the fission) and the capture cross sections at the EAR1, using the **TAC and ionization chambers** developed at the University of Lodz. One CIEMAT PhD student (Adrián Sánchez) has started to work in this topic.
- The ^{239}Pu **samples will be manufactured at JRC-Geel**. These samples will be placed inside the fission detector, in the center of the TAC.
- We request **$5.0 \cdot 10^{18}$ protons on target** to perform this measurement.
 - **10 thin samples (1 mg each)**. We expect to determine the capture cross section and the α -ratio of ^{239}Pu with an accuracy of **$\sim 3\%$ below 100 eV and 4-6% between 100 eV and 1 keV**.
 - **One sample of 50-100 mg**. With this sample the measurement could be extended up to ~ 10 keV, with an accuracy in the capture cross section of **3-4%**. If we succeed, then we request an additional **$1.0 \cdot 10^{18}$ protons**, i.e. a total of **$5.0 \cdot 10^{18}$ protons on target**.

Acknowledgments

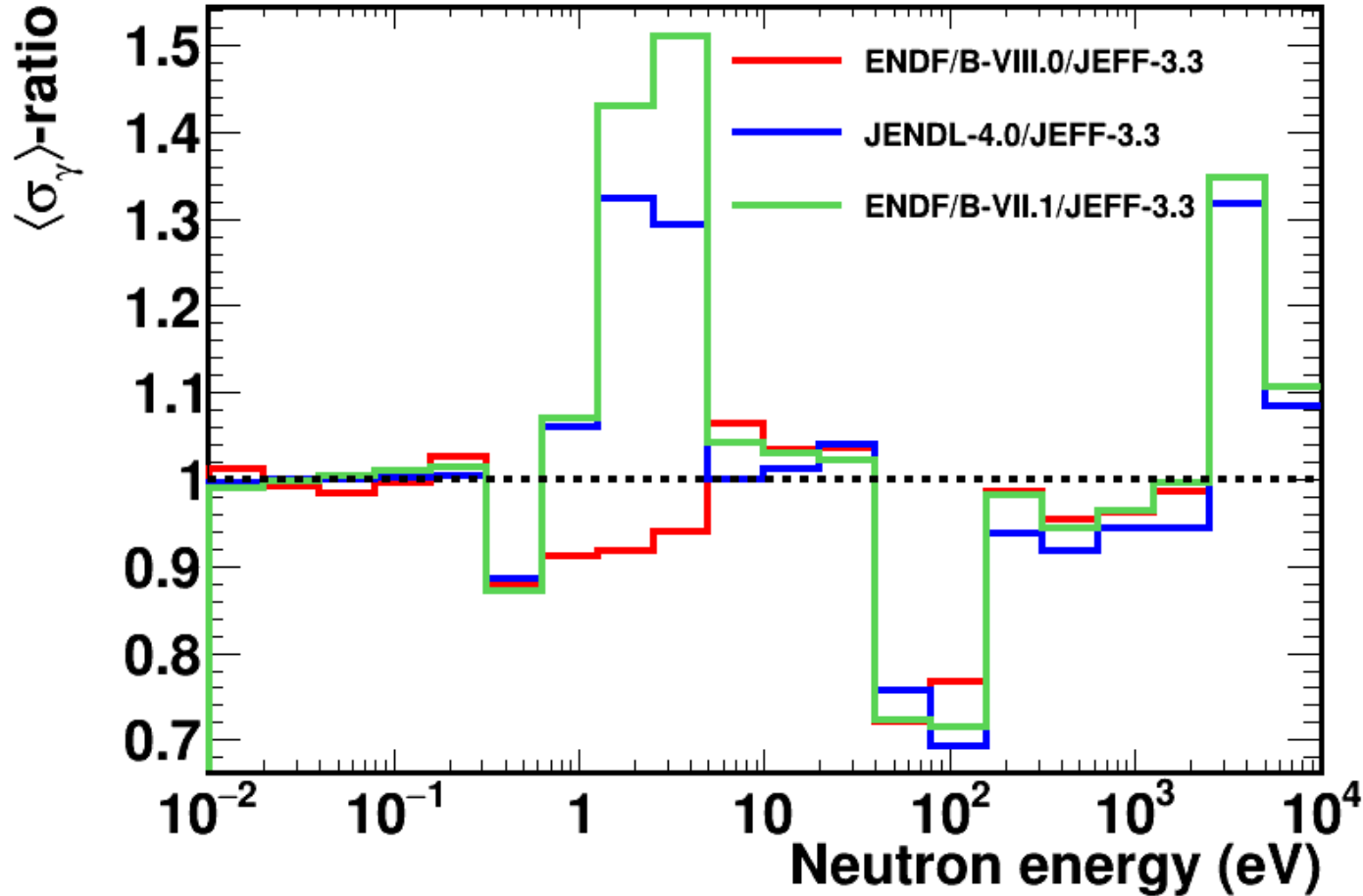
- This work is partially supported by the EC **H2020 SANDA** project (subtask 2.2.1) to perform this measurement and the Spanish Ministry of Science grant PGC2018-096717-B-C21.
- Additional funds have been provided for a **four years contract for a student which will perform his PhD in this measurement.**

Spare slides

Uncertainty in the ^{239}Pu mass in SNF after discharge

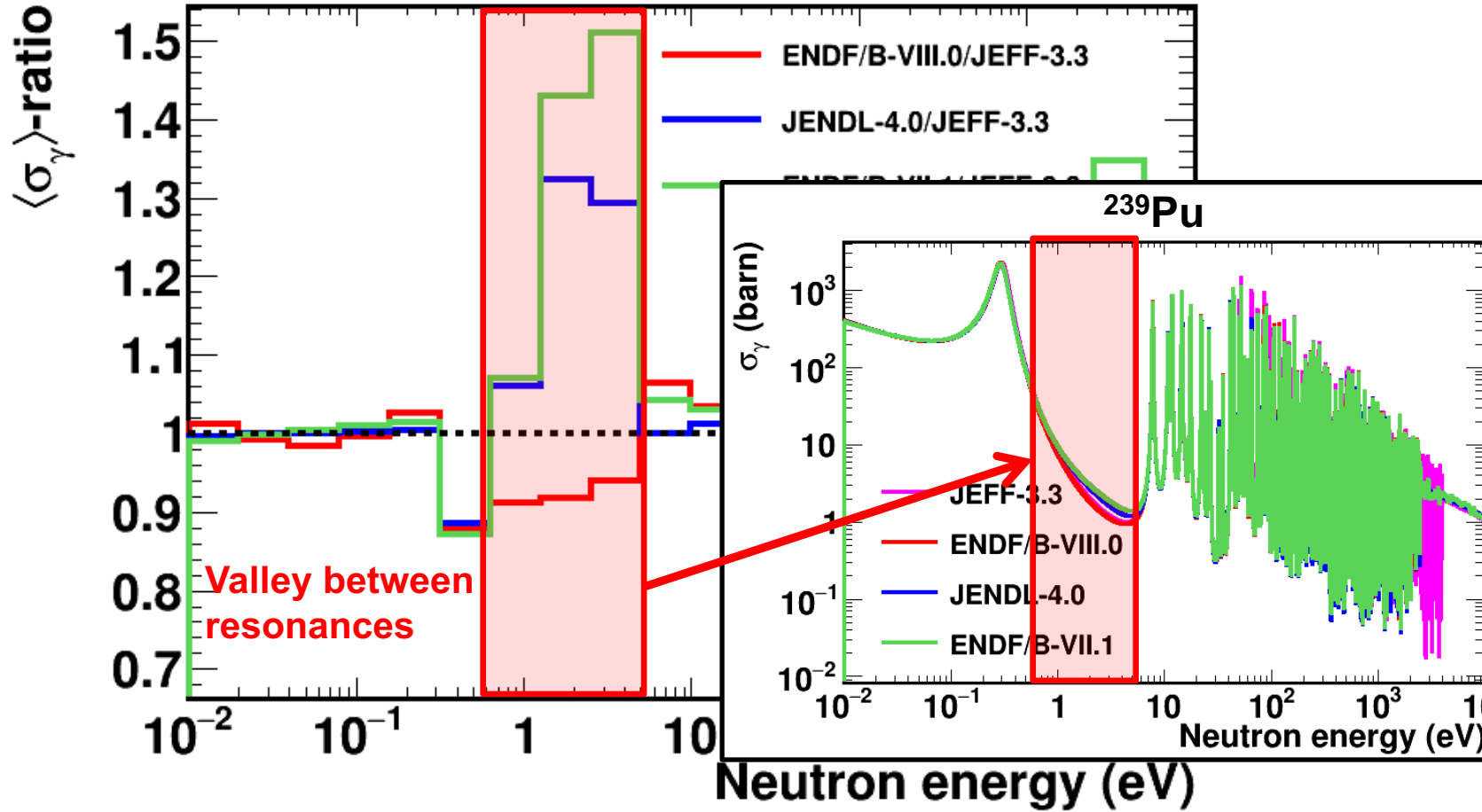


Status of the $^{239}\text{Pu}(n,\gamma)$ cross section



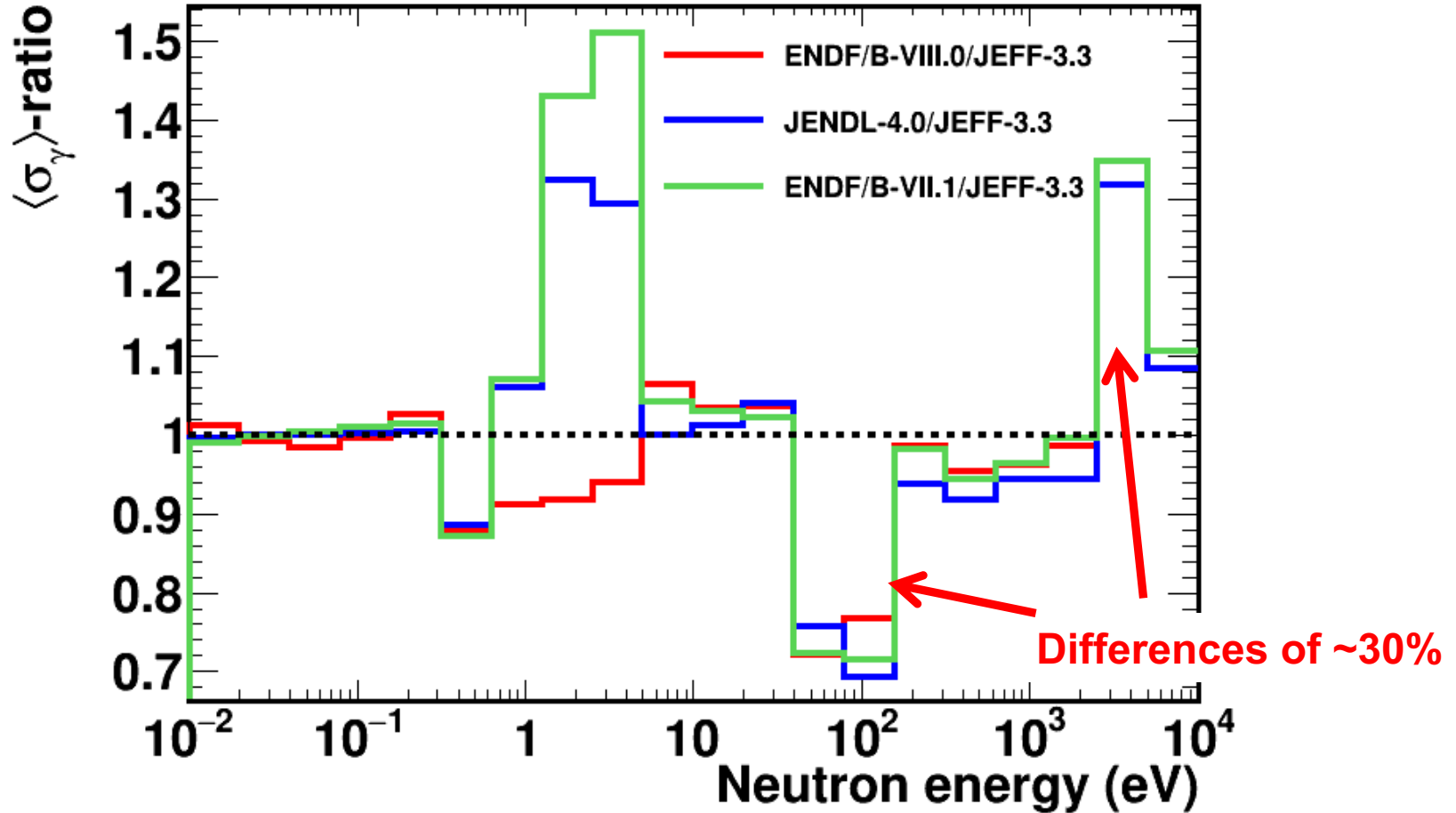
Ratio between the $^{239}\text{Pu}(n,\gamma)$ cross sections of ENDF/B-VIII.0, JENDL-4.0 and ENDF/B-VII.1 and JEFF-3.3.

Status of the $^{239}\text{Pu}(n,\gamma)$ cross section



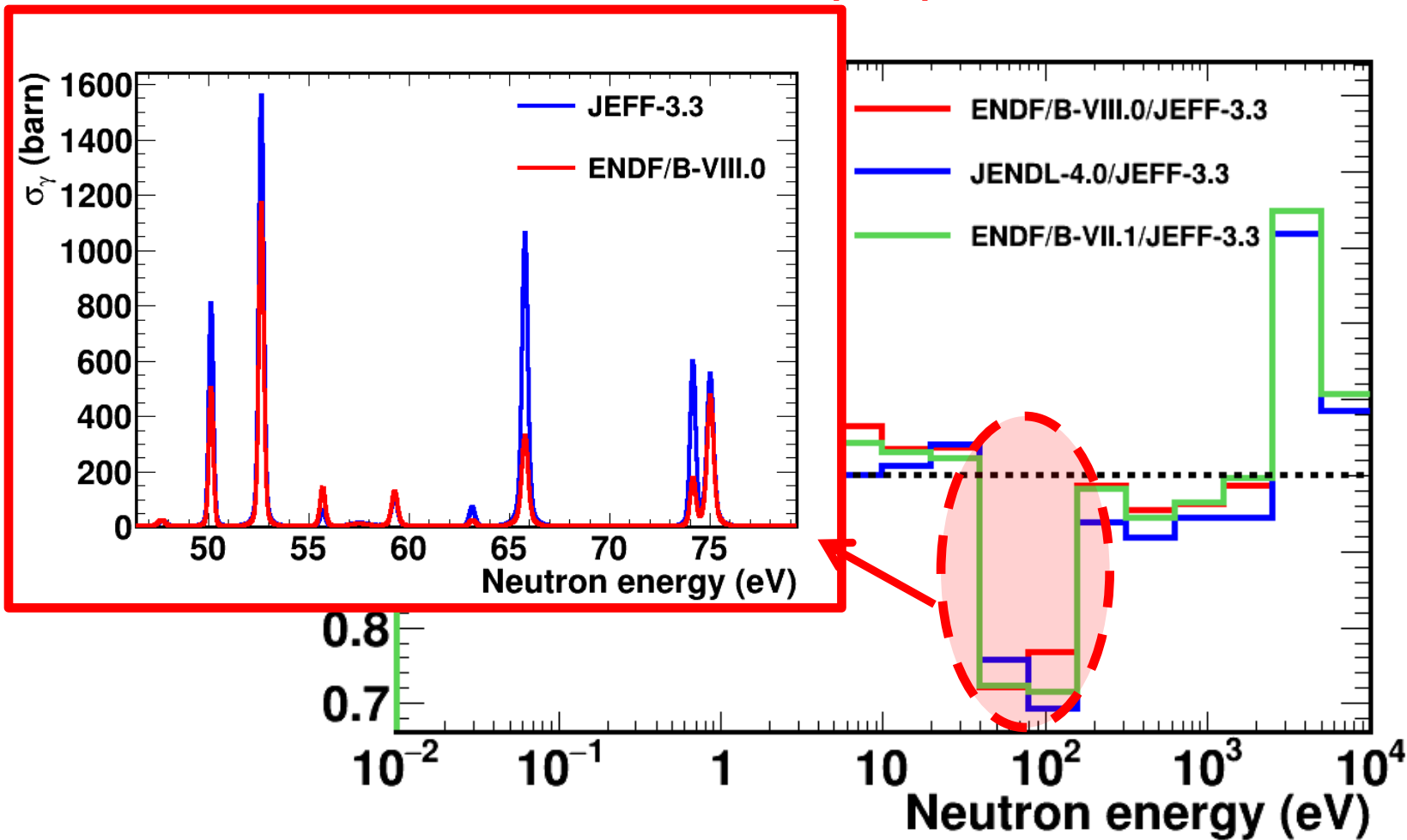
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Status of the $^{239}\text{Pu}(n,\gamma)$ cross section



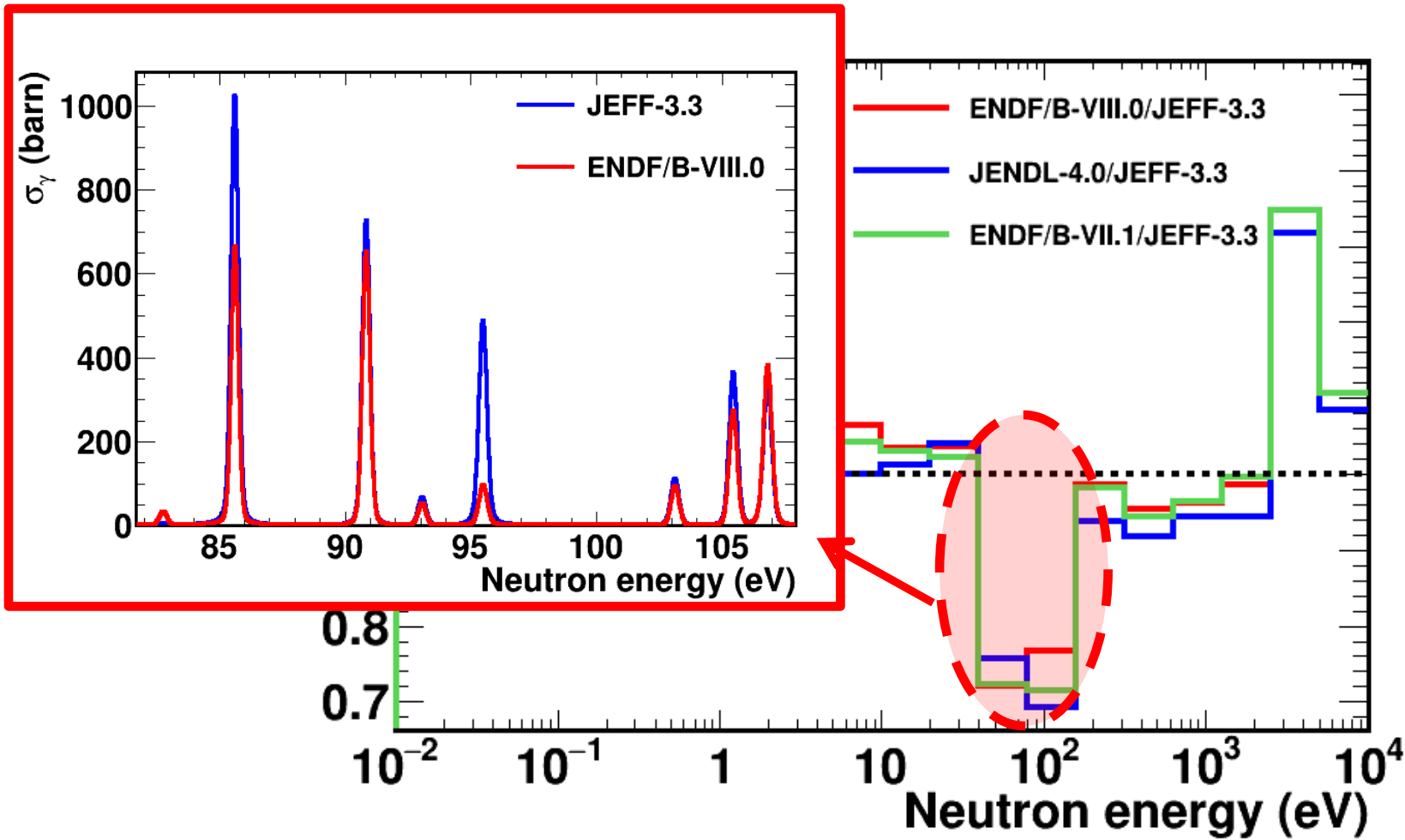
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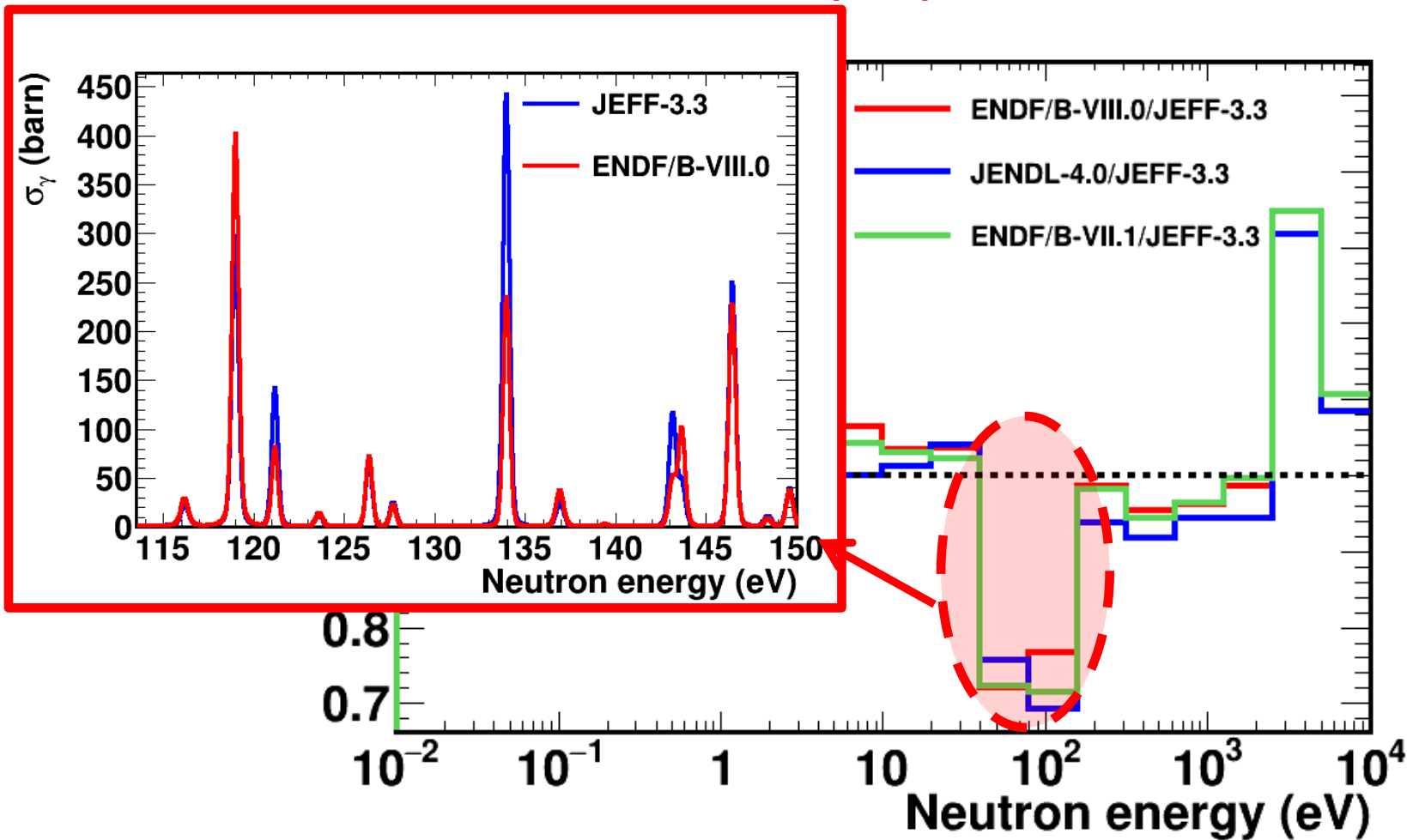
Ratio between the $^{239}\text{Pu}(n,\gamma)$ cross sections of ENDF/B-VIII.0, JENDL-4.0 and ENDF/B-VII.1 and JEFF-3.3.

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Ratio between the $^{239}\text{Pu}(n,\gamma)$ cross sections of ENDF/B-VIII.0, JENDL-4.0 and ENDF/B-VII.1 and JEFF-3.3.

The measurement technique

We will use the **fission tagging technique**, already developed for the $^{235}\text{U}(n,\gamma)$ measurement (see the *Balibrea et al.* draft submitted recently to PRC for more details).

We have performed a very detailed analysis to estimate the uncertainties we will have in the α -ratio and in the capture cross section after performing the measurement. The estimations have been validated with the results of the $^{235}\text{U}(n,\gamma)$ measurement.

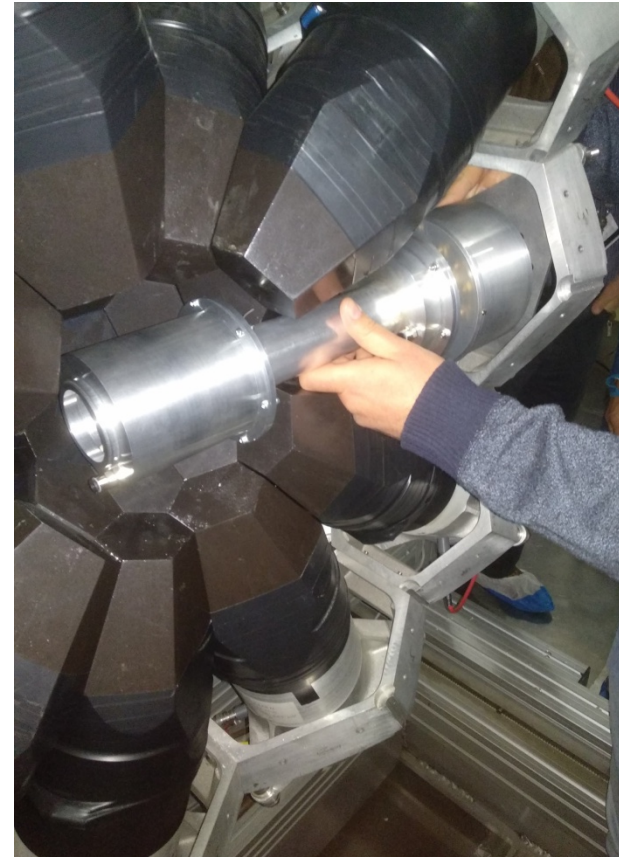
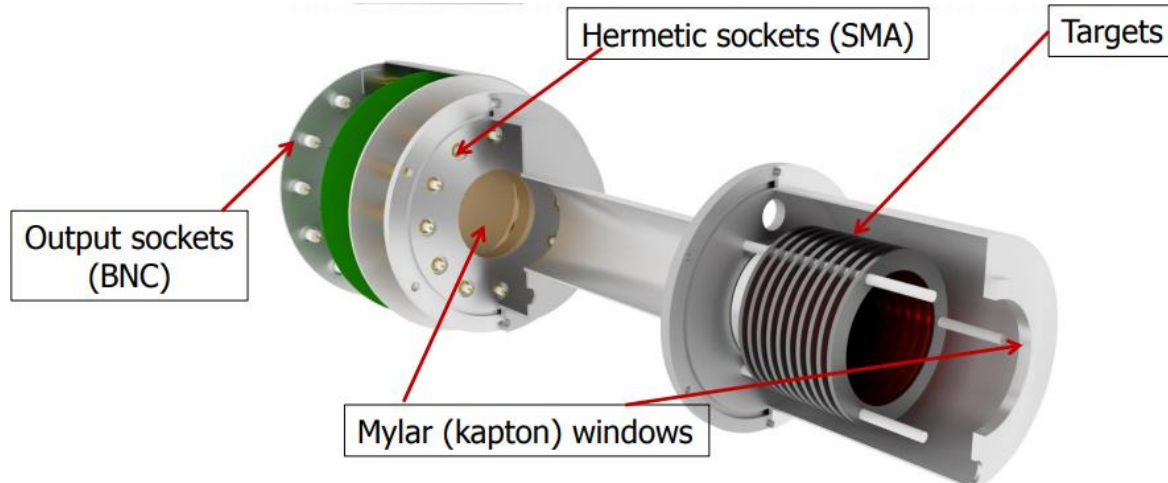
According to these estimations:

- With a **10 mg** sample: σ_γ and α -ratio can be determined with **~3% below 100 eV and 4-6% between 100 eV and 1keV**.
- With an **80 mg** sample : σ_γ and α -ratio can be determined with a bit less accuracy below 100 eV, but **3-4% between 100 eV and 10 keV**.

The fission chamber

The fission chamber has been already designed and built at the University of Lodz. It has been already presented at:

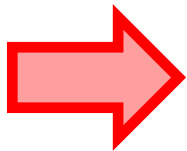
- n_TOF Detectors Meeting, CERN, Feb 2020 → [link](#)
- n_TOF Collaboration Meeting, Prague, October 2019 → [link](#)



The fission chamber

The fission chamber has been designed taking into account the following important characteristics:

1. **Low mass** intercepting the neutron beam, to minimize the background in the TAC due to capture and elastic scattered neutrons.
2. Good **discrimination** between **alphas** and **fission fragments**.
3. Small **pile-up** effects.

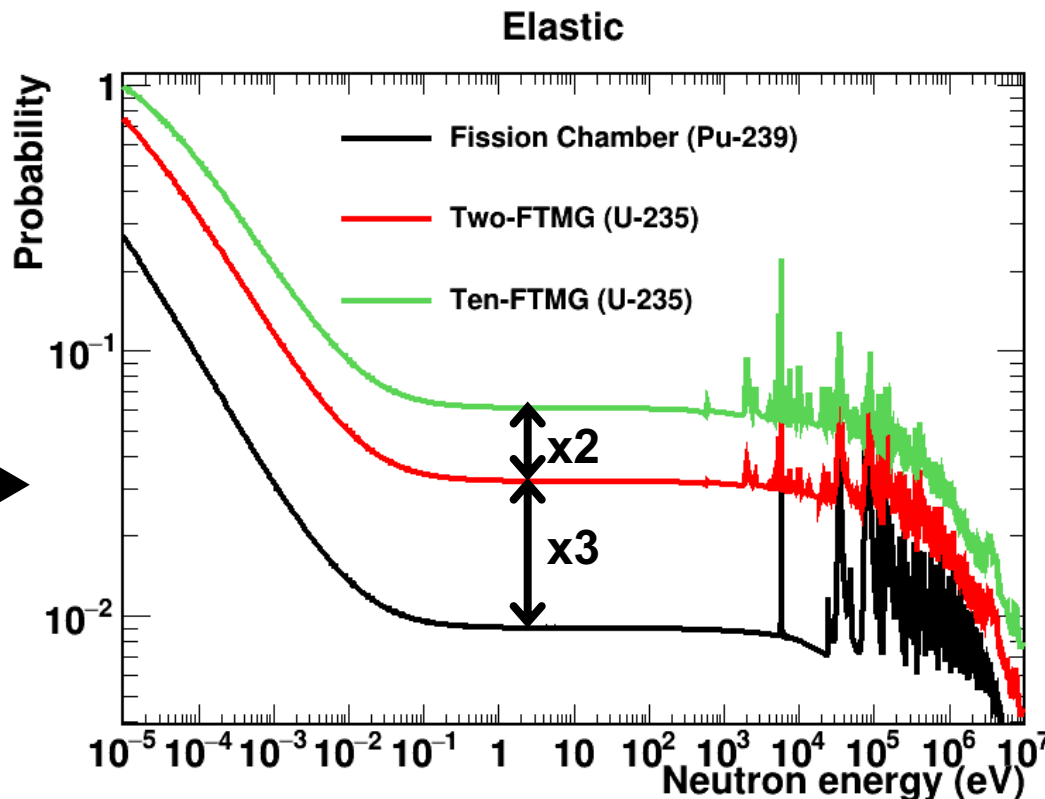


Before the measurement at n_TOF, **the fission chamber will be tested** in a $^{239}\text{Pu}(n,f)$ measurement at **JRC-Geel**.

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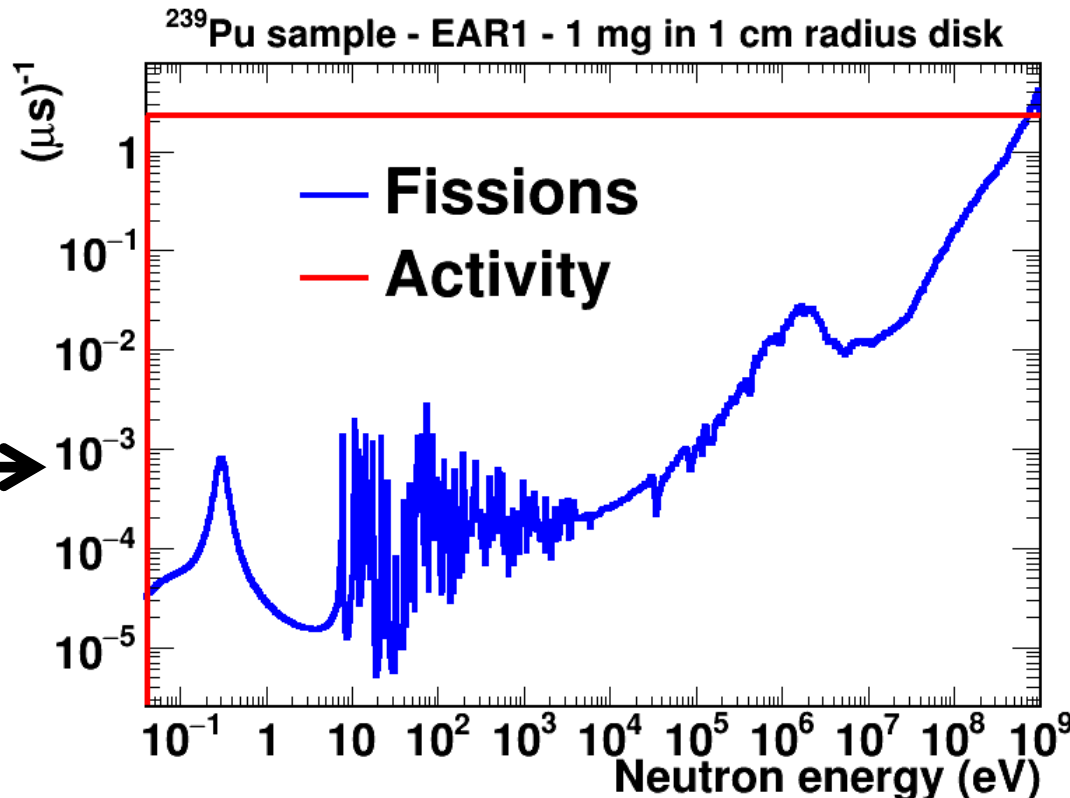


Probability of elastic scattering as a function of the neutron energy, in the new fission chamber (black) and in the fission detectors used in the ^{235}U measurement (red and green).

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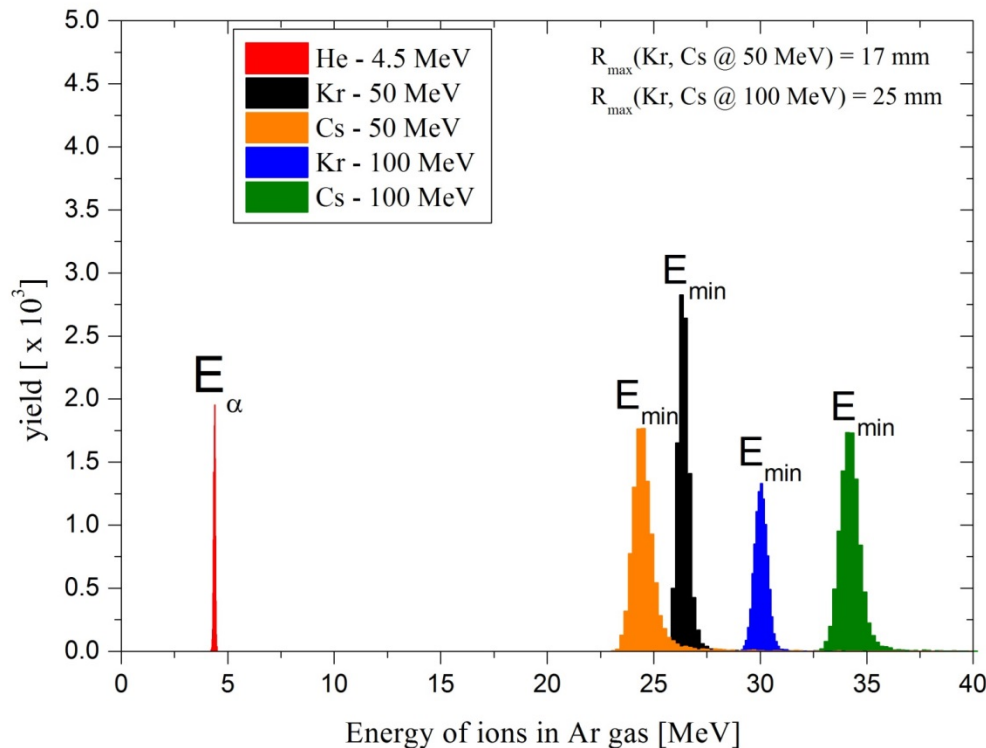


Expected α -activity in one sample (1 mg) together with the expected fission rates, as a function of the neutron energy.

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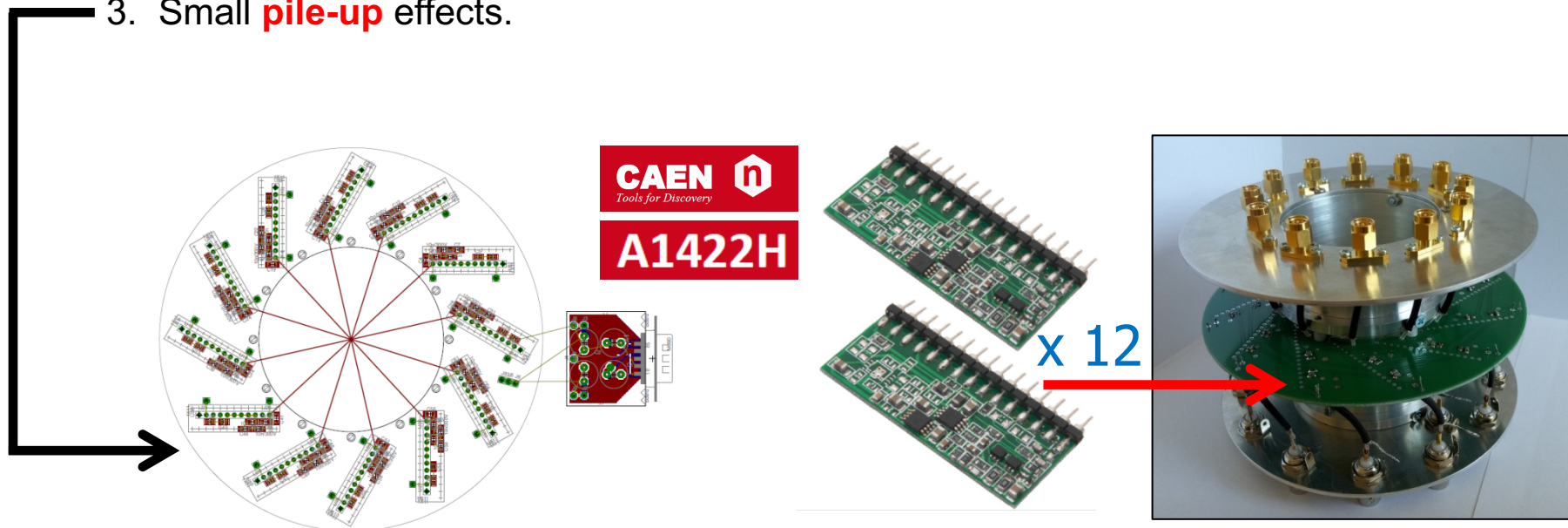


The simulations of energy distributions in 5 mm of gas of α particles and the chosen fission fragments (Kr, Cs) prepared with using the SRIM code.

The fission chamber

The fission chamber has been designed taking into account the following important characteristics:

1. **Low mass** intercepting the neutron beam, to minimize the background in the TAC due to capture and elastic scattered neutrons.
2. Good **discrimination** between **alphas** and **fission fragments**.
3. Small **pile-up** effects.

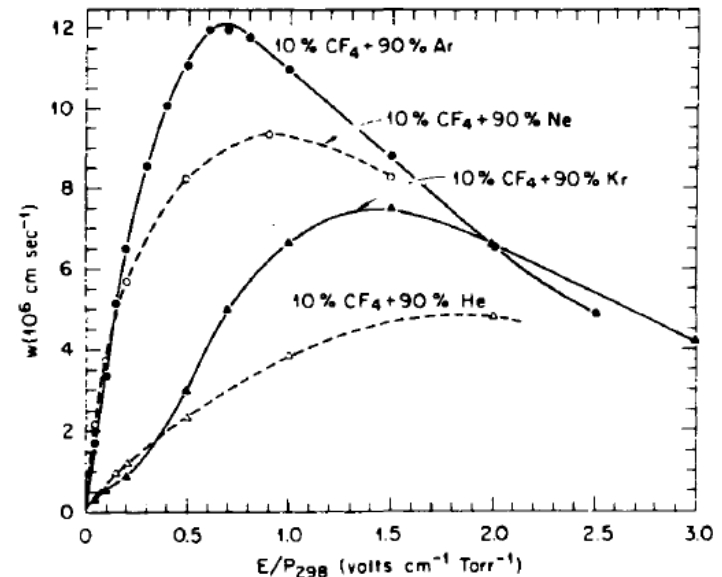
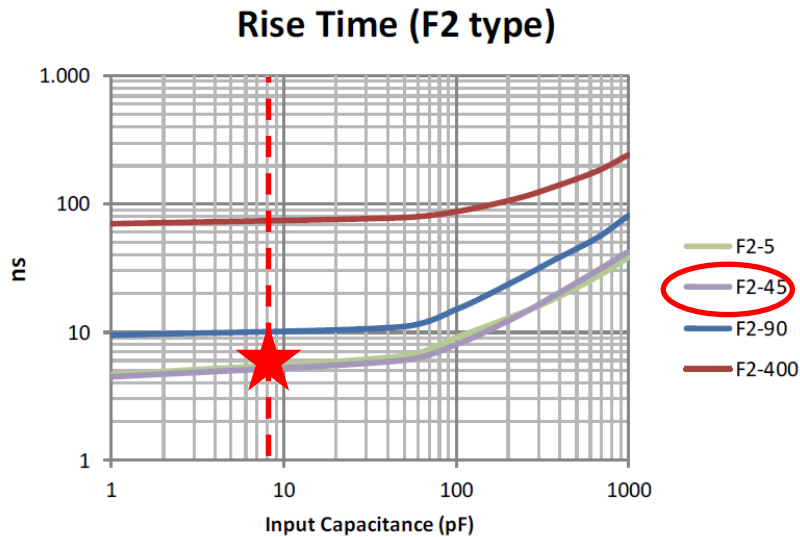


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L.G.Christophorou, et. al., Nuclear Instruments and Methods, 163 (1979) 141

$$Y_\gamma = \frac{c_{tot,\gamma}(E_n) - \frac{c_{tag}(E_n)}{\epsilon_f^*(E_n)} - c_{oth,\gamma}(E_n)}{\epsilon_f(E_n)\phi_N(E_n)}$$

$$c_{tot,\gamma}(E_n) = c_{aco,\gamma}(E_n) + c_{tag}(E_n)$$

- c_{tot} = counts in the TAC.
- c_{tag} = counts in the TAC in coincidence. with the fission chamber.
- c_{aco} = counts in the TAC in anti-coincidence. with the fission chamber.
- ϵ_f^* = fission tagging efficiency.
- ϵ_γ = capture detection efficiency.
- ϵ_f = fission detection efficiency.

$$Y_\gamma = \frac{c_{aco,\gamma}(E_n) - \frac{1 - \epsilon_f^*(E_n)}{\epsilon_f^*(E_n)} c_{tag}(E_n) - c_{oth,\gamma}(E_n)}{\epsilon_\gamma(E_n)\phi_N(E_n)}$$

and

$$\alpha = \frac{\epsilon_f}{\epsilon_\gamma} \frac{c_{aco,\gamma}(E_n) - \frac{1 - \epsilon_f^*(E_n)}{\epsilon_f^*(E_n)} c_{tag}(E_n) - c_{oth,\gamma}(E_n)}{c_{aco,f}(E_n) + c_{tag}(E_n) - c_{bkg f}(E_n)}$$

σ(n,γ) data on actinides measured after 2000 (EXFOR)

Isotope	Facility	Detector	E _{low} (eV)	E _{high} (eV)	EXFOR	Publication
U-233	n_TOF-1	TAC	0,7	1000	Yes	E. Berthoumieux et al., Conf. on Nuclear Data for Science and Technology, Nice 2007, p.571 (2007)
U-233	n_TOF-1	TAC	?	?	No	M. Bacak et al., ND2016, EPJ Conf. 146, 03027 (2017)
U-235	LANSCE	TAC + PPAC	4	1,00E+06	Yes	M. Jandel et al., Phys. Rev. Lett. 109, 202506 (2012)
U-235	n_TOF-1	TAC + MGAS	1	22	Yes	C. Guerrero et al., Eur. Phys. Jour. A 48, 29 (2012)
U-235	n_TOF-1	TAC + MGAS	0,2	200	No	J. Balibrea et al., Nucl. Data Sheets 119, 10 (2014)
U-235	RPI	NaI - TAC	0,02	3000	No	Y. Danon, et al., Nucl. Sci. and Eng. 187, 191 (2017)
U-238	LANSCE	TAC	1	6,30E+05	Yes	J.L. Ullmann et al., Phys. Rev. C 89, 034603 (2014)
U-238	GELINA	C ₆ D ₆ TED	3,5	1200	Yes	H.I. Kim et al., Eur. Phys. Jour. A 52, 170 (2016)
U-238	n_TOF-1	C ₆ D ₆ TED	1	700	Yes	F. Mingrone et al., Phys. Rev. C 95, 034604 (2017)
U-238	n_TOF-1	TAC	1	8,00E+04	Yes	T. Wright et al., Phys. Rev. C 96, 064601 (2017)
Np-237	KURRI	C ₆ D ₆ - TED	0.005	1,00E+04	Yes	K. Kobayashi et al., Jour. Nucl. Sci. Tech. 39, 111 (2002)
Np-237	KURRI	BGO - TED	0,02	100	Yes	O. Shcherbakov et al., Jour. Nucl. Sci. Tech. 42, 135 (2005)
Np-237	KURRI	Ge	0,02	14	Yes	M. Mizumoto et al., Conf. on Nuclear Data for Science and Technology, Nice 2007
Np-237	LANSCE	TAC	0,02	5,00E+05	Yes	E.I. Esch et al., Phys. Rev. C 77, 034309 (2008)
Np-237	n_TOF-1	TAC	0,7	2000	Yes	C. Guerrero et al., Phys. Rev. C 85, 044616 (2012)
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Pu-239	LANSCE	TAC	10	1000	Yes	S. Mosby et al., Phys. Rev. C 89, 034610 (2014)
Pu-240	n_TOF-1	TAC	0,7	2000	No	C. Guerrero et al., Conf. on Nuclear Data for Science and Technology, Nice 2007
Pu-242	LANSCE	TAC + PPAC	0,027	3,60E+04	Yes	M.Q. Buckner et al., Phys. Rev. C 93, 044613 (2016)
Pu-242	n_TOF-1	C ₆ D ₆ - TED	2	4000	Yes	J. Lerendegui-Marco et al., Phys. Rev.C 97, 024605 (2018)
Am-241	LANSCE	TAC	0,02	3,20E+05	Yes	M. Jandel et al., Phys. Rev.C 78, 034609 (2008)
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Am-241	J-PARC	C ₆ D ₆ TED	0,025	100	No	K. Terada et al., Jour. Nucl. Sci. Tech. 55, 1198 (2018)
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Am-243	n_TOF-1	TAC	0,7	2500	Yes	E. Mendoza et al., Phys. Rev. C 90, 034608 (2014)
Cm-244	J-PARC	Ge	2	300	Yes	A. Kimura et al., Jour. Nucl. Sci. Tech. 49, 708 (2012)
Cm-244	n_TOF-2	C ₆ D ₆ TED	1	300	No	V. Alcayne et al., WONDER-2018, Aix-en-Provence France, October 2018
Cm-244	n_TOF-1	TAC	1	50	No	V. Alcayne et al., WONDER-2018, Aix-en-Provence France, October 2018
Cm-246	J-PARC	Ge	2	300	Yes	A. Kimura et al., Jour. Nucl. Sci. Tech. 49, 708 (2012)
Cm-246	n_TOF-2	C ₆ D ₆ TED	1	300	No	V. Alcayne et al., WONDER-2018, Aix-en-Provence France, October 2018

Data sets from n_TOF

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CIEMAT's contribution (11 of 35)

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