

Proposal for the (n, γ) cross section measurements of ^{244}Cm and ^{246}Cm at n_TOF EAR-2

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Motivation

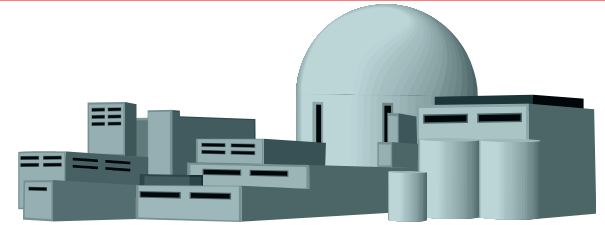
Es242 40 s EC, α	Es243 21 s EC, α	Es244 37 s EC, α	Es245 1.1 m (3/2-) EC, α	Es246 7.7 m (4-,6+) EC, α	Es247 4.55 m (7/2+) EC, α	Es248 27 m (2-,0+) EC, α	Es249 102.2 m 7/2(+) EC, α	Es250 8.6 h (6+) EC, α *	Es251 33 h (3/2-) EC, α
Cf241 3.78 m EC, α	Cf242 3.49 m 0+ α	Cf243 10.7 m (1/2+) EC, α	Cf244 19.4 m 0+ α	Cf245 45.0 m (5/2+) EC, α	Cf246 35.7 h 0+ EC, α ,sf,...	Cf247 3.11 h (7/2+) EC, α	Cf248 333.5 d 0+ α ,sf	Cf249 351 y 9/2- α ,sf	Cf250 13.08 y 0+ α ,sf
Bk240 4.8 m EC	Bk241 (7/2+)	Bk242 7.0 m EC	Bk243 4.5 h (3/2-) EC, α	Bk244 4.35 h (1-) EC, α	Bk245 4.94 d 3/2- EC, α	Bk246 1.80 d 2(-) EC, α	Bk247 1380 y (3/2-) EC, α	Bk248 9 y (6+) EC, α *	Bk249 320 d 7/2+ β^- , α ,sf,...
Cm239 2.9 h (7/2-) EC, α	Cm240 27 d 0+ EC, α ,sf,...	Cm241 32.8 d 1/2+ EC, α	Cm242 162.8 d 0+ α ,sf	Cm243 29.1 y 5/2+ EC, α ,sf,...	Cm244 18.10 y 0+ α ,sf	Cm245 8500 y sf	Cm246 4730 y α ,sf	Cm247 1.56E+7 y α	Cm248 3.4E+5 y α ,sf
Am238 98 m 1+ EC, α	Am239 11.9 h (5/2-) EC, α	Am240 50.8 h (3-) EC, α	Am241 432.2 y 5/2- α ,sf	Am242 16.02 h 1- EC, β^- *	Am243 7370 y 5/2- α ,sf	Am244 9.1 h 5/2- EC, β^- *	Am245 2.05 h (5/2+) β^-	Am246 39 m (7-) β^- *	Am247 23.0 m (5/2) β^-
Pu237 45.2 d 7/2- EC, α *	Pu238 87.7 y 0+ α ,sf	Pu239 24110 y 1/2- α ,sf	Pu240 6563 y sf	Pu241 14.35 y β^- , α ,sf,...	Pu242 3.733E+5 y α ,sf	Pu243 4.9E6 h β^-	Pu244 8.08E+7 y 0+ α , β^- , β^- ,sf,...	Pu245 10.5 h (9/2-) β^-	Pu246 10.84 d 0+ β^-
Np236 1.54E5 y (6-) EC, β^- , α ,...	Np237 2.144E+6 y 5/2+ α ,sf	Np238 2.117 d 2+ β^-	Np239 2.145 d 5/2- β^-	Np240 61.9 m (5+) β^- *	Np241 13.9 m (5/2+) β^-	Np242 5.5 m (6) β^- *	Np243 1.8 m (5/2-) β^-	Np244 229 m (7-) β^-	
U235 7.038E+8 y 7/2- α , β^- ,sf,...	U236 2.342E7 y 0+ α ,sf	U237 6.75 d 1/2+ β^-	U238 4.468E+9 y 0+ α , β^- , β^- ,sf,...	U239 3.285 m 5/2- β^-	U240 14.1 h 0+ β^-	U241	U242 16.8 m 0+ β^-		

Neutron Capture Cross Sections of

- **minor actinides (MAs) and**
- **long-lived fission products (LLFPs)**

are important.

- for improving the performance and safety of our actual reactors,
- for designing new types of reactors,
for reducing the high-level radioactive Waste (transmutation),



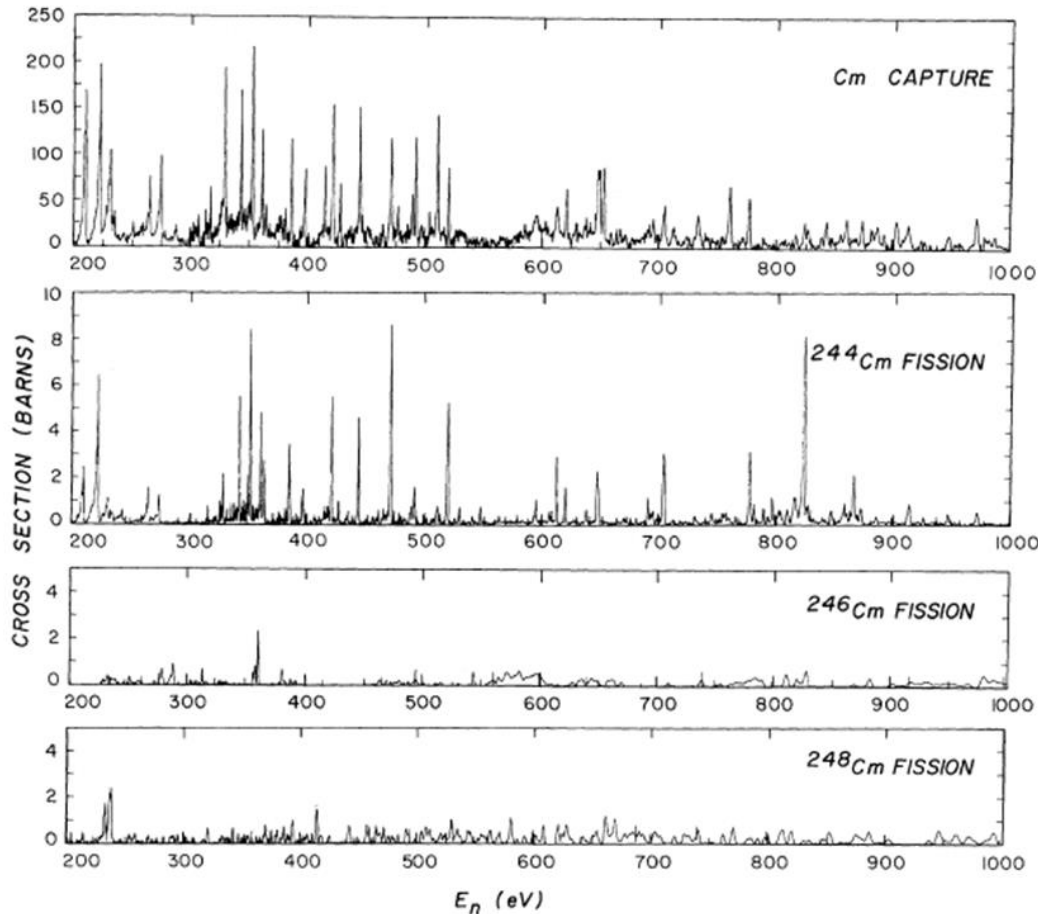
The reported uncertainties of c.s. Libraries are (too) often questionable!



Especially, ^{244}Cm and ^{246}Cm are **very important**:

- Share nearly 50% of the total actinide **decay heat** in spent reactor fuels even after three years of cooling.
- ^{244}Cm is one of the main **neutron emitters** in the irradiated nuclear fuel (fuel safety).
- Both capture and fission cross sections (transmutation) are **known poorly**.
- Cm isotopes open the path to the production of higher Z elements: Bk, Cf...
- **Only two previous measurements** available (extreme difficulties).

Experiment by Moore et al.



This experiment was performed with the neutron time-of-flight (TOF) method in 1969 **using a under-ground nuclear explosion (one single pulse)** as a pulsed neutron source and Moxon-Rae detectors operated in current mode.

The data had to be corrected with some assumptions, because the **sample** had been **shifted from the correct position** in their experiment.

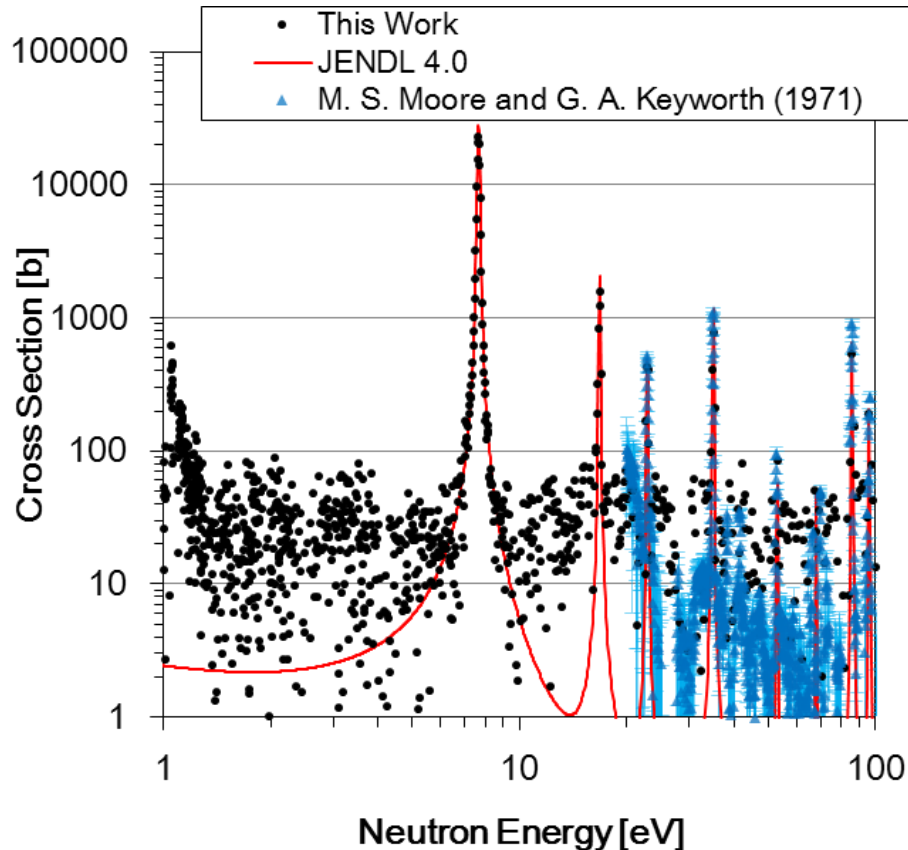
→ **Accuracy questionable due to systematic uncertainties**

[1] M. S. Moore and G. A. Keyworth, Physical Review C, 3, 1656 -1667 (1971)

Experiment by Kimura et al.

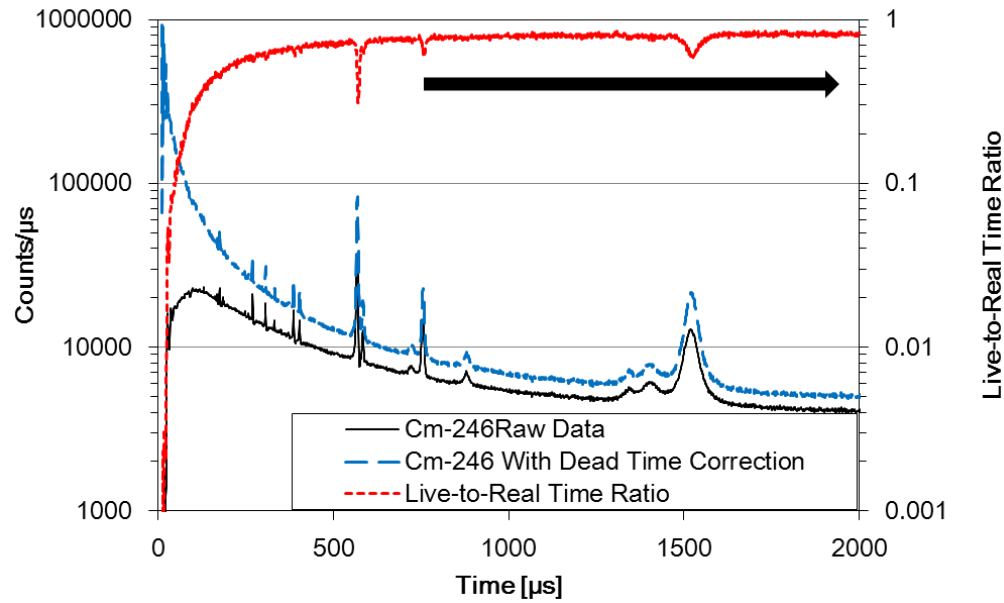
In the experiment at J-PARC Ge detectors were used for detecting the (n, γ) cascades.

- In the neutron energy range **above 100 eV**, the measurement at J-PARC required severe **dead time corrections (up to 90%)** due to the high counting rates and the long decay times of the detectors.
- Dependence on the electromagnetic de-excitation pattern.

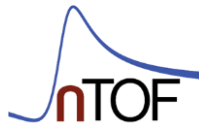


As a result the upper-limits of the measured neutron energy range were restricted to 100 eV.

A. Kimura et al., J. Nucl. Sci. Tech. 49, 708 (2012).



The experiment proposed at EAR-2



The uncertainties of the reported data sets are much larger than the required accuracy of 4.1–25.7% especially in the neutron energy range over 100eV.

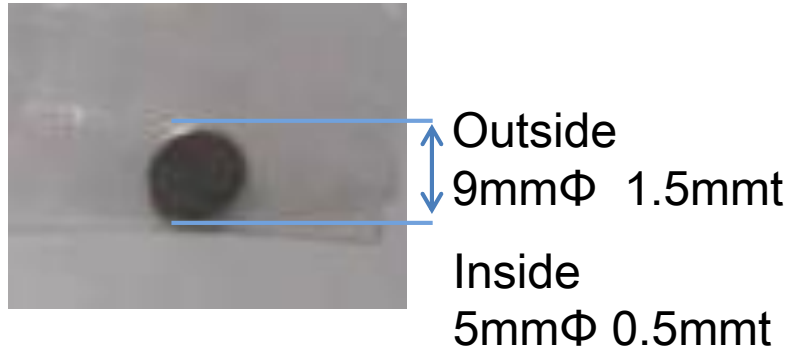
Advantages/improvements of the experiment proposed at n_TOF EAR-2:

- The EAR-2 **neutron rate is 250 times larger than at EAR-1** (25 time more fluence in 10 times less time) which permits to reduce to reduce the influence of background due to decay γ -rays (i.e. smaller time windows). The n_TOF EAR-2 was built for the measurement of highly radioactive samples.
- The use of a **high performance digital acquisition system** (12/14 bits resolution, 1 Gsample/s)
- The use of the **total energy detector technique** based on very **fast (40 ns vs ~100 μ s)** C_6D_6 detectors.

We expect as well to be able to obtain results in the range up to 300 eV.

The Cm samples

Two sealed ^{244}Cm samples and one ^{246}Cm sample are available and will be provided by JAEA.



Samples:

^{244}Cm ($T_{1/2}=18.1\text{y}$: MA)

Net weight = 0.6 mg

Activity = 1.8 GBq (x2)

^{246}Cm ($T_{1/2}=4753\text{y}$: MA)

Net weight = 2.1 mg

Activity = 12.1 MBq
(^{244}Cm : 1.7GBq)

Both of the samples

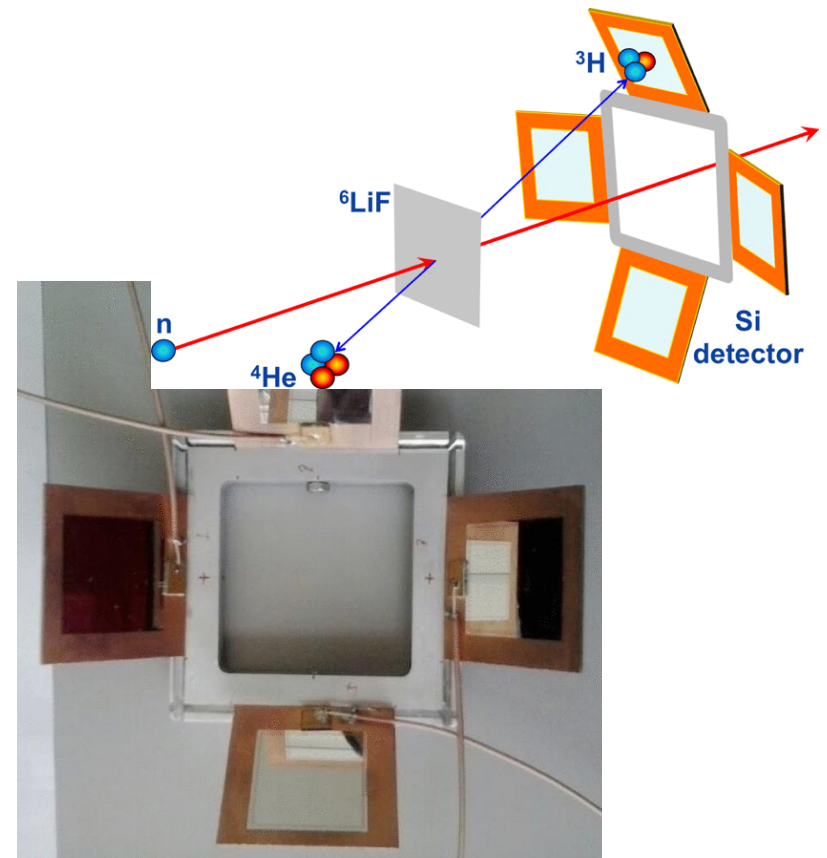
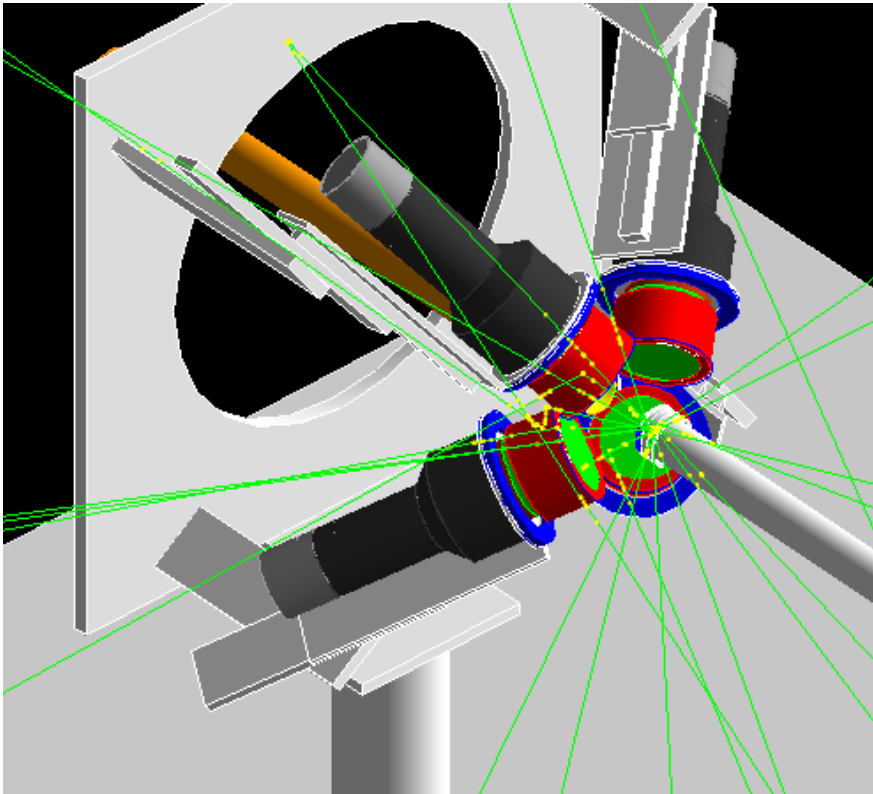
Chemical form = CmO_2

Container = Al capsule

	^{244}Cm sample (mole %)	^{246}Cm sample (mole %)
^{244}Cm	88.5 \pm 1.7 (0.46 mg)	24.1 \pm 0.4 (0.50mg)
^{245}Cm	3.14 \pm 0.40 (0.02 mg)	1.11 \pm 0.29 (0.02 mg)
^{246}Cm	8.36 \pm 0.39 (0.04 mg)	62.3 \pm 1.4 (1.30mg)
^{247}Cm	-----	2.99 \pm 0.37 (0.06mg)
^{248}Cm	-----	9.53 \pm 0.25 (0.20mg)
^{240}Pu	32.1 \pm 0.8 (0.17mg)	7.42 \pm 0.15 (0.15mg)

The detector setup

- We will use a set of four C_6D_6 detectors (20% efficiency). These detectors have been used successfully during the first (n, γ) cross measurements performed at EAR-2 in 2015.
- The neutron beam intensity will be monitored with a neutron transparent silicon flux monitor.

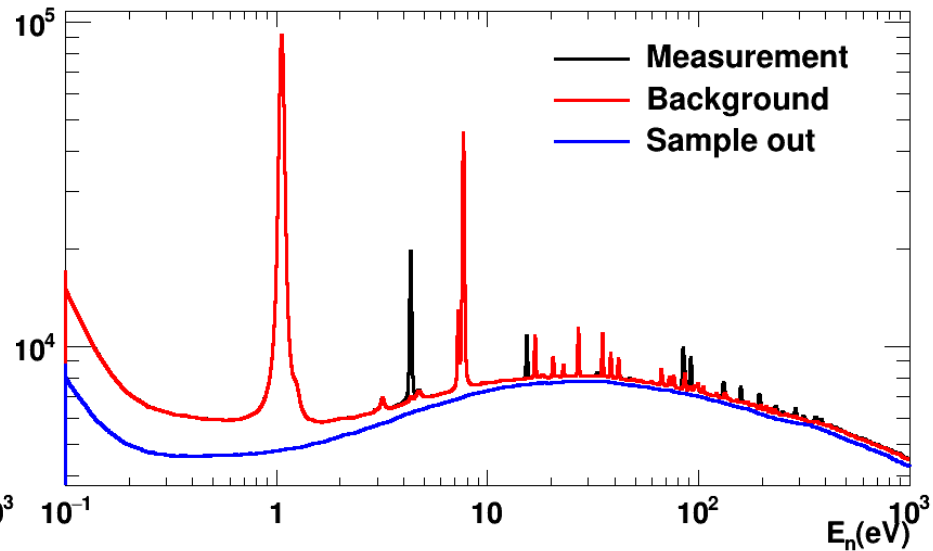
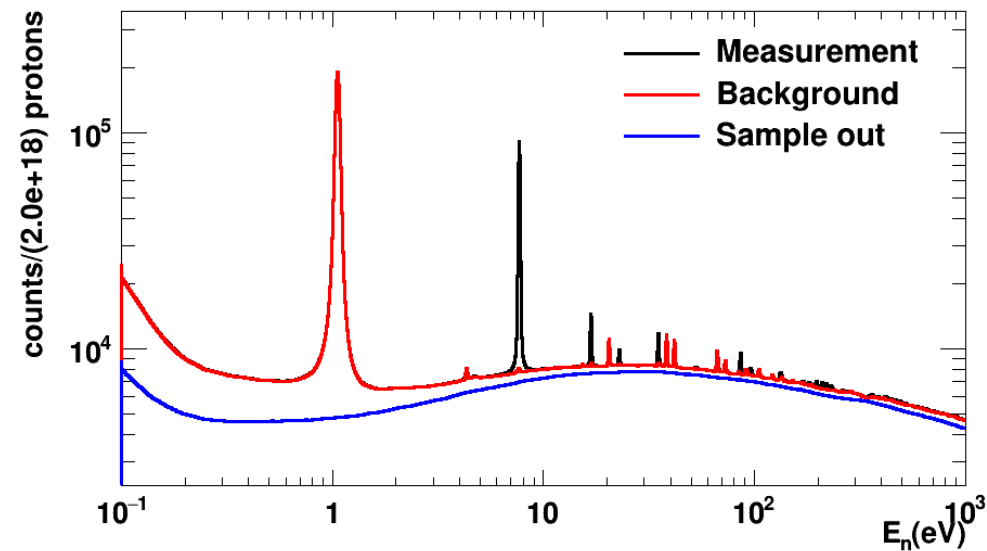


Rate estimates for the proposal

- A 20% total detection efficiency for the setup with 4 x C_6D_6 detectors
- The **EAR-2 background** has been **determined experimentally (in 2015)**. An attempt for **optimising the C_6D_6 setup** for reducing the background (with active/passive shielding) is currently **in progress**.
- The **background** due to **elastic** (and fission) **reactions** has been estimated with a 0.1% neutron sensitivity of the C_6D_6 detectors.
- An efficiency for detecting **fission reactions** 2 times larger as the efficiency for capture.

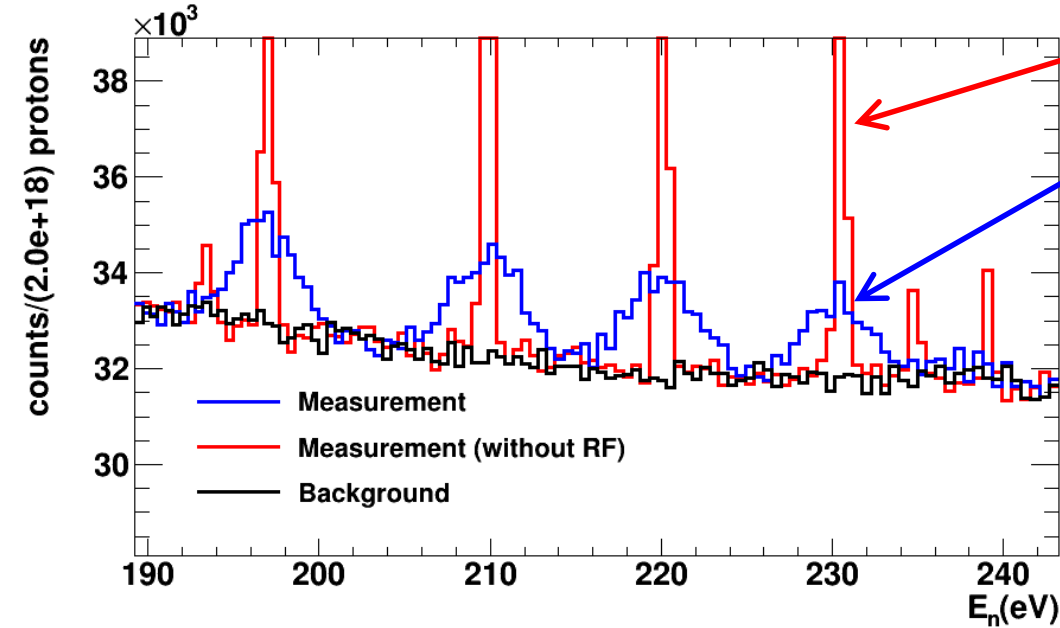
5000 BPD

5000 BPD



The effect of the resolution function

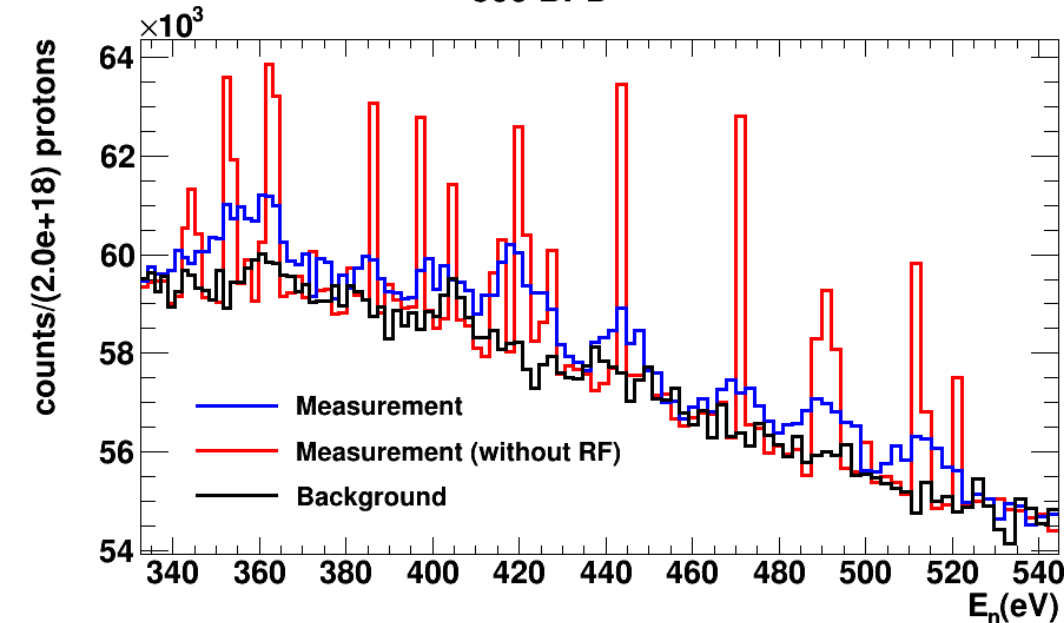
1000 BPD



Good statistics is required because:

- The **resolution function** worsens the peak to background ratio.
- The **background** will have to be subtracted.

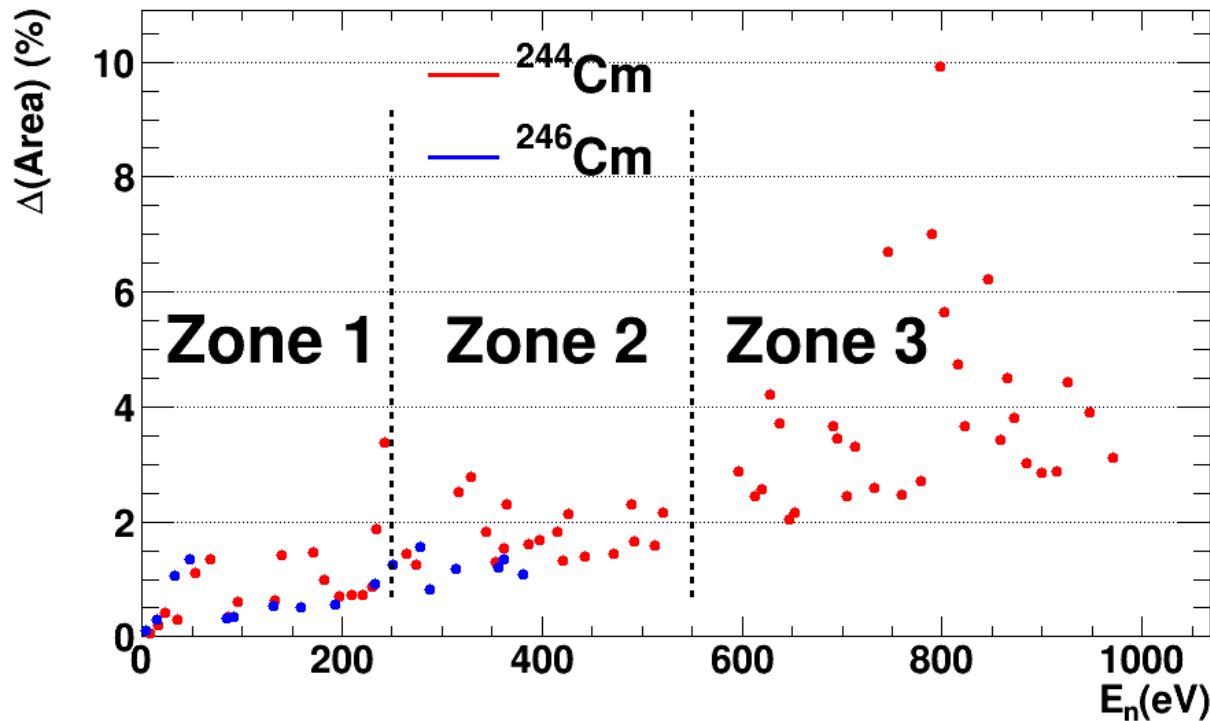
500 BPD



(*) expected number of counts per 2×10^{18} protons in the ^{244}Cm measurement, using 500 bins per decade.

Estimated statistical uncertainties

Estimated statistical uncertainties in the area of the resonances with 2×10^{18} protons.



- **Zone1**, between 1 eV and 200 eV. The measurement proposed at EAR-2 will allow to get resonance integrals with an overall uncertainty below 5%.
- **Zone2**, between 200 eV and 550 eV. The uncertainty in the results in this region will depend largely on the real cross section values and background.
- **Zone3**, between 550 eV and 1 keV. If the experimental conditions are much better than expected, this region could be analysed as well.

Proton request

Number of protons requested for each measurement in EAR-2.

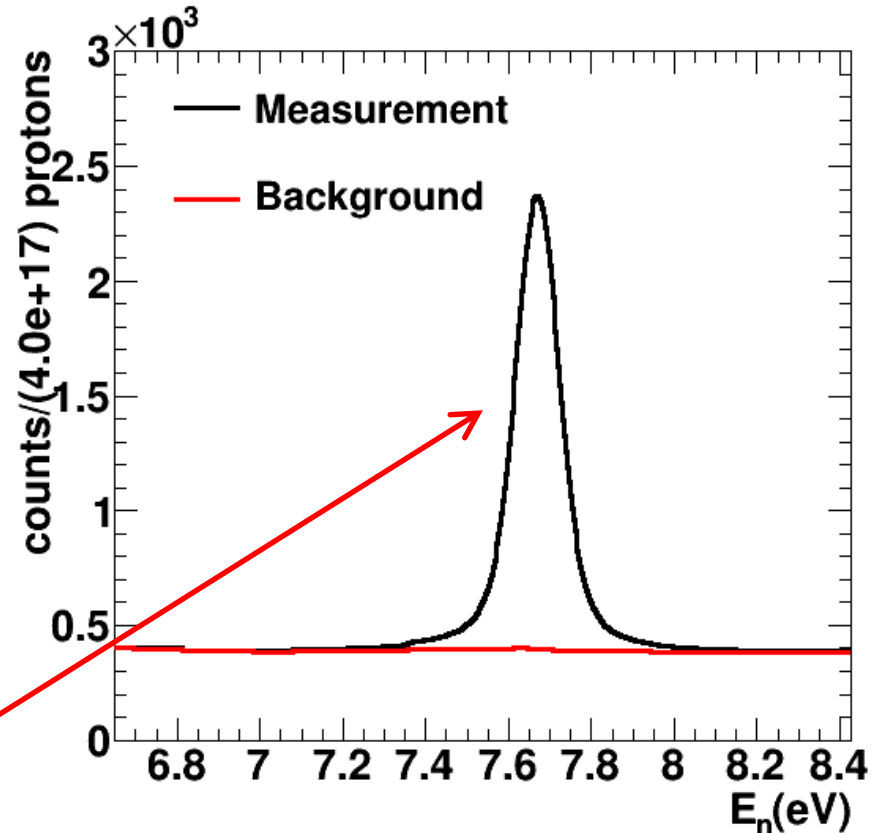
Sample	Purpose	Protons
$^{244}\text{Cm}(n,\gamma)$	capture cross section measurement	2.0×10^{18}
$^{246}\text{Cm}(n,\gamma)$	capture cross section measurement	2.0×10^{18}
Dummy – ^{244}Cm	Canning related background	1.0×10^{18}
Dummy – ^{246}Cm	Canning related background	1.0×10^{18}
Sample out	Beam related background	1.0×10^{18}
Graphite	Neutron Sensitivity	0.5×10^{18}
$^{197}\text{Au}(n,\gamma)$	Validation of the capture measurement	0.5×10^{18}
$^{244}\text{Cm}/^{246}\text{Cm}/-$	No beam related background	0
Total		8.0×10^{18}

Additional Measurement at EAR-1

5000 BPD

Short measurement with the TAC in EAR-1:

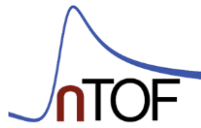
- To have an **alternative (total absorption) normalization** to the ^{240}Pu 1 eV resonance. The systematic uncertainty in the normalization is the largest contribution in the overall uncertainty.
- To obtain **spectroscopic information** about the **γ -ray cascades** following the $^{244}\text{Cm}(n,\gamma)$ and $^{240}\text{Pu}(n,\gamma)$ reactions.



Number of expected counts in the TAC in strongest resonance in the $^{244}\text{Cm}(n,\gamma)$ c.s. for 4.0×10^{17} protons.

Sample	Purpose	Protons
$^{244}\text{Cm}(n,\gamma)$	Capture cascades of the strongest ^{240}Pu and ^{244}Cm resonances	0.4×10^{18}
Dummy – ^{244}Cm	Beam related background	0.1×10^{18}
^{244}Cm	Background with no correlation with the beam	0
Total		0.5×10^{18}

Summary and conclusions



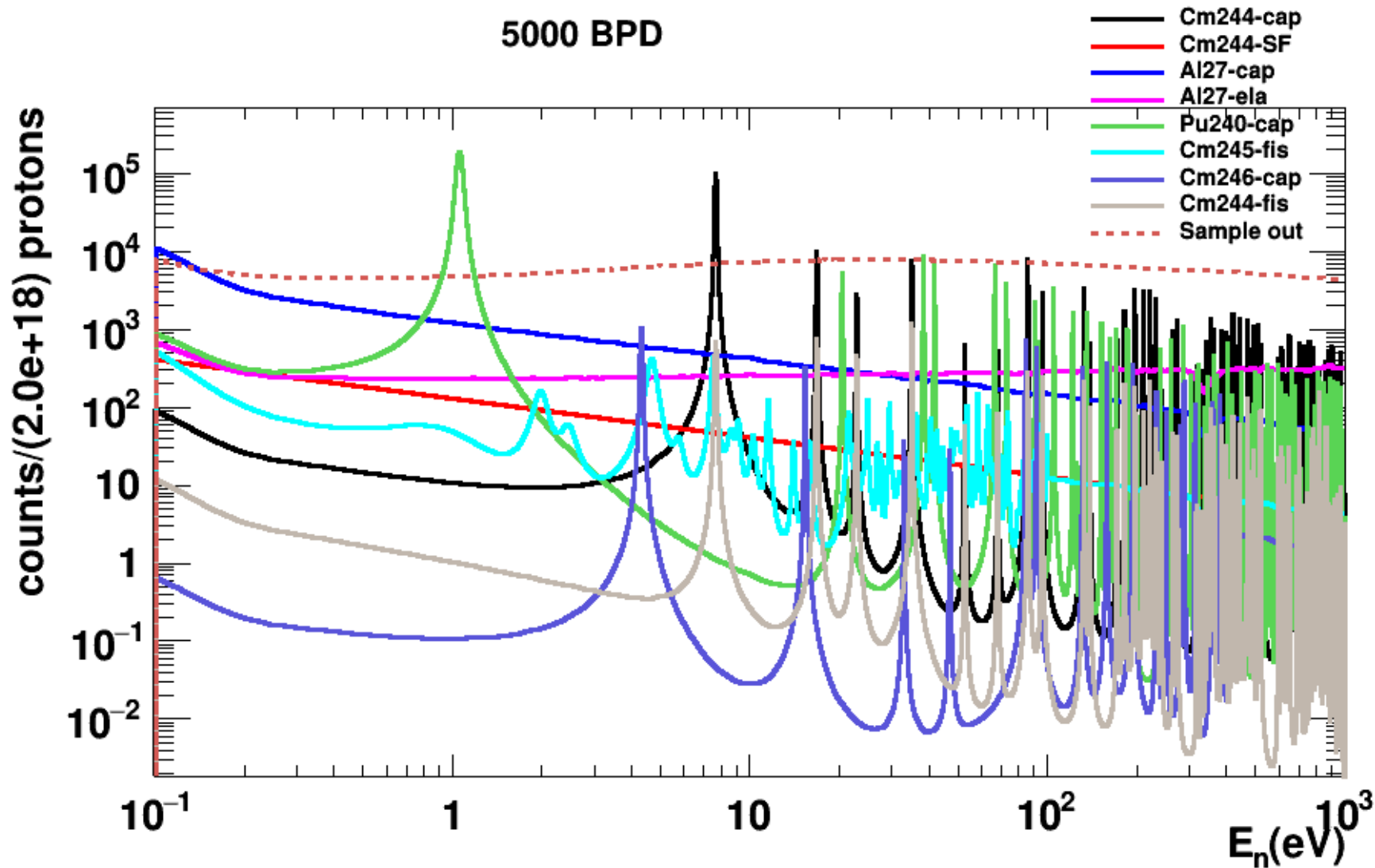
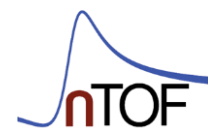
- We propose to measure the capture cross sections of ^{244}Cm and ^{246}Cm at the n_TOF EAR-2 and perform a short measurement at EAR-1 for getting an addition normalization and spectroscopic information (statistical de-excitation models of the nucleus, photon strength functions).
- Both measurements have to be performed with the same systematics since the two isotopes appear in the two samples with different weights.
- Both ^{244}Cm and ^{246}Cm cross sections are required to estimate the production and transmutation Cm and heavier isotopes.
- This is the type of measurements for which EAR-2 was built (250 times more neutron rate than at EAR-1) and will have a large impact/visibility in the nuclear data for nuclear technologies community.

We request a total of:

- 8×10^{18} protons for the measurement at EAR-2
- 0.5×10^{18} protons at EAR-1.

SPARE SLIDES

Background components in the ^{244}Cm experiment



Background components in the ^{246}Cm experiment

