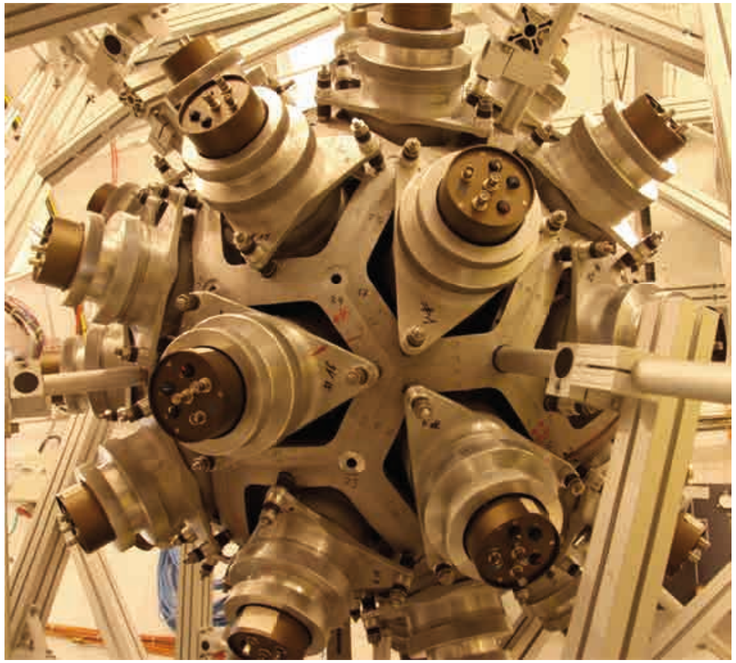
Measurement of the ^{241}Am(n,γ) cross section at the CERN n_TOF facility



E. Mendoza, D. Cano Ott, C. Guerrero and the n_TOF collaboration
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Why to measure the $^{241}Am(n,\gamma)$ cross section

•^{241}Am and ^{237}Np are the minor actinides which contribute most to the long term hazard of the nuclear waste (^{241}Am decays into ^{237}Np via α emission).

•After Np, Am is the second most abundant minor actinide in the spent nuclear fuel. ^{241}Am is the most abundant Am isotope present in the spent nuclear fuel.

•The (n,γ) reactions in Am isotopes are the path to heavier minor actinides like Cm isotopes.

•In a fast spectrum, $^{241}Am(n,\gamma)$ leads to ^{242}Cm production via the decay of the ^{242}Am ($T_{1/2}$=16hr). ^{242}Cm decays via α emission into ^{238}Pu, which is fissile in a fast spectrum.

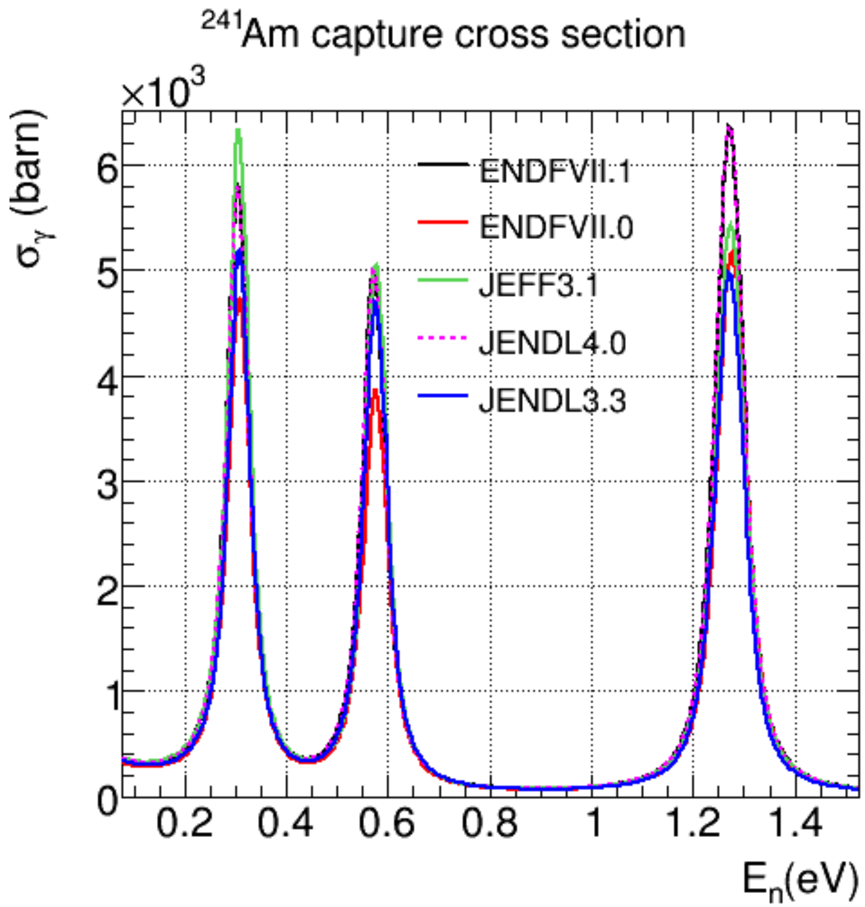
•The reprocessing of Am is technologically more advanced than for other minor actinides. Indeed, the fabrication of Am-oxide blankets for the transmutation of Am in fast reactors is already considered as feasible.

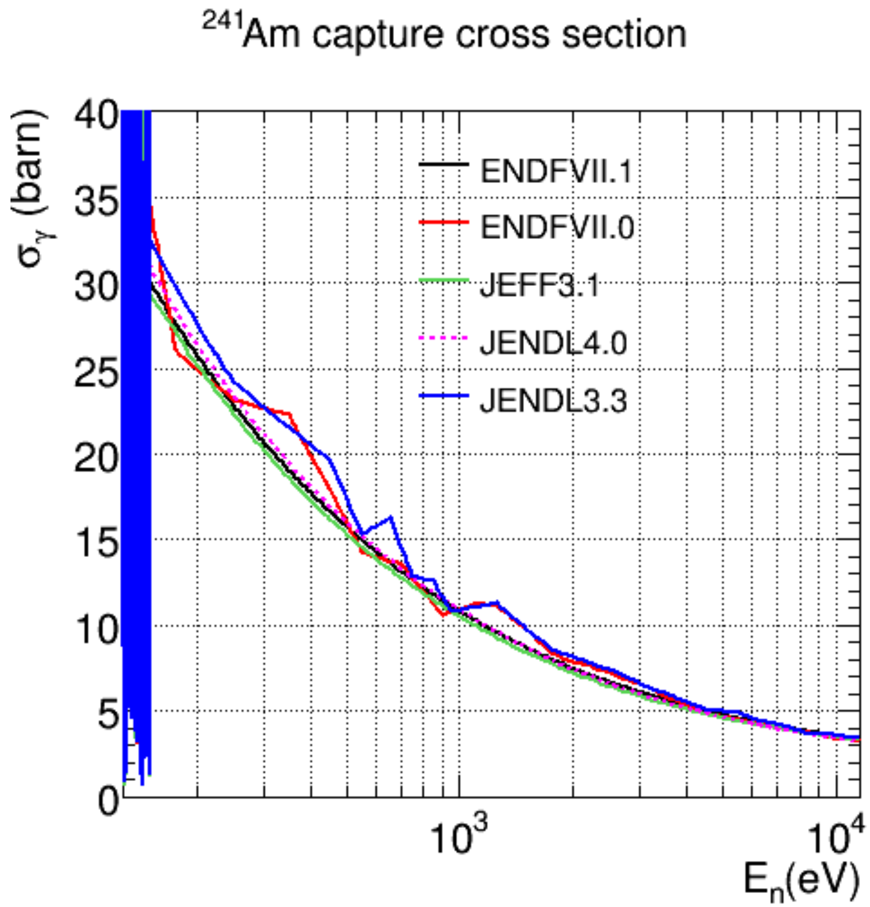
The improvement of the ^{241}Am data is a major effort of the **ANDES project**: 2 capture measurements at n_TOF with the TAC (E. Menodza et al.) and C_6D_6 detectors (K. Fraval et al.) and 1 transmission + 1 capture measurement with C_6D_6 detectors at IRMM – Geel (Lampoudis et al.)

New high quality data for the future evaluations!

SEVENTH FRAMEWORK PROGRAMME

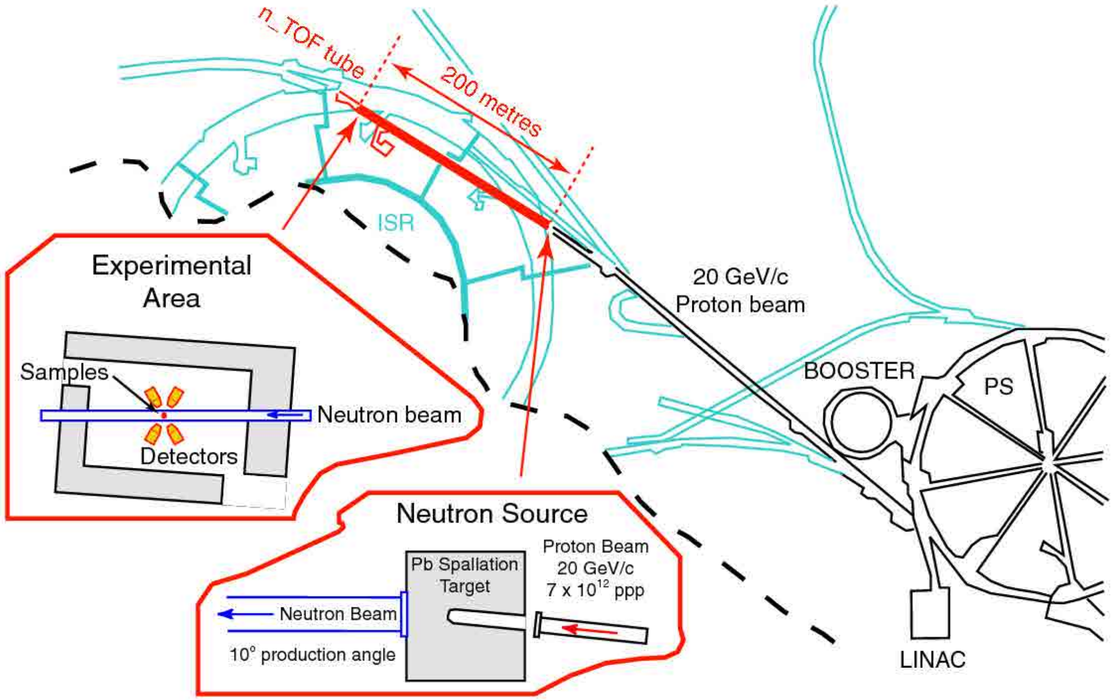
Present status of the evaluated libraries

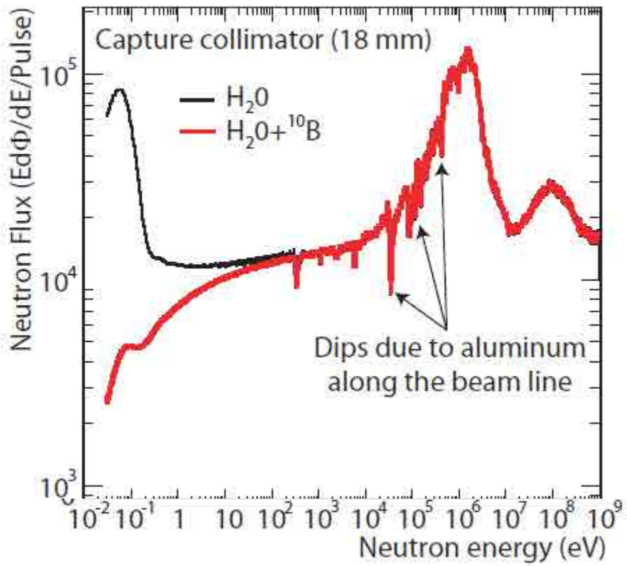




The CERN n_TOF facility

The n_TOF facility is a high instantaneous intensity spallation neutron source driven by the CERN PS synchrotron (20 GeV/c with 8x10^{17} ppp)





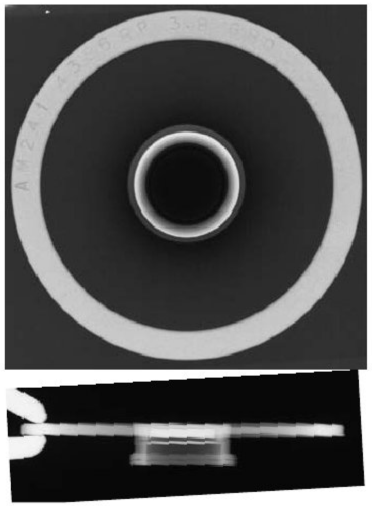
The neutron energy range extends from thermal energies up to GeV (meV up to 1 MeV for the case of capture). Two types of moderators are available:

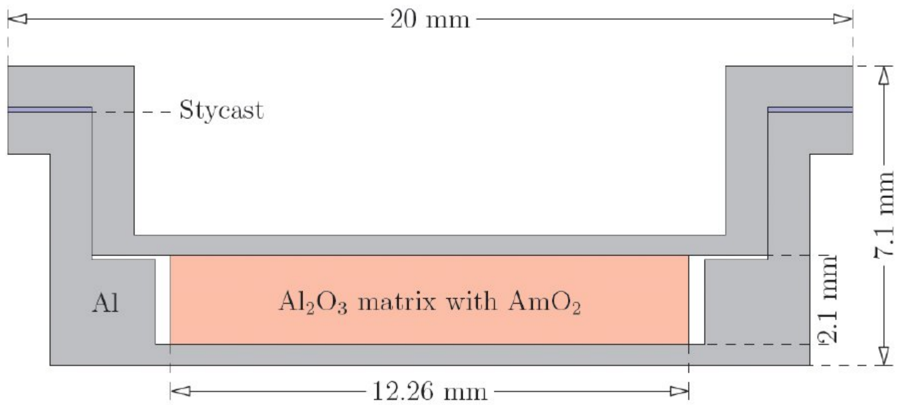
-Water: $^{1}H(n,\gamma)$ background

-Borated water: reduced background.

The ^{241}Am sample

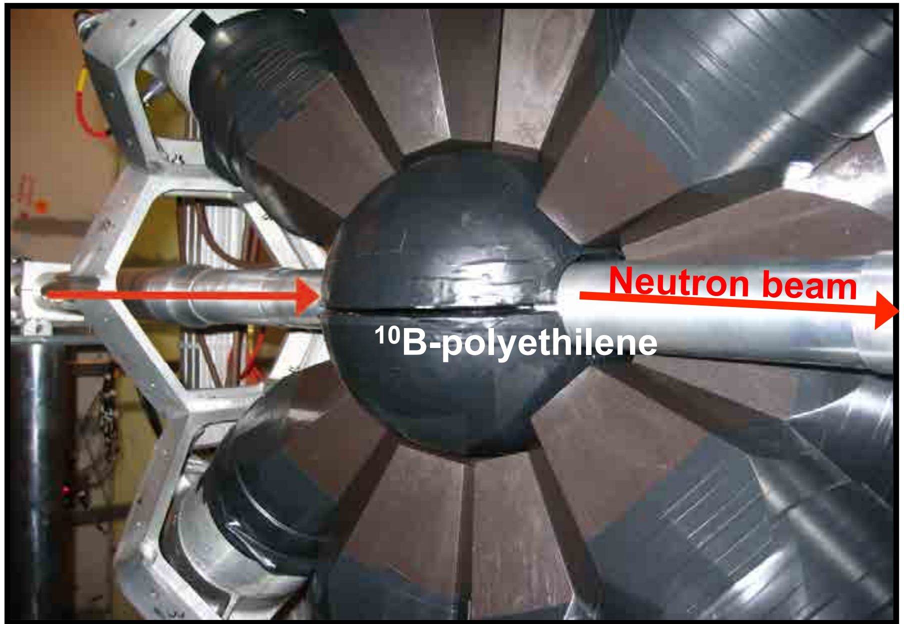
The ^{241}Am has been borrowed from IRMM Geel. The sample (disk-shaped with 12.2 mm diameter) consists of **32.2(±0.19) mg** of ^{241}Am (**~4 GBq**) oxide embedded in a 305 mg Al_2O_3 matrix and encapsulated in a 0.5 mm thick aluminum canning. The sample is part of a set of samples prepared for IRMM at ITU from material given by CEA for (n,γ) and (n,2n) cross section measurements.

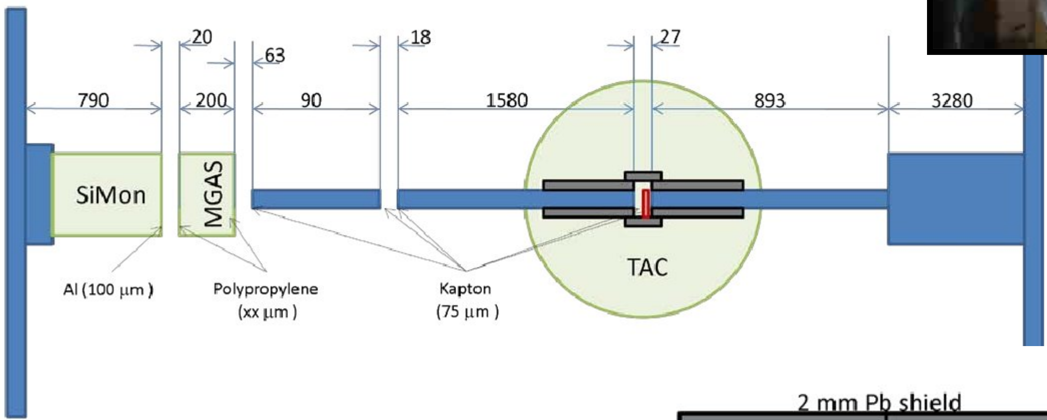


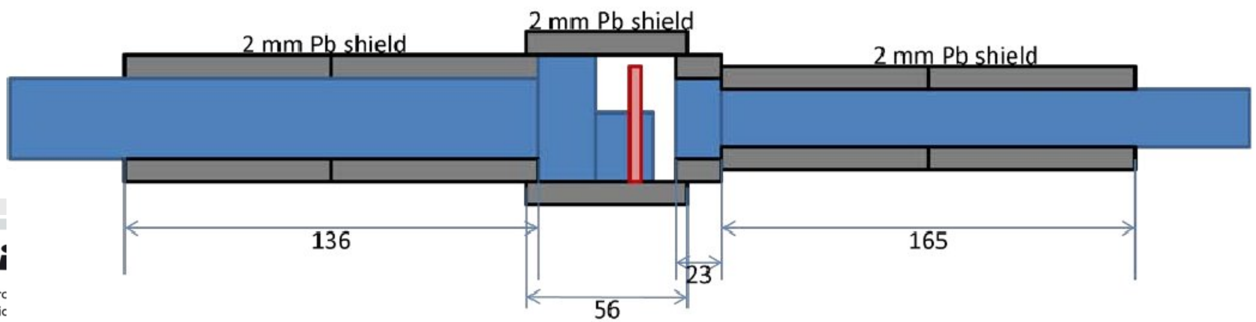


The experimental setup

The 40-fold segmented Total Absorption Calorimeter made of BaF_2 was used for the $^{241}Am(n,\gamma)$ measurement. The huge sample activity required to surround the sample with a 2 mm Pb shielding. Two monitors (Silicon Flux Monitor and ^{235}U loaded micromegas).







The measurements at n_TOF

About 2 months of beam time were dedicated to the ^{241}Am measurement. A large series of measurements were performed with two flash ADC sampling rates: 500 MHz (E_n > 0.7 eV) and 250 MHz (E_n > 0.1 eV), in order to measure the strongest resonances.

Sampling rate	**250 MHz**		**500 MHz**	
Measurement	**Pulses**	**Protons ($x10^{17}$)**	**Pulses**	**Protons ($x10^{17}$)**
^{241}Am	16732	1.44	88344	7.56
Dummy (without ^{241}Am)	10942	0.93	10185	0.87
Canning (no Al_2O_3)	16165	1.40	8696	0.75
Kapton	1470	0.13	5743	0.49
Nothing	6556	0.57	5292	0.45
Carbon	2225	0.18	0	0
Activity	4067	-	251	-
Ambient	4999	-	851	-
Au197	6708	0.46	1634	0.14
Total	**69864**	**5.12**	**120996**	**10.36**

The capture yield

The capture yield is the common starting point of the evaluators (retrieved from EXFOR or elsewhere) in order to perform the cross section evaluations / resonance analysis.

$$Y_\gamma(E_n) = \frac{\text{number of neutrons with energy } \mathrm{E_n} \text{ captured in the sample}}{\text{number of neutrons of energy } \mathrm{E_n} \text{ hitting the sample}} = \frac{N_\gamma(E_n)}{N_T(E_n)}$$

In a capture experiment, we do measure counting rates in the detectors.

$$N_\gamma(E_n) = \frac{ND(E_n) - BK(E_n)}{\varepsilon_\gamma}$$

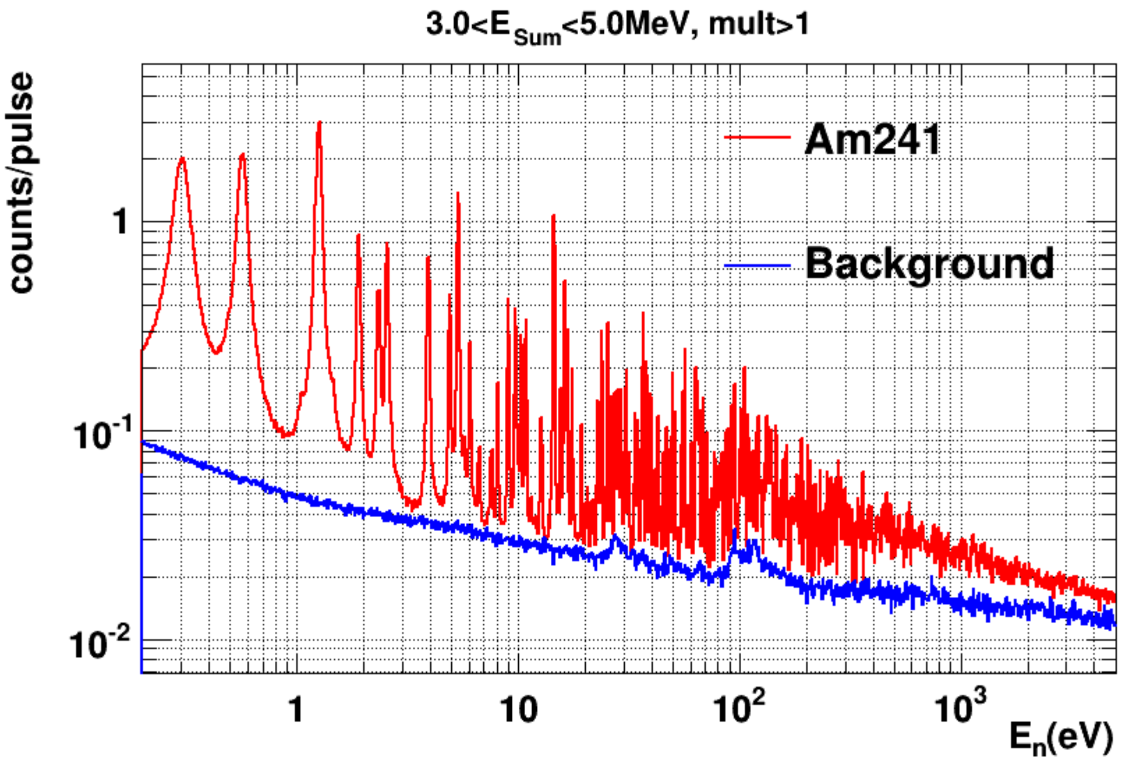
The number of neutrons hitting the sample $N_T(E_n)$ is a normalisation which is determined in relation to standard cross sections, other data (evaluated or experimental) or self normalised.

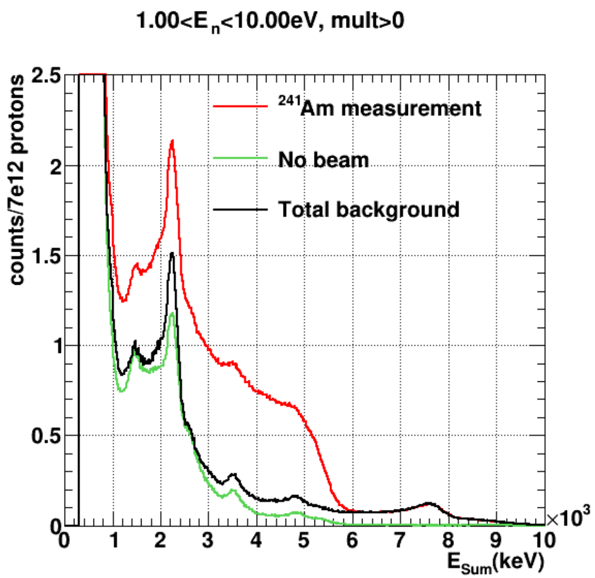
Procedure:

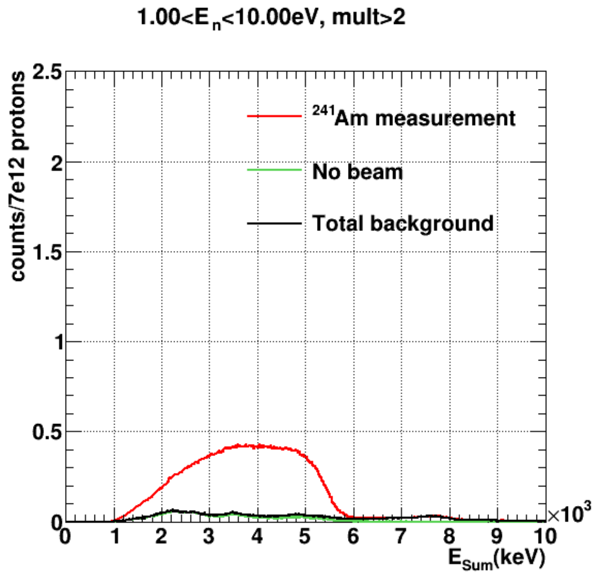
- Determination from the total counting rate as a function of the time of flight
- Subtraction/characterization of the different types of background.
- Normalization of the yield (for the moment, to evaluated data).

The procurement of the TAC capture yield

The TAC crystals register the energy of the γ-rays forming a capture cascade. The total energy deposited in the TAC (sum of the energy of coincident crystals) is obtained as a function of the crystal multiplicity and the TOF.







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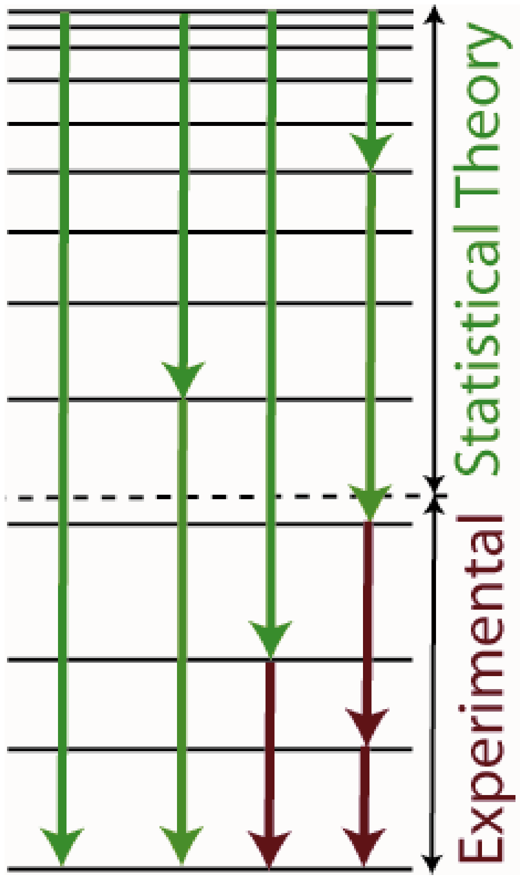
Ingredients of the Monte Carlo simulation



- Very detailed geometric modelling of the TAC. (C. Guerrero et al., Nucl. Instr. Meth. A 671 (2012))
- Pulse pilep / dead time model (E. Mendoza et al. NIMA in preparation)
- DECAYGEN code (J.L.Tan, D.Cano-Ott, Nucl. Instr. Meth. A 571 (2007))

Discrete level scheme: generation of realistic capture cascades taking the ENSDF data –levels, branching ratios, conversion coefficients –.

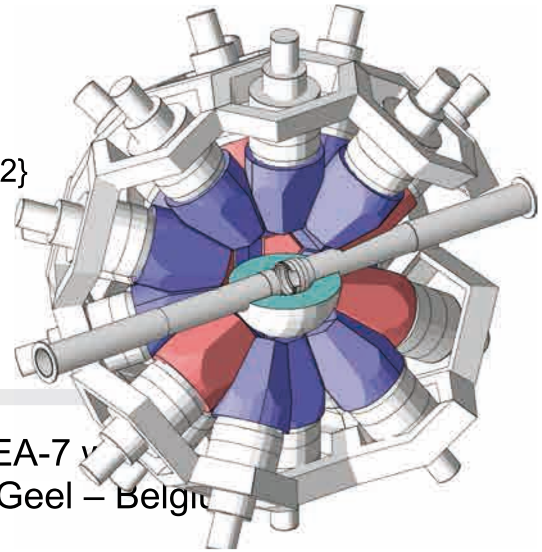
Statistical region: described by the nuclear level density and photon strength functions.

Transition probabilities given by:

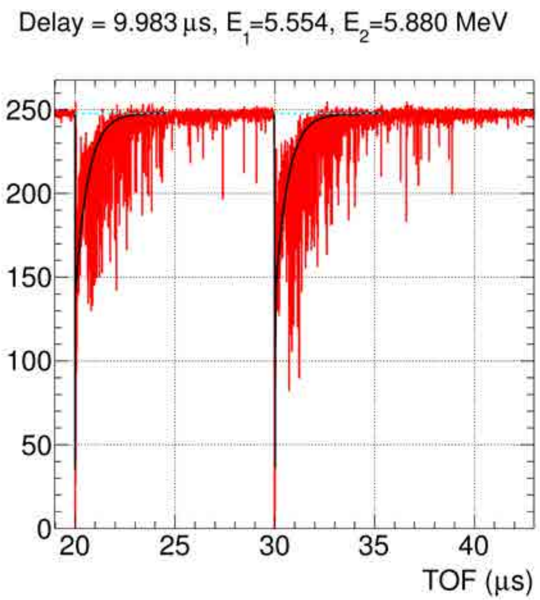
$T_{XL}(E_i,E_f) = f_{XL}(E_g)\ \rho(E_f,I,\Pi)_{XL=\{E1,M1,E2\}}$

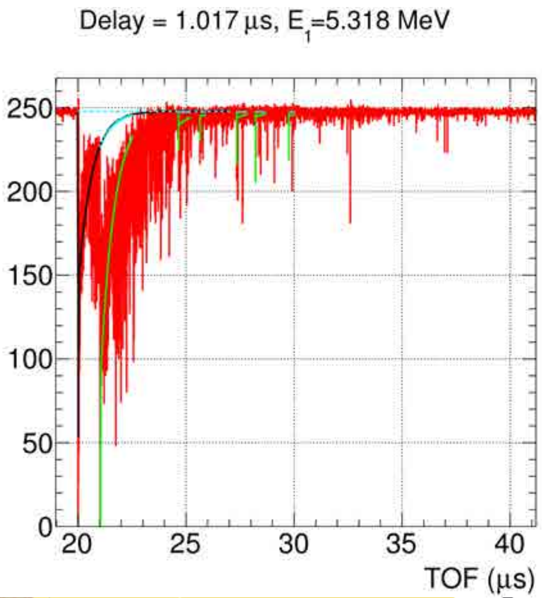
$$\varepsilon = \varepsilon(E_n, E_{sum}, m_{crystal}, CR)$$

Detection efficiency as a function of the neutron energy, deposited energy, crystal multiplicity and counting rate.



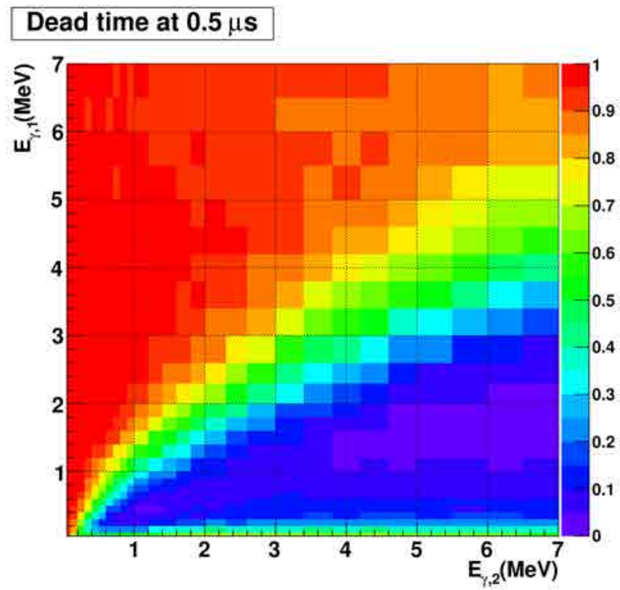
New dead time correction method



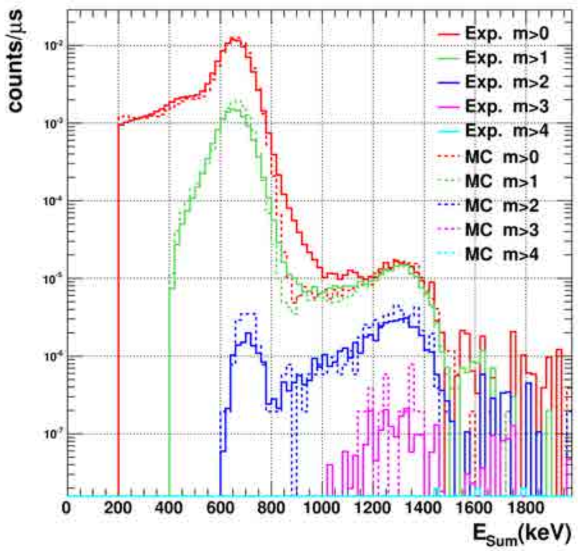


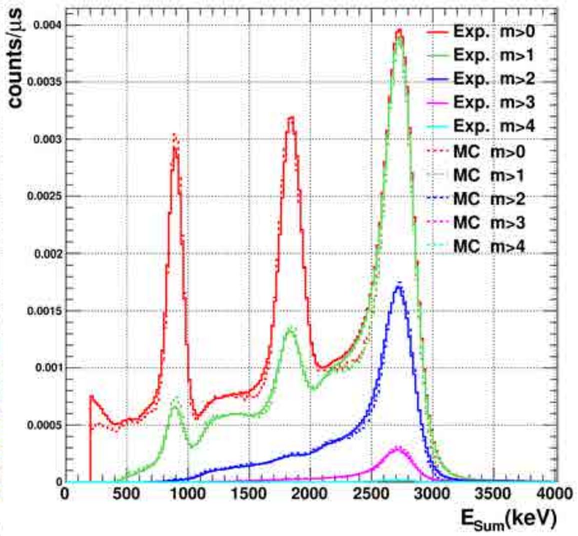
A pulse pile-up and dead time reconstruction method has been developed (*E. Mendoza et al* , NIM in preparation) for treating the ^{241}Am data (also applied to the ^{238}U data).

- Generate pairs of real detector signals with well know amplitudes and separated by well known times.
- Processing of the pairs of signals with the pulse shape analysis routine.
- Characterisation of the effect of the pile-up on the detection probability (E1, E2, DT) and on the reconstructed energy.
- Processing of the pairs of signals with the pulse shape analysis routine.-



Determination of the efficiency by Monte Carlo simulation

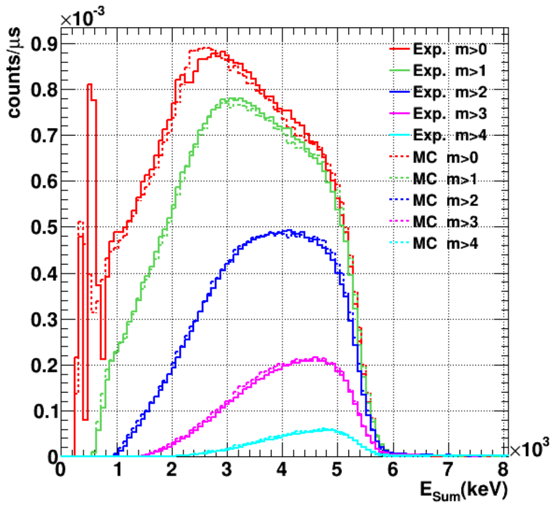


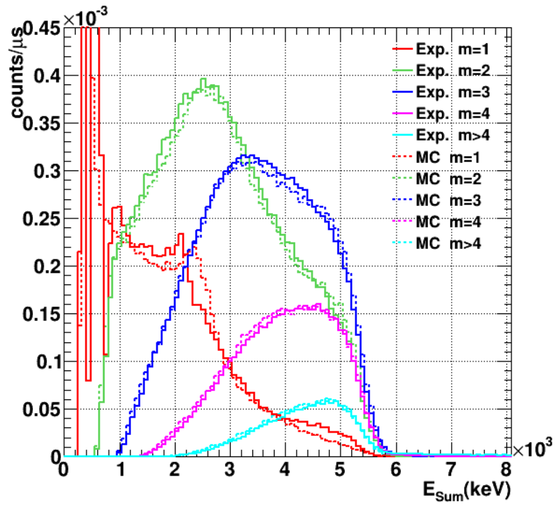


Comparison of the simulations with simple geometries and decay schemes (γ-ray sources like ^{137}Cs, ^{88}Y, ^{24}Na and $^{nat}Ti(n,\gamma)$):

- With/without Pb shielding
- w/wo neutron absorber

An excellent agreement was found.





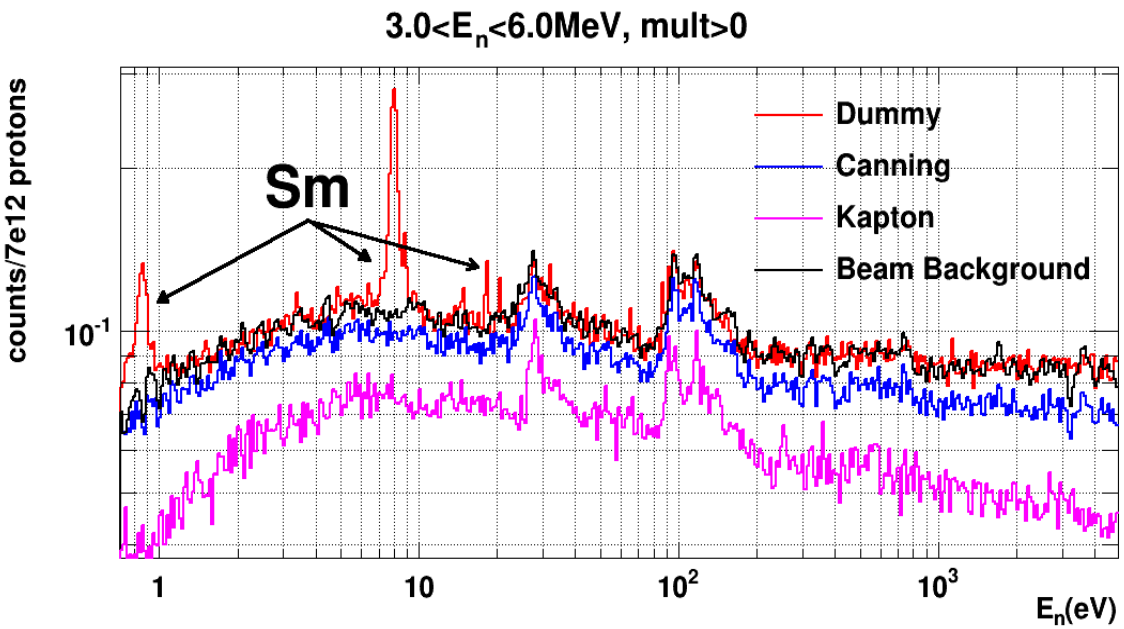
Comparison of the simulations to the ^{241}Am capture data.

-Realistic model for the de-excitation of the nucleus, based on experimental information and statistical parameters (known levels and branching ratios, level densities, Photon Strength Functions…)

-Adjustment of the model parameters.

The background

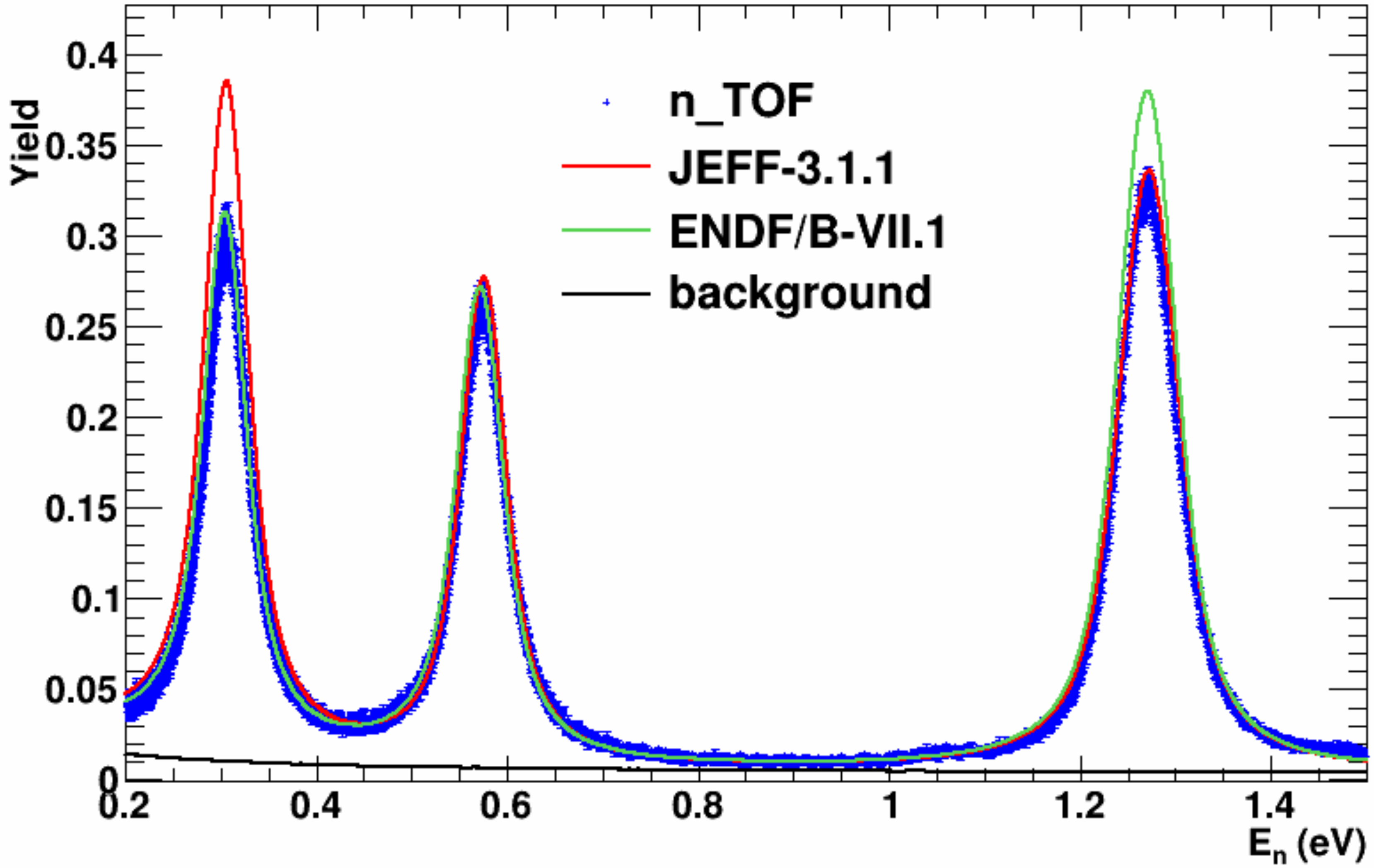
- The background component which is not related with the neutron beam (~4 GBq ^{241}Am activity)
- (n,f) –negligible– or (n,el) reactions in ^{241}Am –determined from the ^{nat}C measurement–.
- Interaction of neutrons with the dead material: other detectors, vacuum windows (Kapton), **dummy canning** (^{nat}Sm contamination in the Al_2O_3 used in the dummy canning fabrication!)



The dummy canning background had to be reconstructed from the canning measurement (just Al, no Al_2O_3) and an Al capture component.

The background is not subtracted from the data but used as a pointwise background in SAMMY.

The yield: first three resonances



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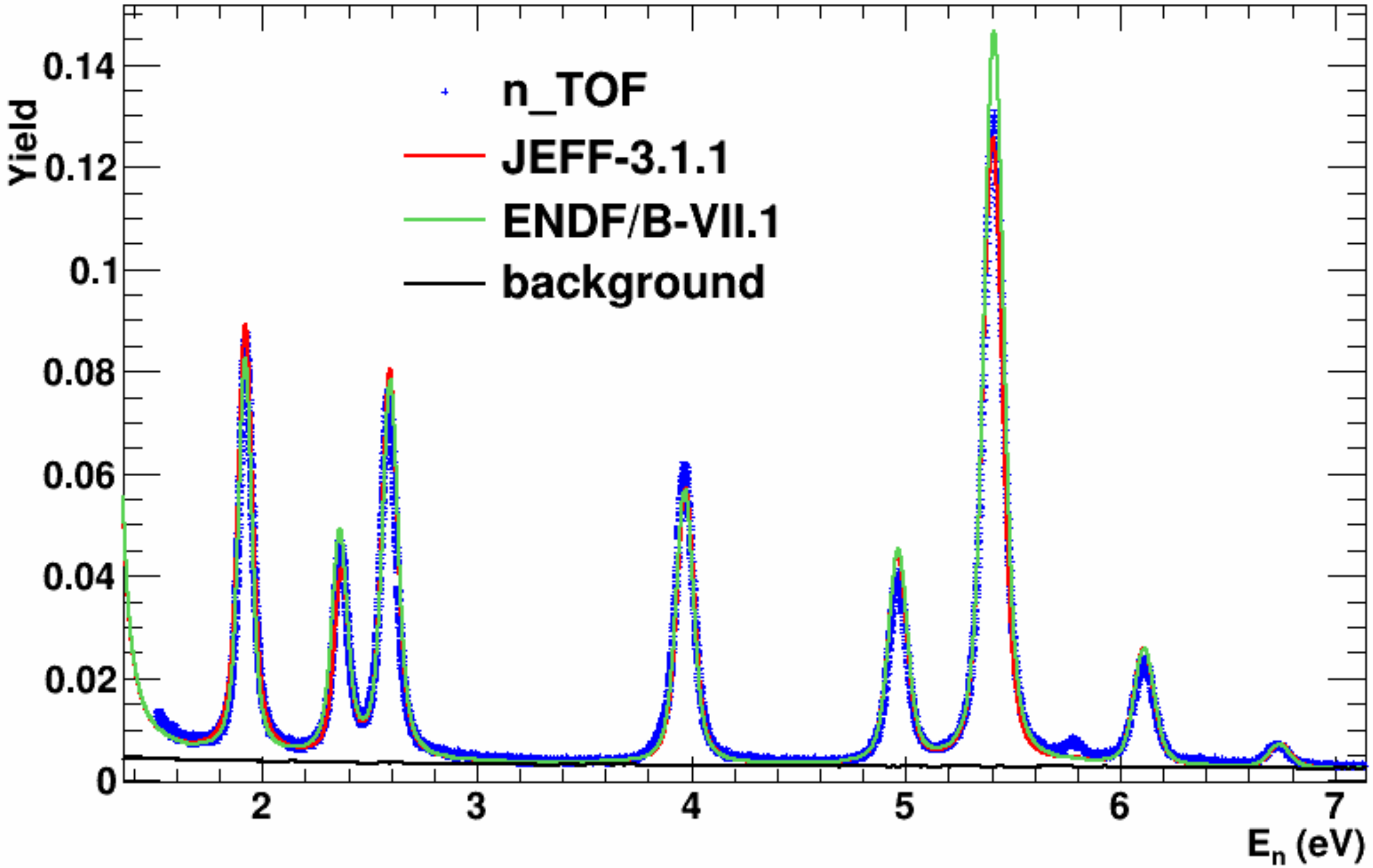
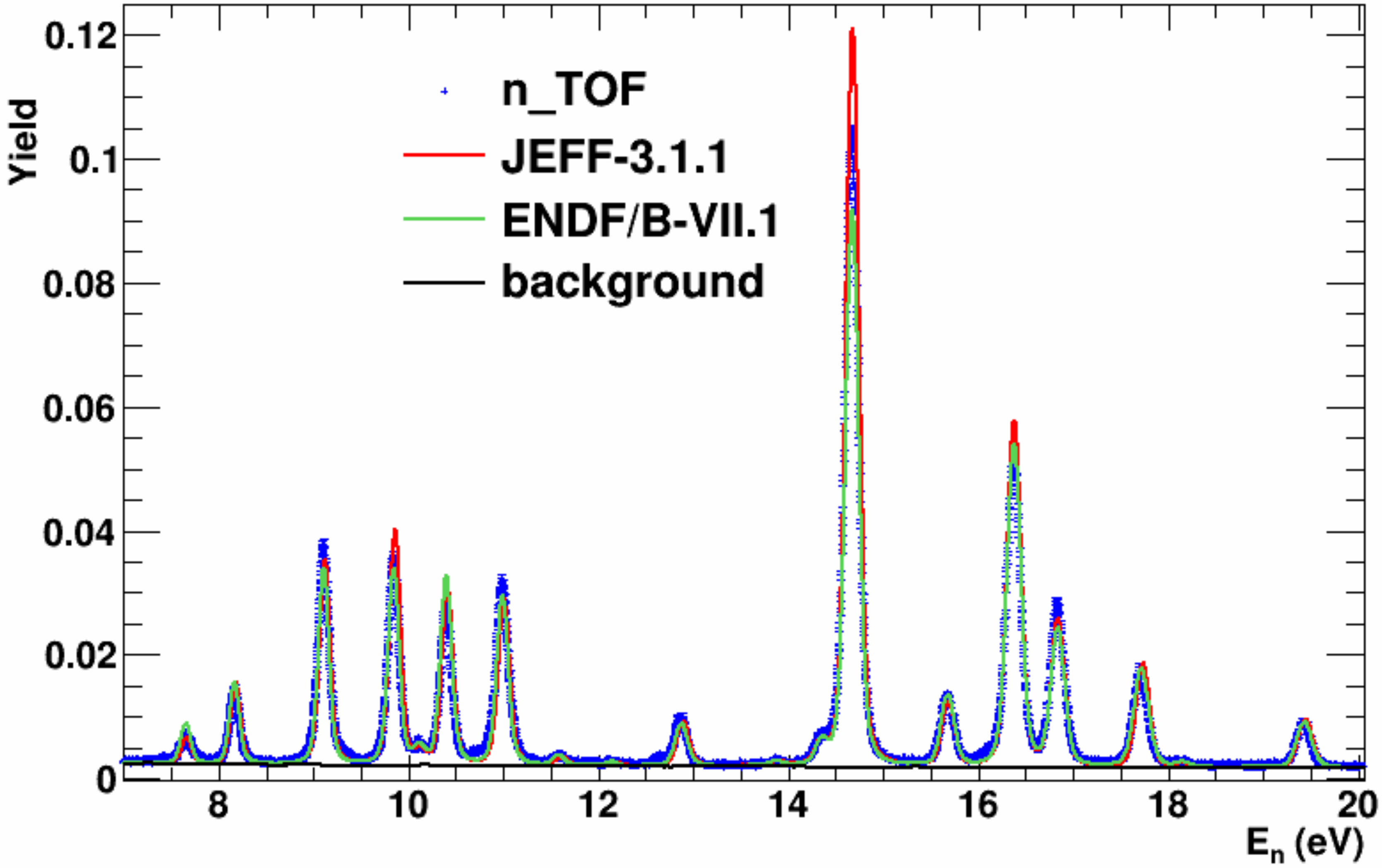
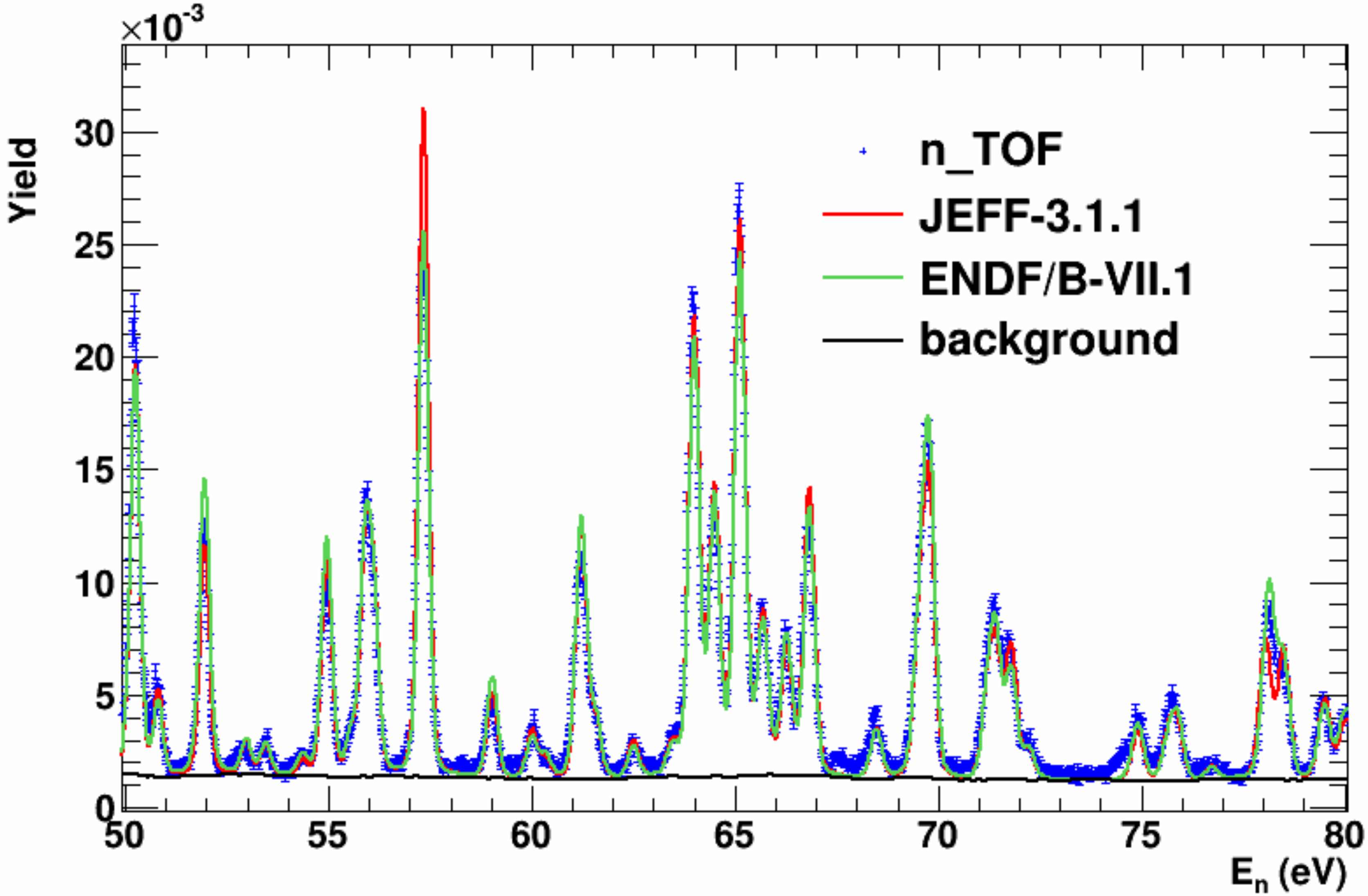
The yield: the resolved resonance region (ii)



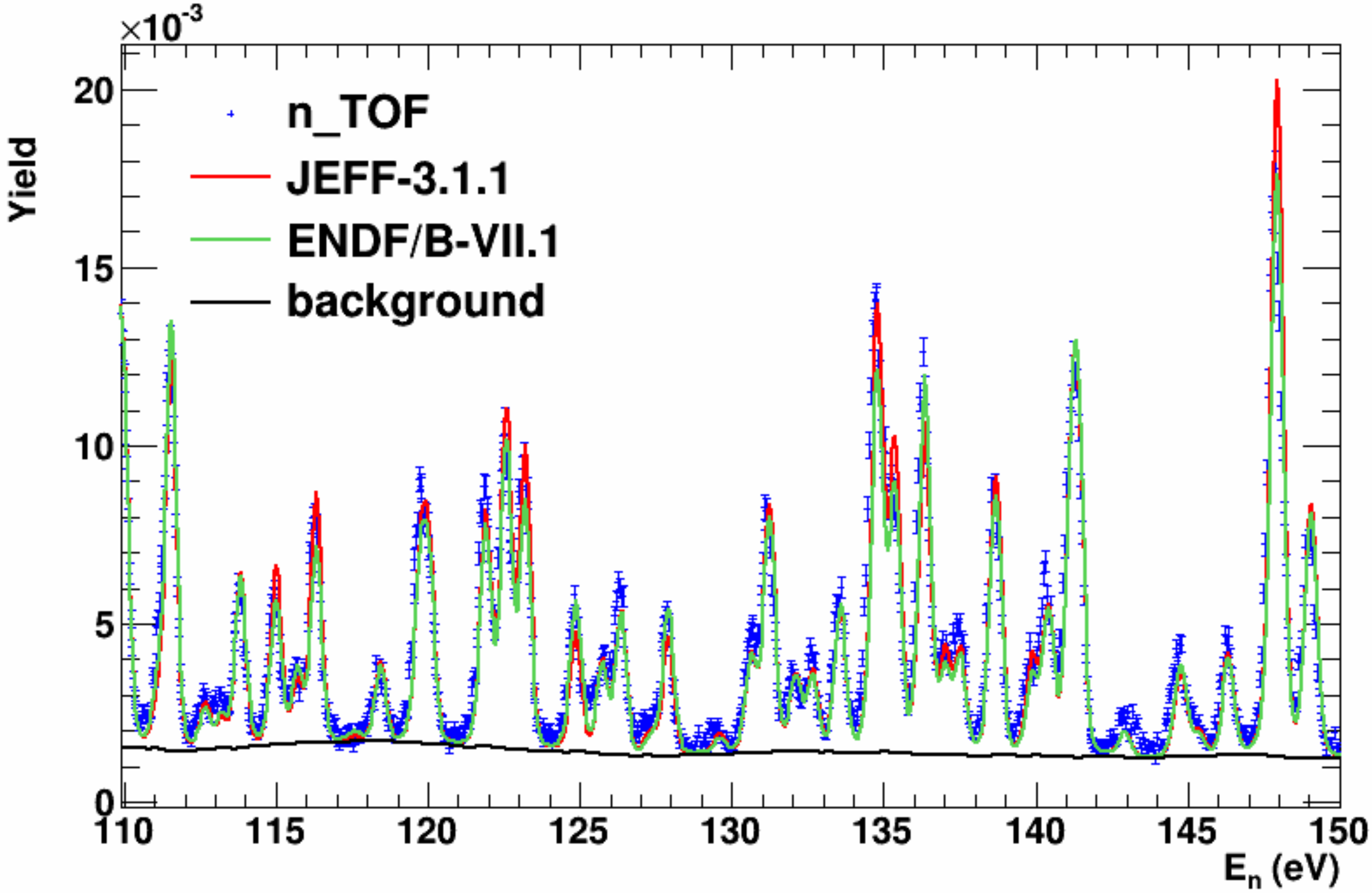
The yield: the resolved resonance region (iii)



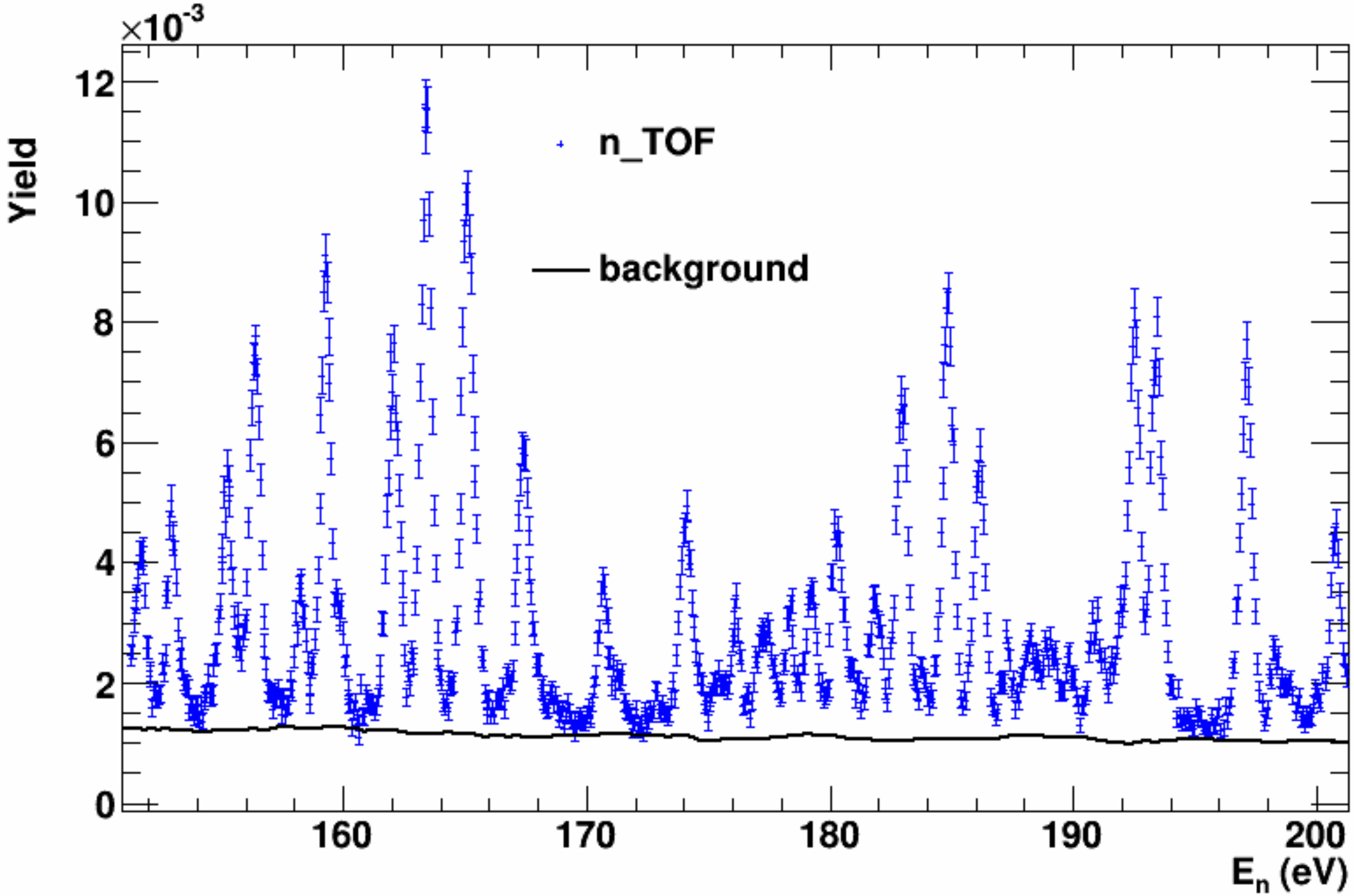
The yield: the resolved resonance region (iv)



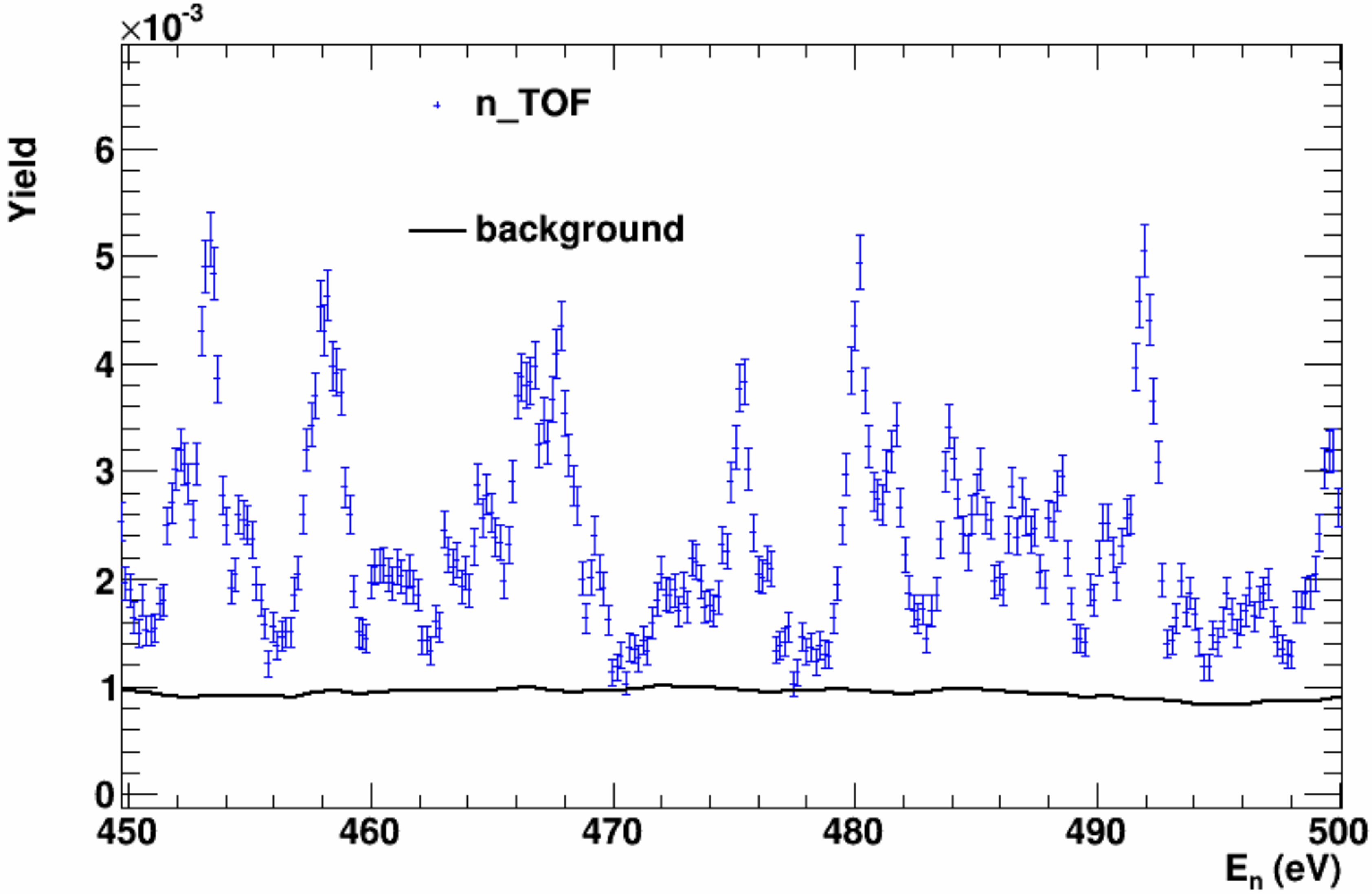
The yield: the resolved resonance region (v)



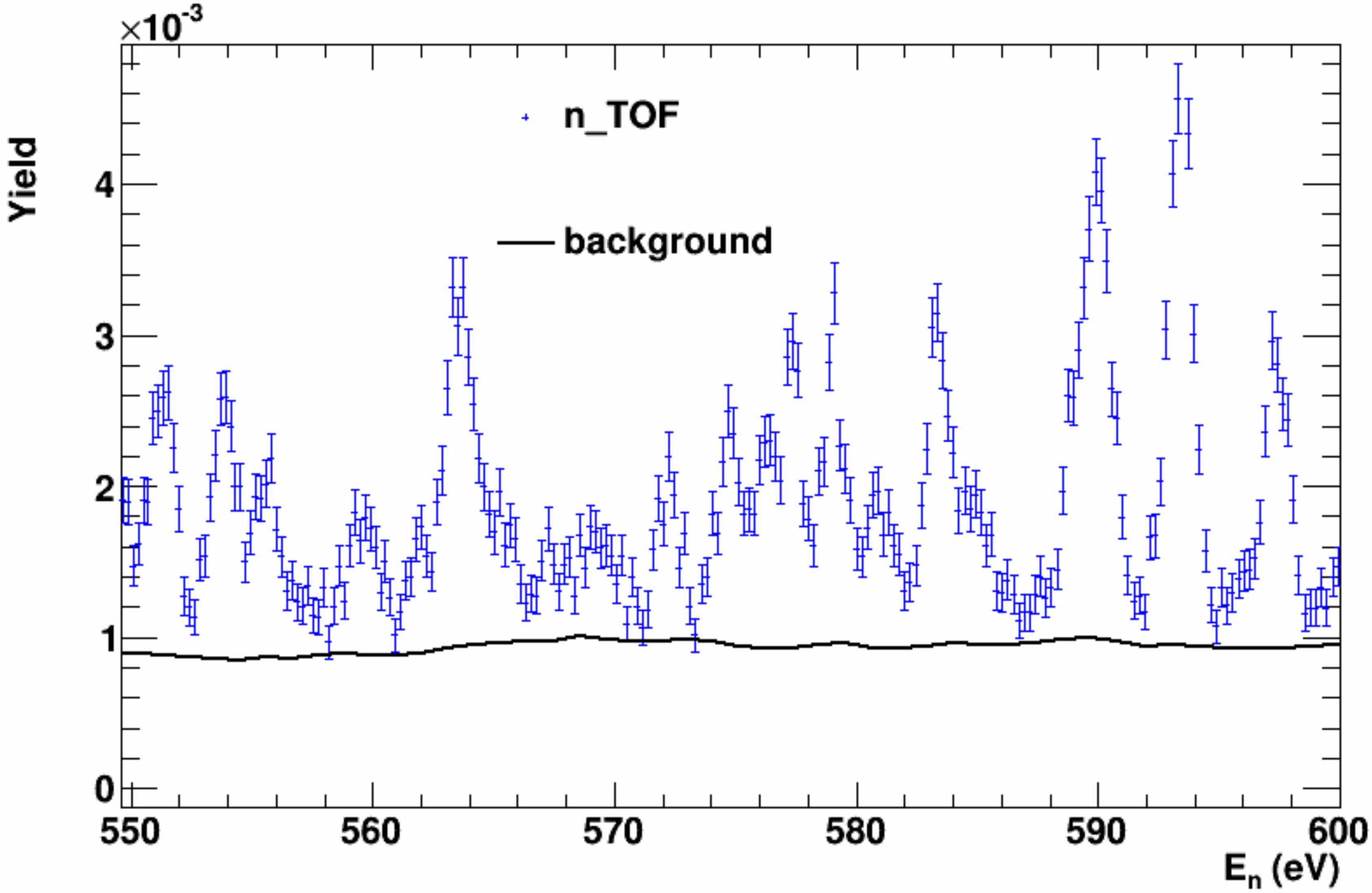
The yield: the extended resolved resonance region (vi)



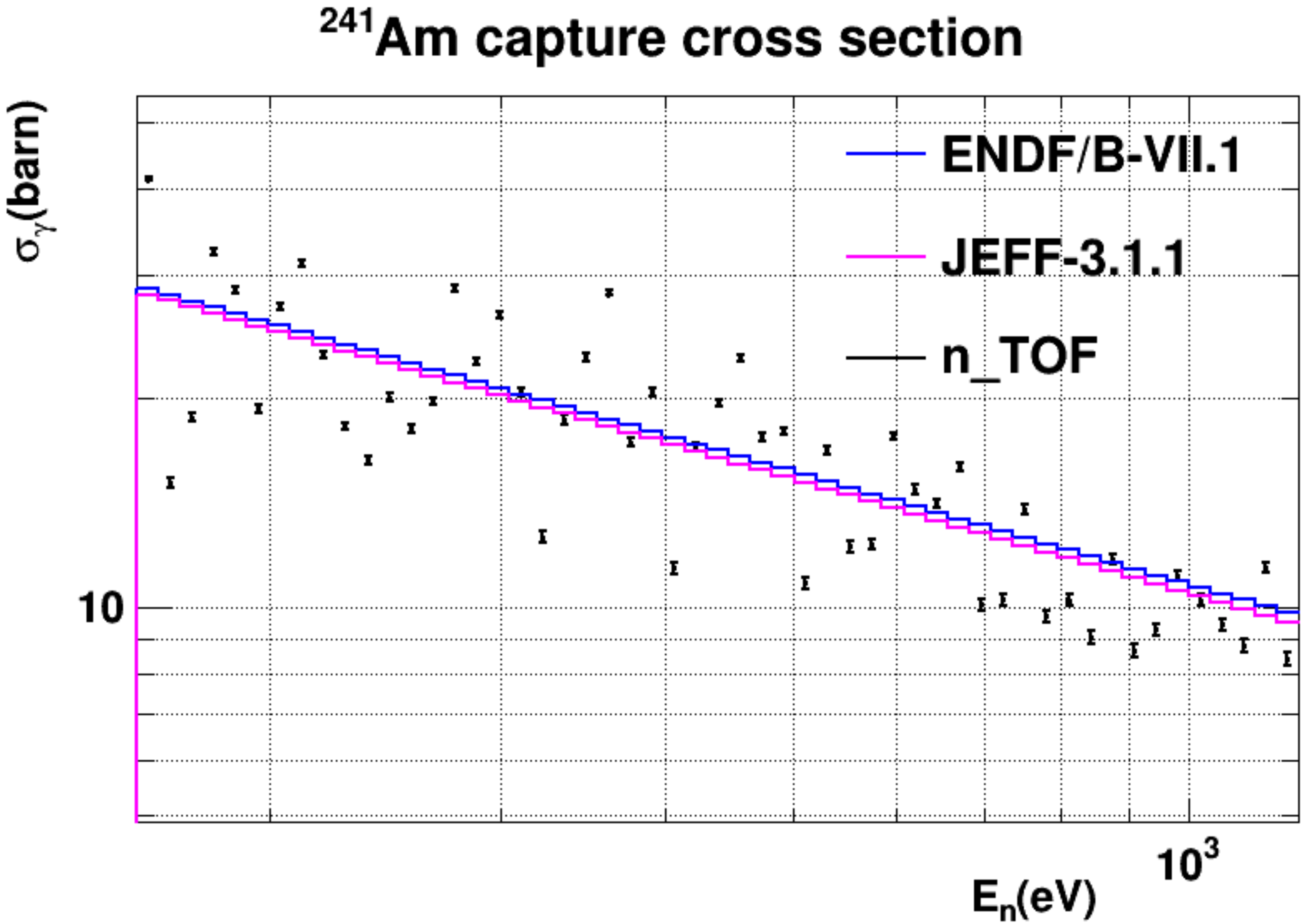
The yield: the extended resolved resonance region (vii)



The yield: the extended resolved resonance region (viii)



The yield: the unresolved resonance region (iv)



Summary and conclusions

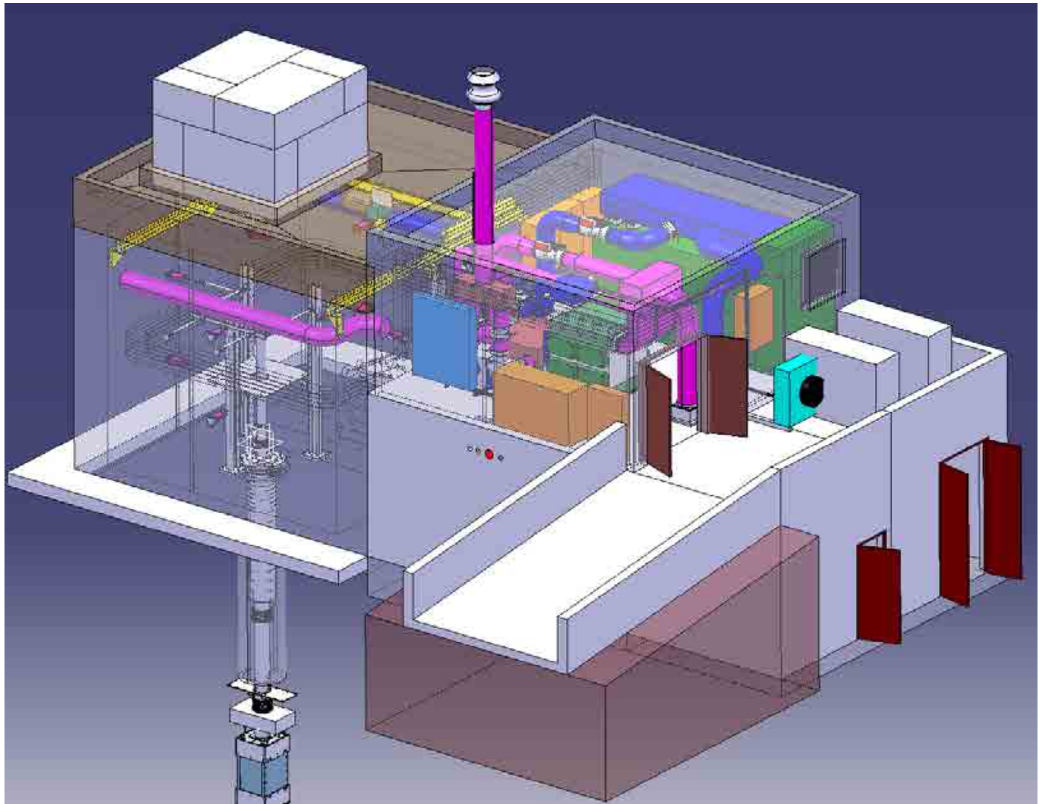
A new $^{241}Am(n,\gamma)$ cross section measurement has been performed successfully at the n_TOF facility at CERN with the Total Absorption Calorimeter. The $^{241}Am(n,\gamma)$ capture yield was obtained in the range between 0.2 eV and 2 keV. The work has been part of the **FP7 EC Project ANDES**.

The obtained capture yield will be evaluated together with recent transmission (and capture) data for reaching an accuracy better than 5%. Without any external normalisation, the technique applied provides data with an average accuracy of 7%.

For the the discussion of the results, the capture yield has been normalized to the capture cross section of the JEFF-3.1.1 evaluated library in the 2-10 eV energy range. The resulting yield has been compared with the ENDF/B-VII.1 and the JEFF-3.1.1 evaluations, finding a reasonable agreement in the entire Resolved Resonance Region, up to 150 eV.

The obtained capture yield allows to extend the description of the cross section in terms of Resolved Resonances from 150 eV up to higher energies, probably up to 400-500 eV. This is a consequence of the large resolving power of the n_TOF facility, together with the excellent statistics achieved in the $^{241}Am(n,\gamma)$ measurement.

The new n_TOF experimental area: EAR-2



25 times more fluence than at EAR-1

10 times shorter flight path (20 m versus 185 m).

A factor of 250 in the signal to background ratio for radioactive samples (lower masses or larger sample activities.

Supported by the FP-7 EC project CHANDA

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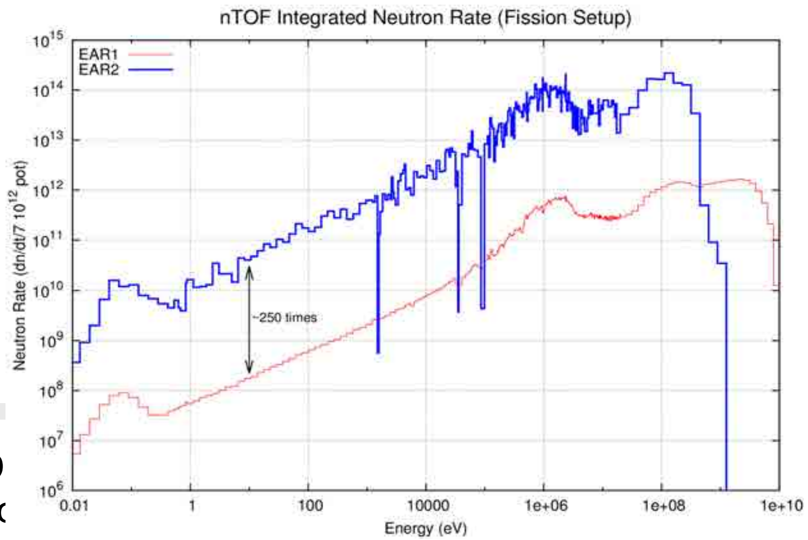

Cross section measurements related to Nuclear Technologies

Periods 2000 – 2004 and 2009 – 2012	
(n,γ)	(n,f)
$^{54,56,57}Fe$	
^{151}Sm	
$^{206,207,208}Pb$	
^{209}Bi	^{209}Bi
^{232}Th	^{232}Th
$^{233,234,235,236,238}U$	$^{233,234,235,236,238}U$
^{237}Np	^{237}Np
^{240}Pu	$^{240,242}Pu$
$^{241,243}Am$	$^{241,243}Am$
	^{245}Cm

Main results:

- Capture and fission cross sections.
- Information on the photon strength functions (Calorimeter and C_6D_6 data)
- Prompt fission γ-rays (calorimeter data).
- Fission fragment angular distributions.

Several new proposals on ^{239}Pu(n,γ) and (n,f), ^{242}Pu(n,γ), ^{244}Cm(n,γ)… are on the way for the two experimental areas. Start in 2014.