

Measurement of the capture cross section of ^{244}Cm & ^{246}Cm

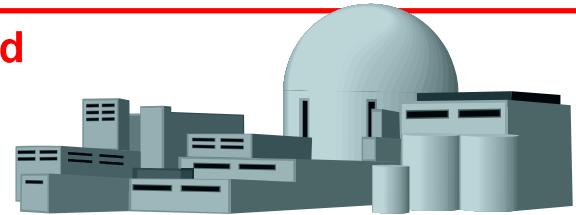
¹V. Alcayne, ²A. Kimura, ¹E. Mendoza, ¹D. Cano Ott
and the n_TOF Collaboration

¹CIEMAT– Spain
²JAEA – Japan

Motivation

Neutron Capture Cross Sections of minor actinides (MAs) and long-lived fission products (LLFPs) are important.

- Improving the performance and safety of actual reactors.
- Designing new types of reactors, for reducing the high-level radioactive waste (transmutation).



The reported uncertainties of C.S. libraries are often questionable.

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

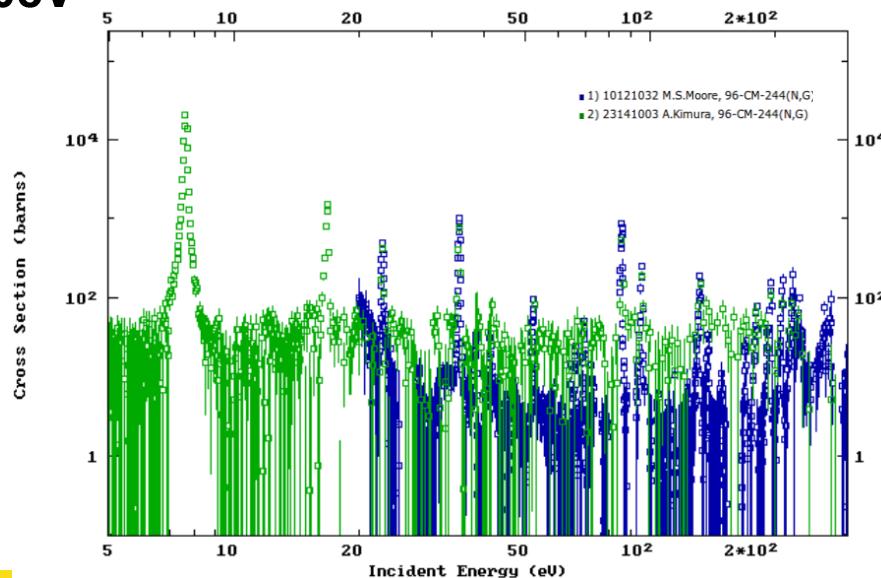
- Share nearly 40-50% of the total actinide decay heat in spent reactor fuels even after three years of cooling.
- ^{244}Cm is one of the main **neutron emitters** in the irradiated nuclear fuel (fuel safety).
- Cm isotopes open the path to the production of higher Z elements: Bk, Cf...
- Both capture and fission cross sections (transmutation) are **known poorly**.
- **Only two previous measurements** available.

Only 2 sets of previous data

Experiment by Moore et al.

- 1969 Using underground nuclear explosion
- Moxon-Rae detectors
- Accuracy questionable due to systematic uncertainties
- No data under 20eV

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



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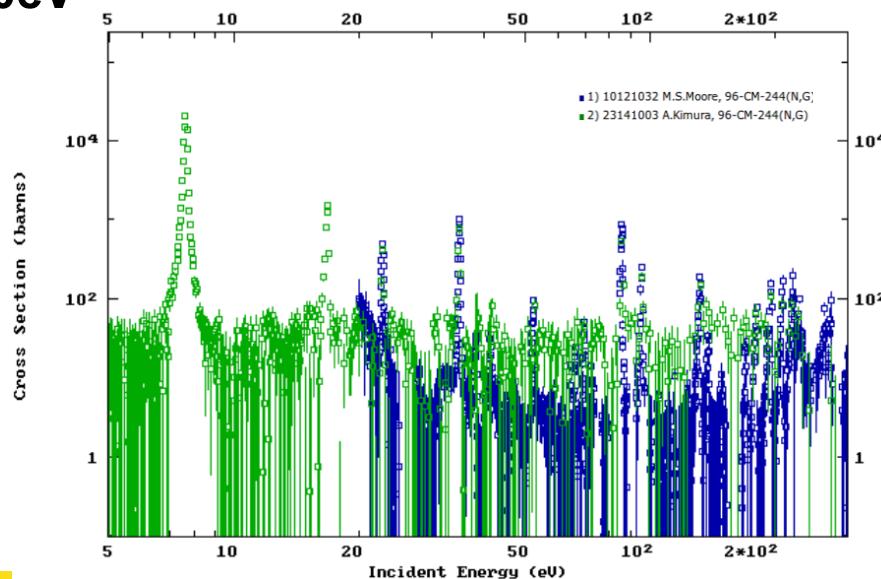
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Experiment by Moore et al. Experiment by Kimura et al.

- 1969 Using underground nuclear explosion
- Moxon-Rae detectors
- Accuracy questionable due to systematic uncertainties
- No data under 20eV

- 2010 at J-PARC
- Two cluster-Ge detectors
- Above 100 eV, the measurement required severe dead time corrections (up to 90%)
- Resonance analysis up to 30 eV



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

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

Measurement at n_TOF

Collaboration between CIEMAT, JAEA and n_TOF

~1 mg ^{244}Cm and ^{246}Cm samples with high activity (~1 GBq)

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C₆D₆ in EAR2

TED

High intensity / 20m flight path

Worse RF

TAC in EAR1

TAC

Lower intensity / 185m flight

Better RF

Information EM cascade



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Measure the Cross Sections of ^{244}Cm and
 ^{246}Cm for improving the uncertainty
assessment and extending the energy range

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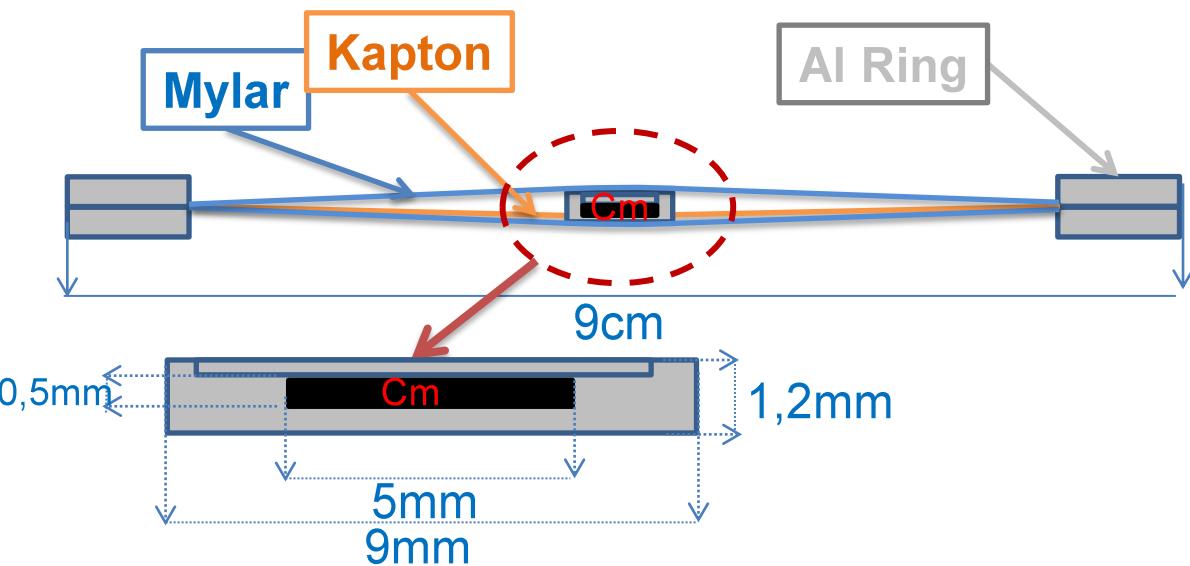
Measurement during **May-September 2017**
Relative measurement to $^{240}\text{Pu}(n,\gamma)$

Samples



Japan Atomic
Energy Agency

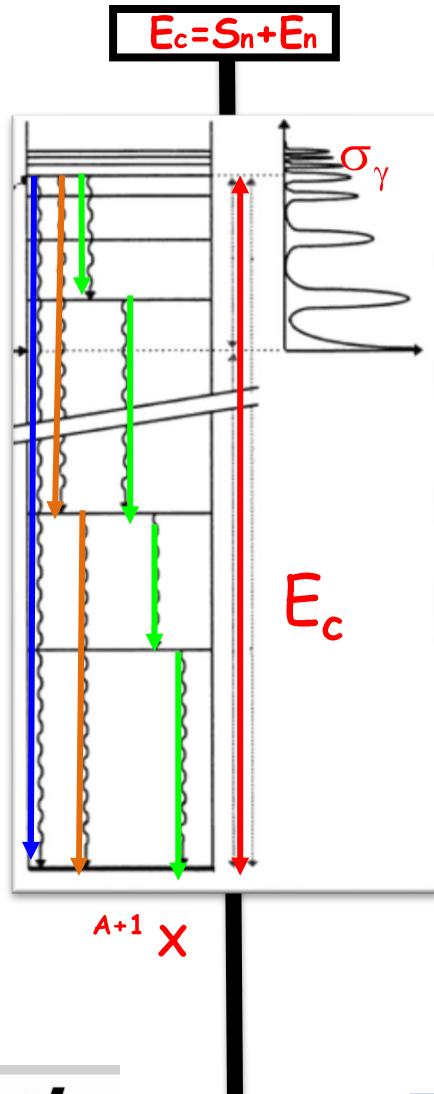
	Two ^{244}Cm sample (%)	^{246}Cm sample (%)
^{244}Cm	60.6 ± 1.1 (~0.80 mg)	20.3 ± 0.5 (~0.38mg)
^{245}Cm	2.38 ± 0.30 (~0.02 mg)	1.03 ± 0.29 (~0.02 mg)
^{246}Cm	6.35 ± 0.55 (~0.08 mg)	57.7 ± 1.5 (~1.10mg)
^{247}Cm	----	2.8 ± 0.4 (~0.05mg)
^{248}Cm	----	8.8 ± 0.2 (~0.17mg)
^{240}Pu	31.1 ± 0.6 (~0.40mg)	9.30 ± 0.15 (~0.17mg)



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Neutron Capture measurement techniques



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J.Lerendegui (U.S.) Private communication

Neutron Capture measurement techniques

TED
(Total Energy Detector)

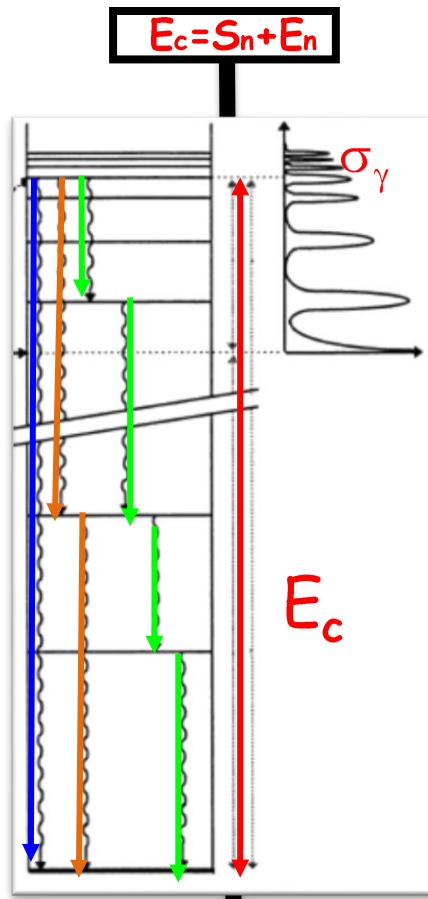
I.) Low Efficiency Detectors:

II.) Efficiency to detect a γ -ray is proportional to its energy

III.) Proportionality fulfilled with Weighting factors

$$\varepsilon_c = \sum_{i=1} \varepsilon_{\gamma i} = k \sum_{i=1} E_{\gamma i} = kE_c$$

The detection efficiency is proportional to E_c



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Neutron Capture measurement techniques

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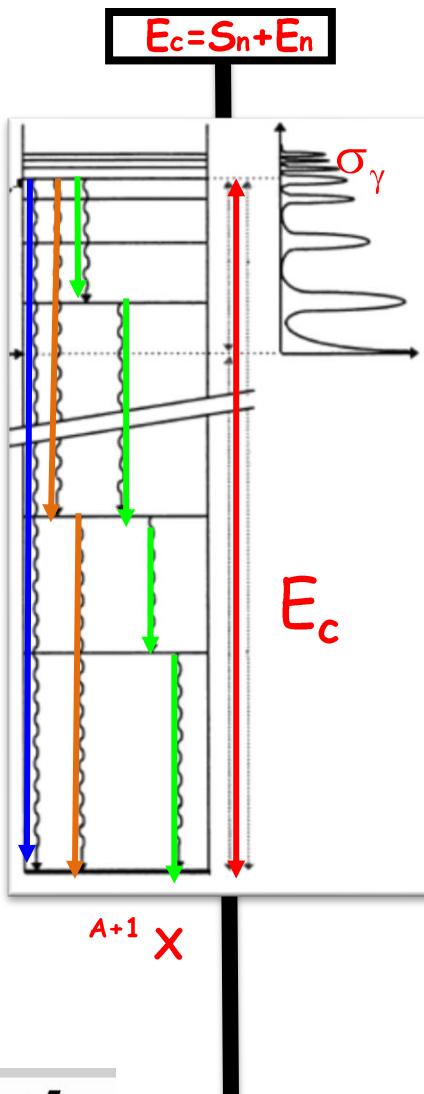
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$$\varepsilon_c = \sum_{i=1}^n \varepsilon_{\gamma i} = k \sum_{i=1}^n E_{\gamma i} = kE_c$$

The detection efficiency is proportional to E_c

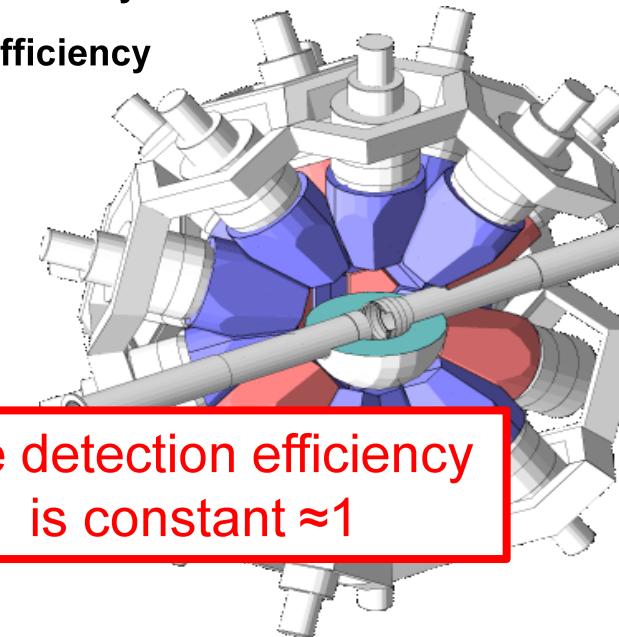


TAC
(Total Absorption Calorimeter)

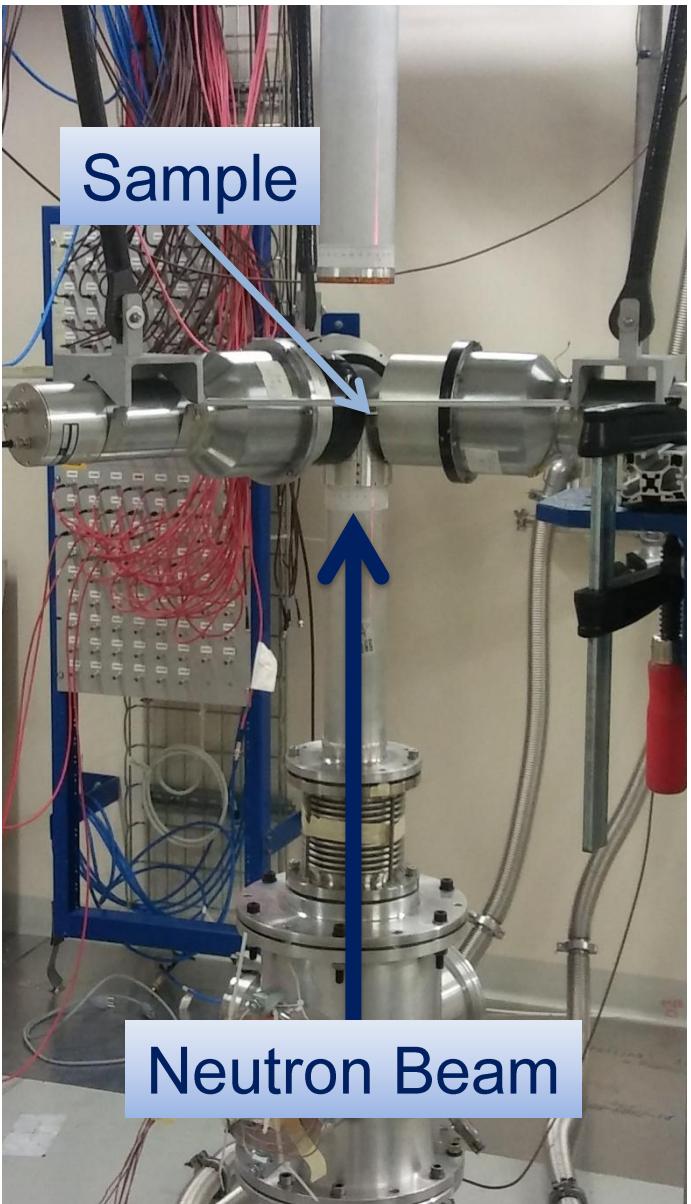
If intrinsic and angular efficiencies are large:

I.) Total efficiency of the cascade : 4π

II.) Peak efficiency



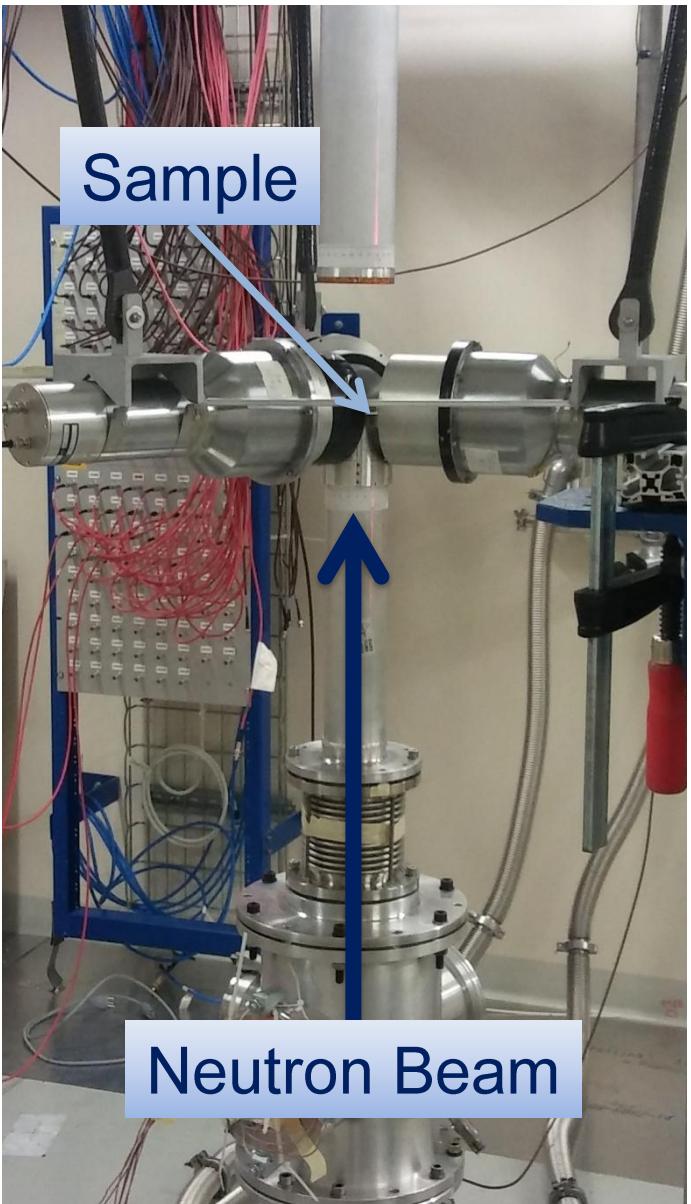
The detection efficiency is constant ≈ 1



C₆D₆ Setup EAR2 (~20 m)

- Three detectors
- Efficiency proportional to energy ($\varepsilon_{\gamma i} \propto E_{\gamma i}$) achieved using PHWT.
- Good time resolution (~1ns).





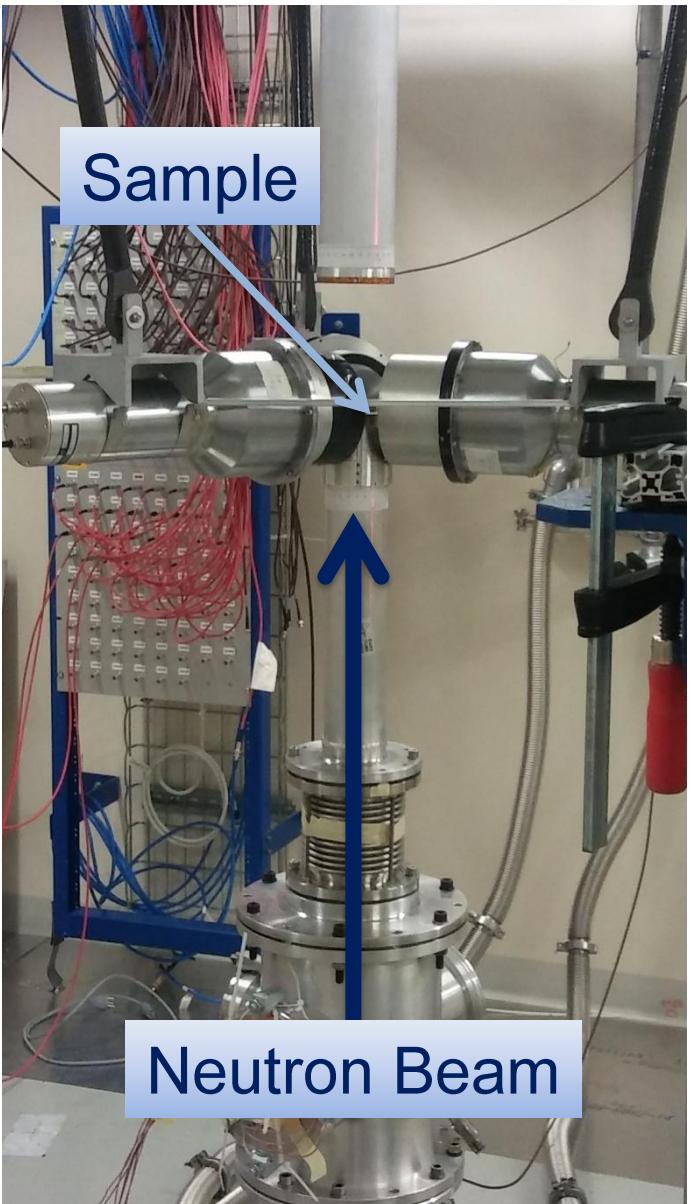
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• Experiment

- 3 months.
- $8 \cdot 10^{18}$ protons of 20 GeV/c.
- ~ 50% of the beam measuring ²⁴⁴Cm and ²⁴⁶Cm rest dummy measurements and check.





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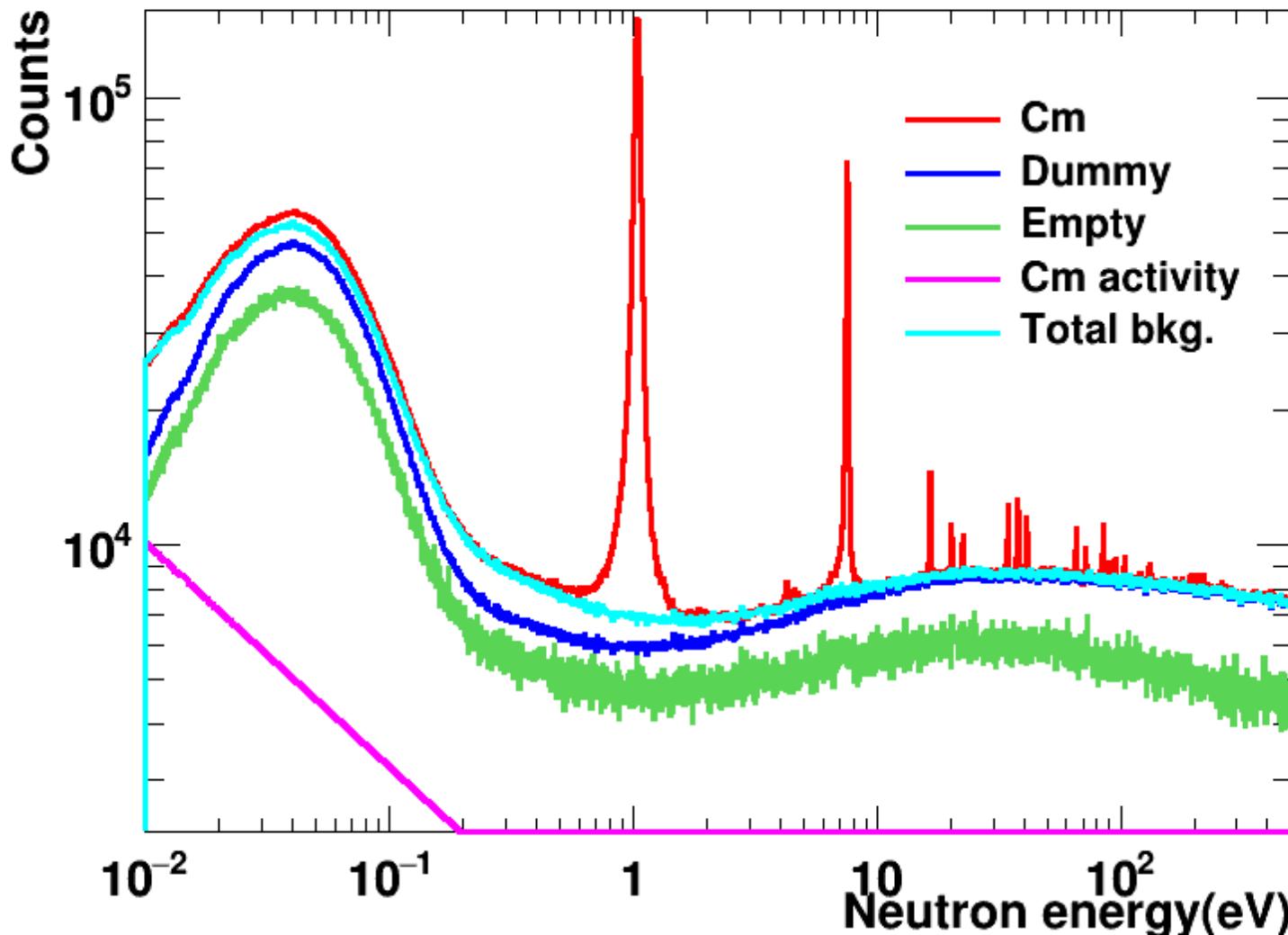
- Analysis
- Precise background subtraction.
- Energy calibration (¹³³Ba, ¹³⁷Cs, ⁶⁰Co, ⁸⁸Y, AmBe and CmC)
- Gain drifts (12 sets)



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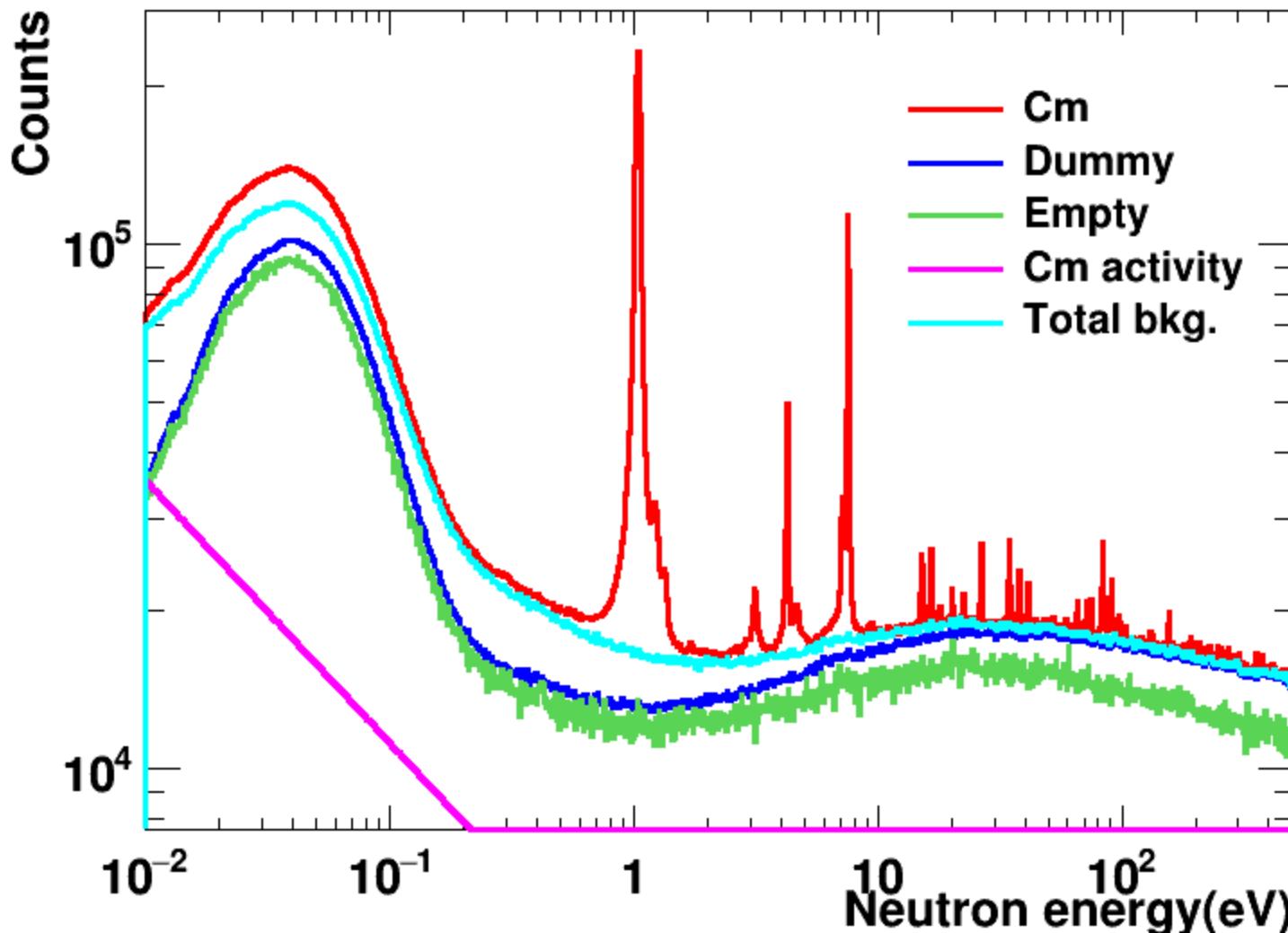
^{244}Cm Experimental results EAR2



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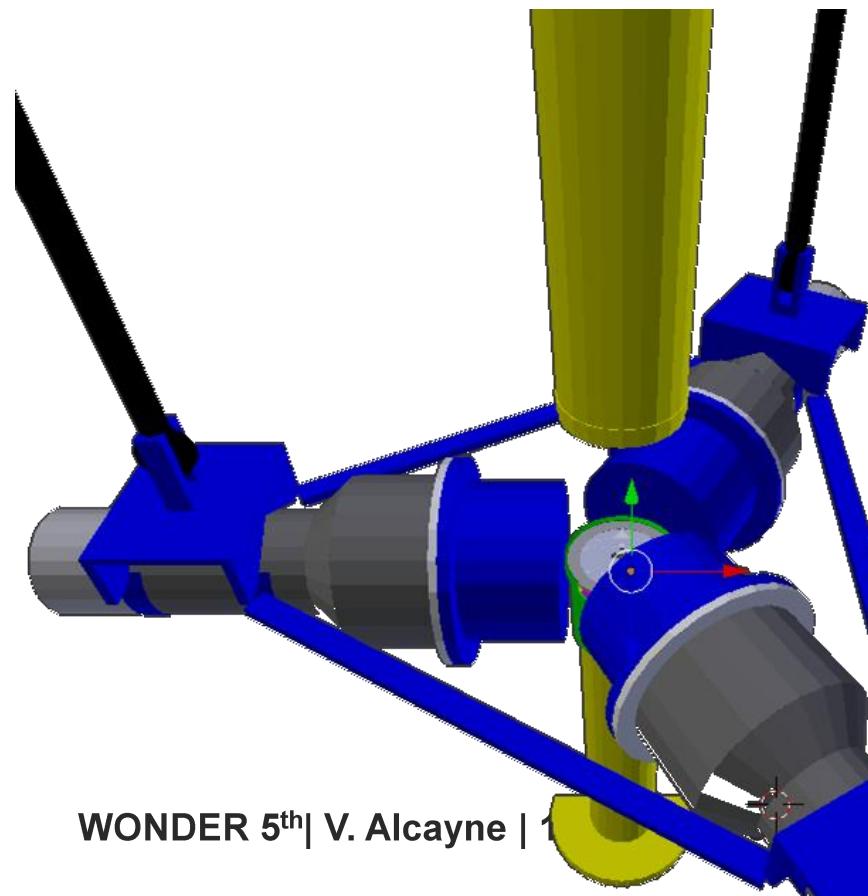
^{246}Cm Experimental results EAR2



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Geant4 Simulations



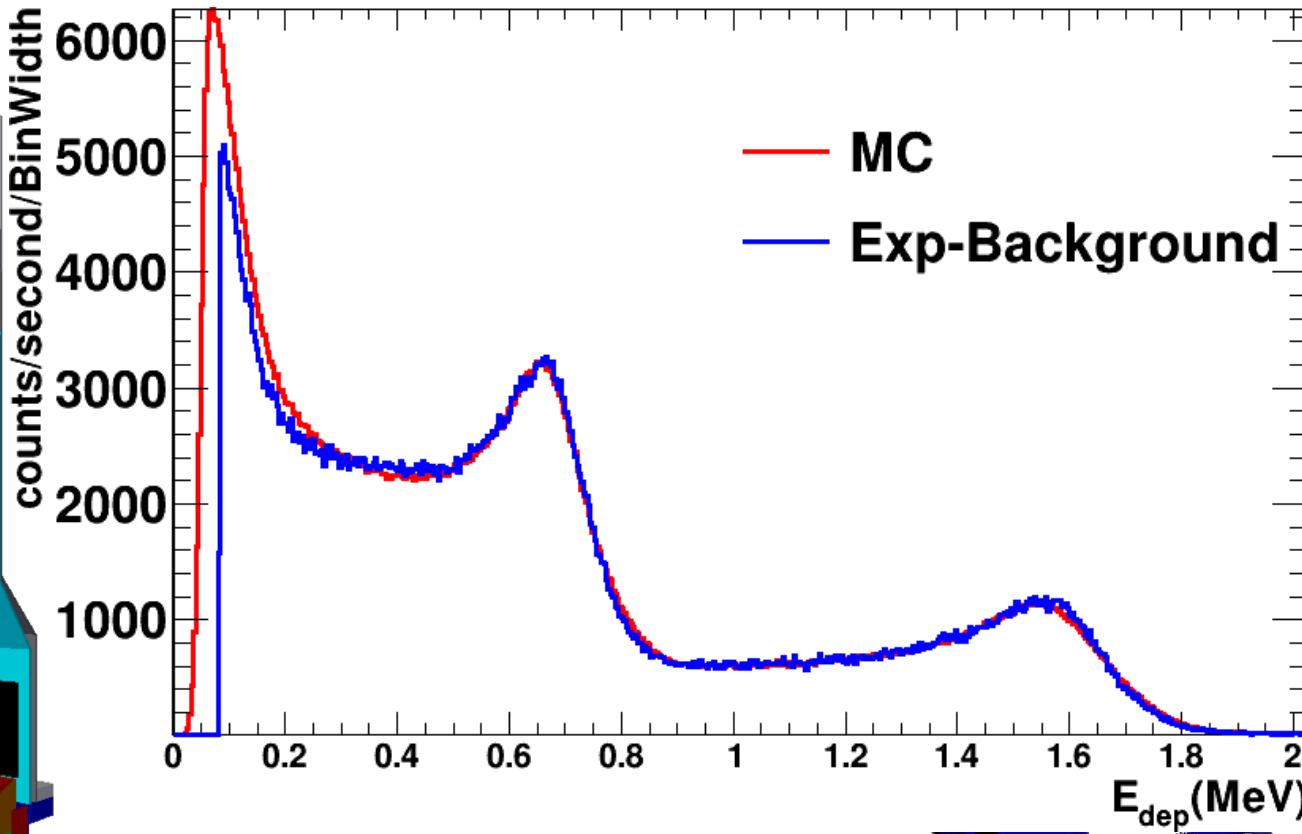
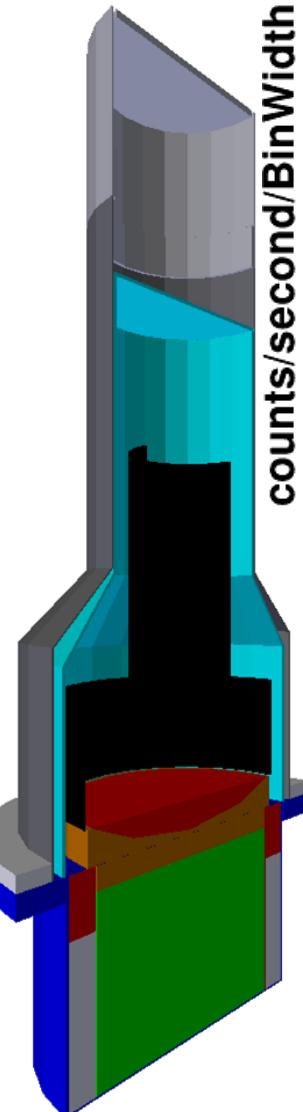
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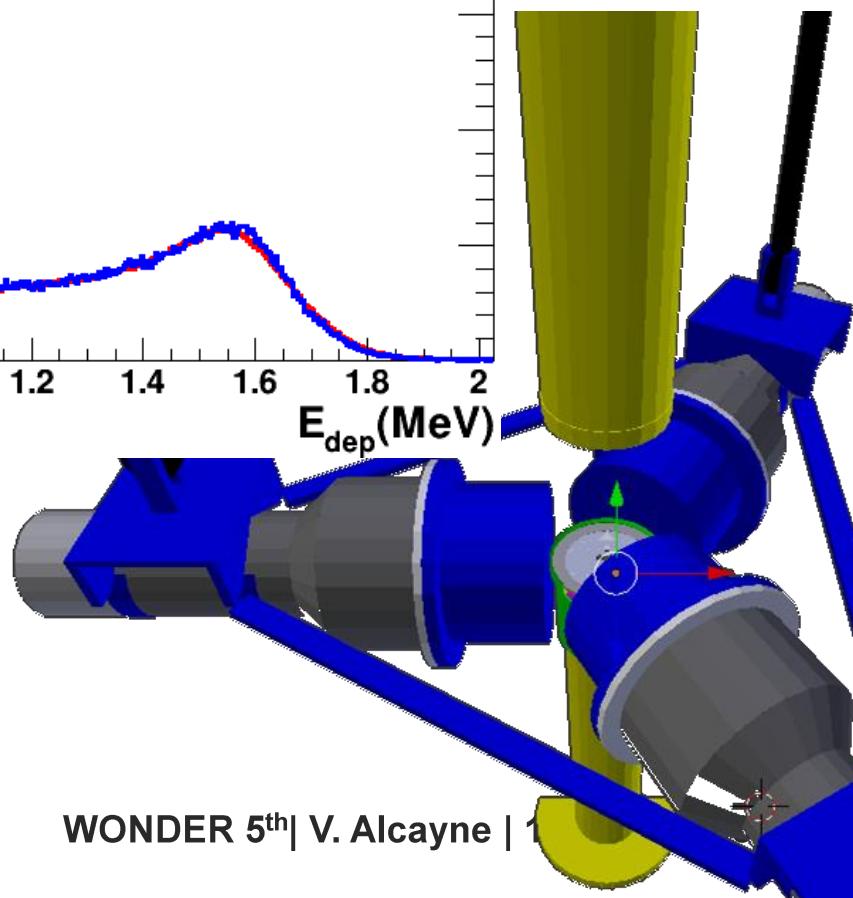
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Geant4 Simulations



Y-88



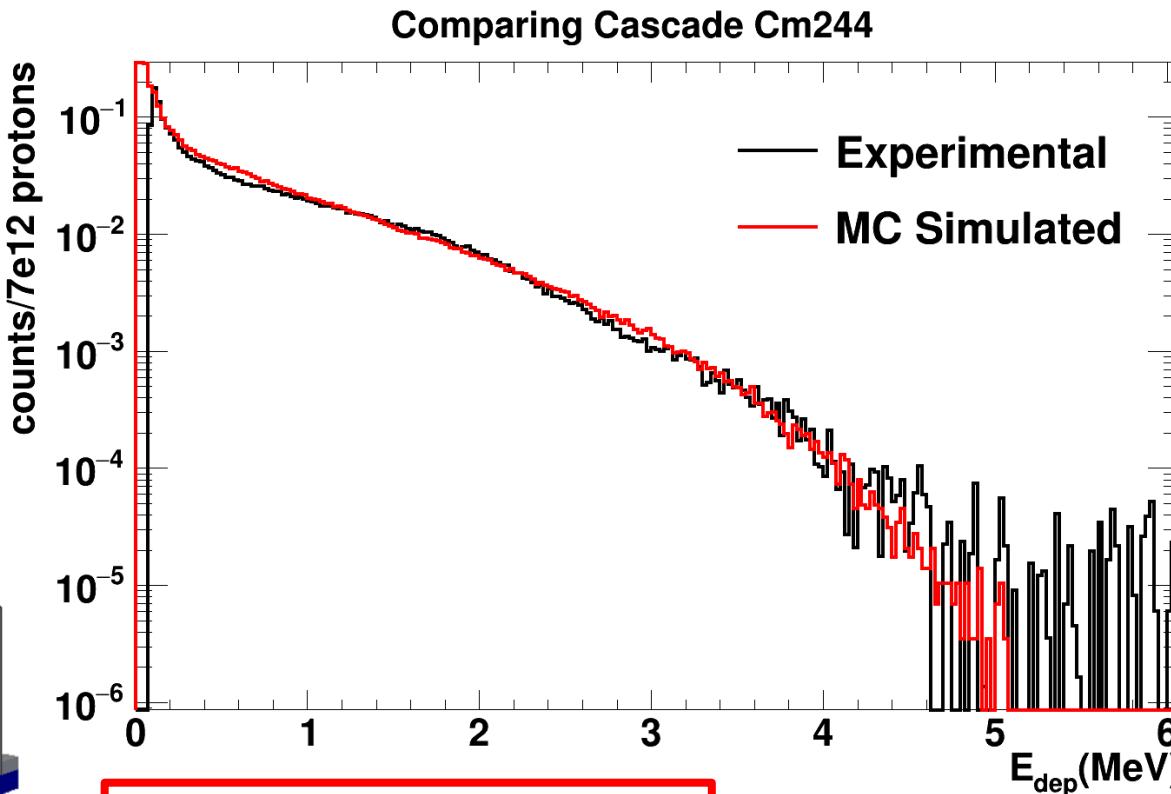
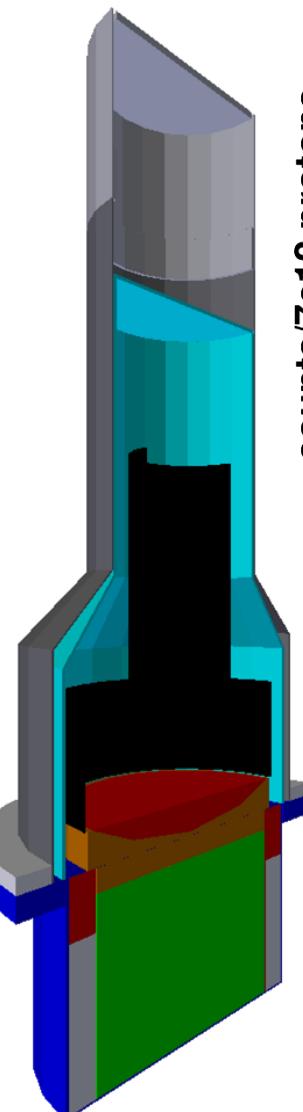
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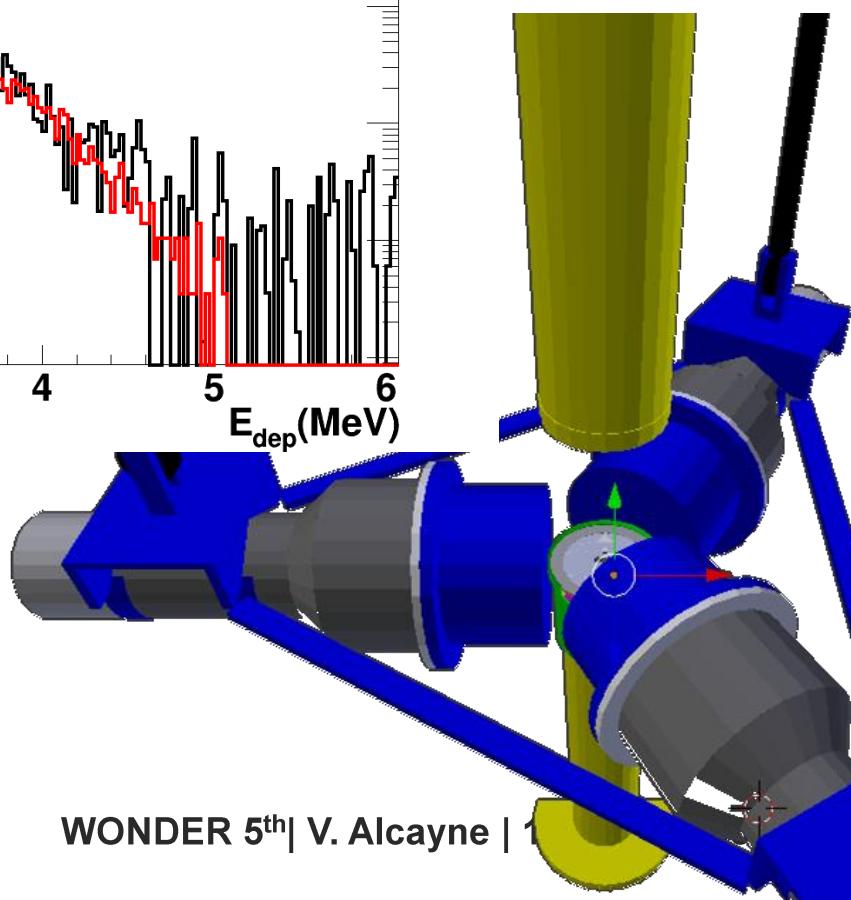
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Geant4 Simulations



Cascade
Simulation for
WF correction



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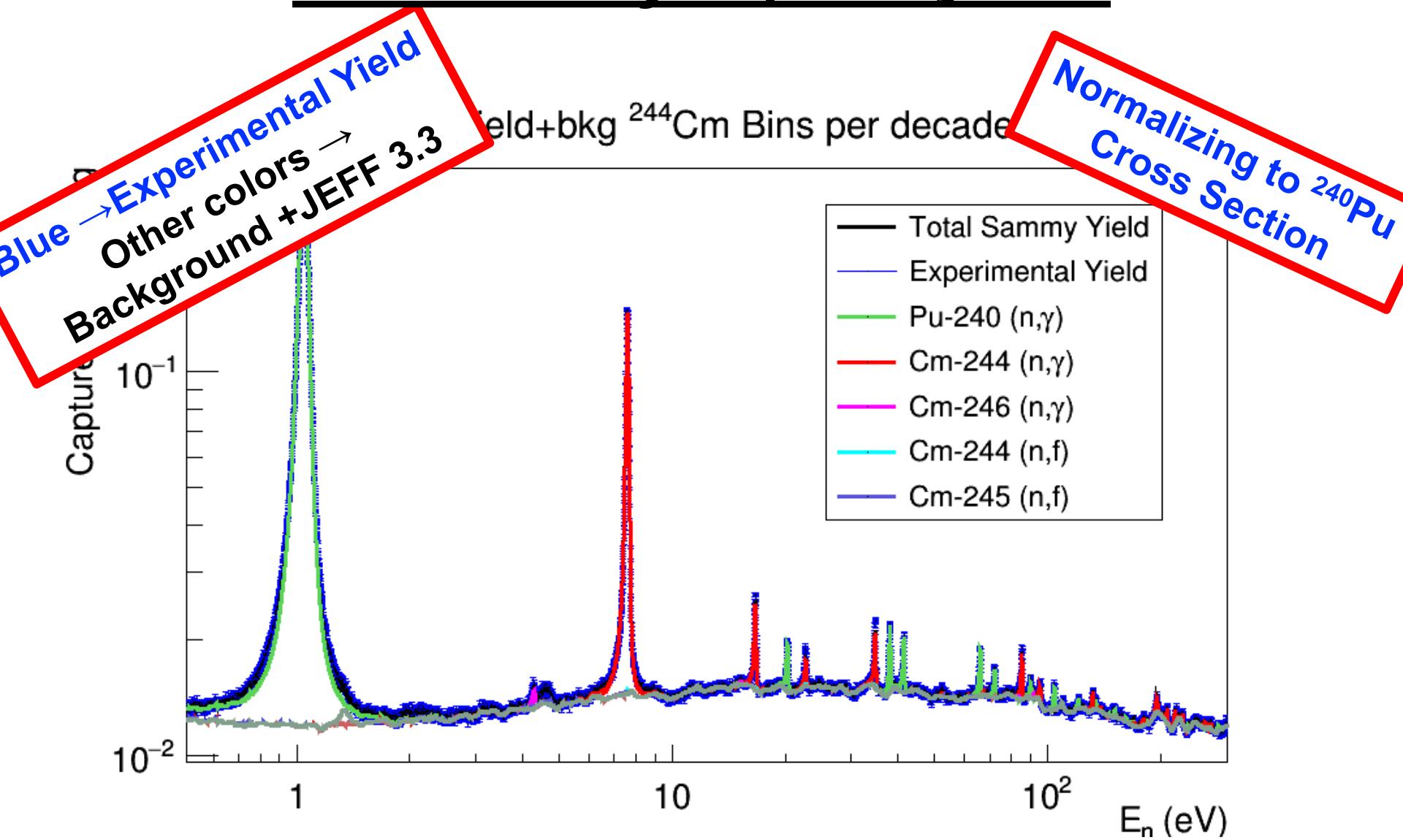


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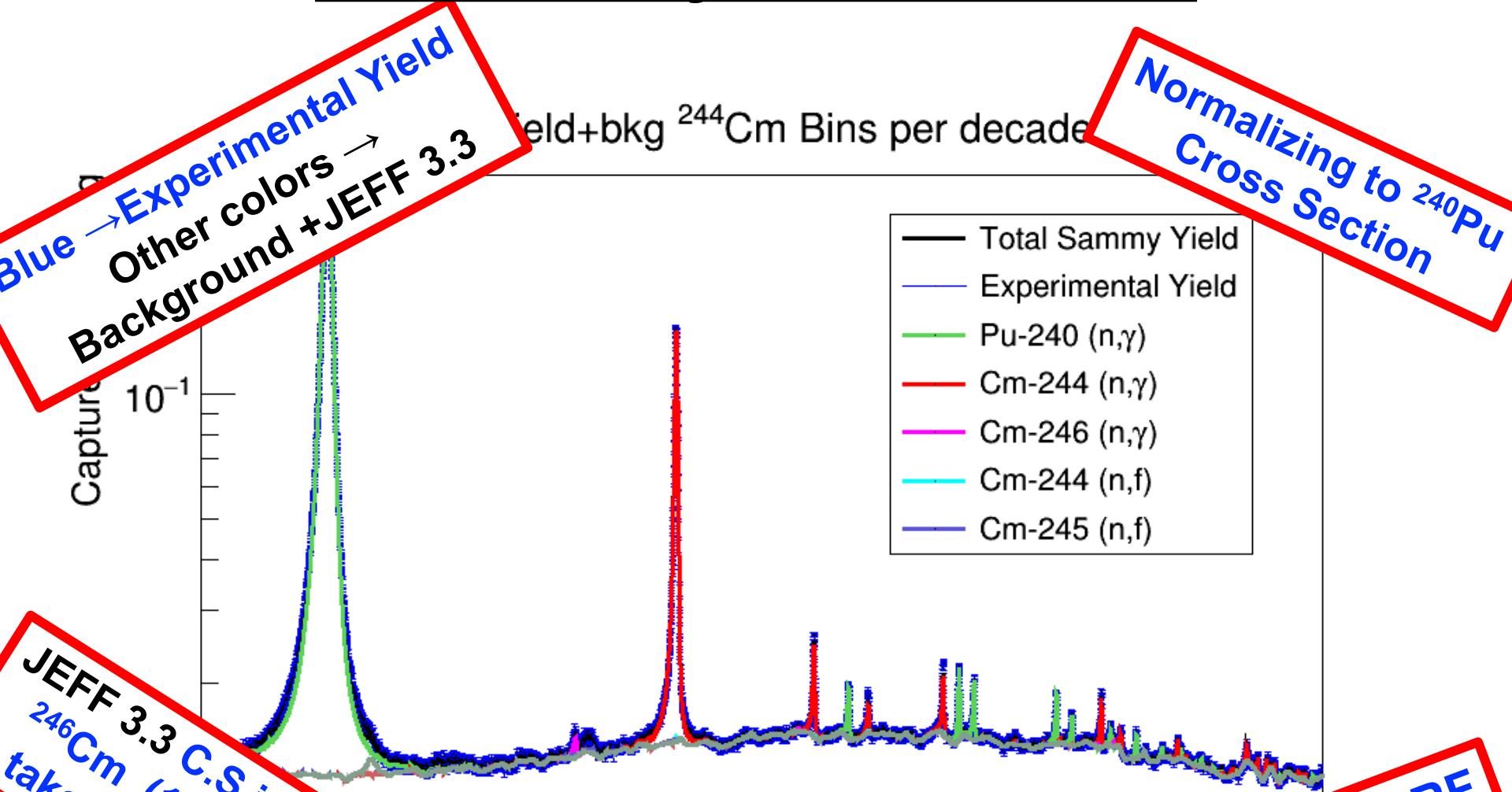
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Preliminary capture yields

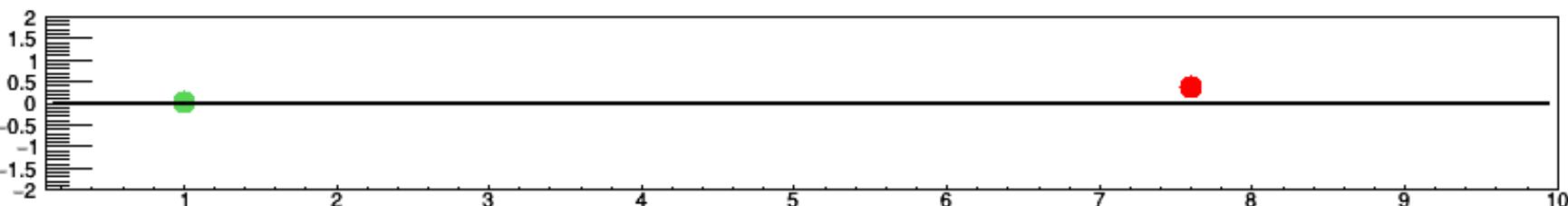
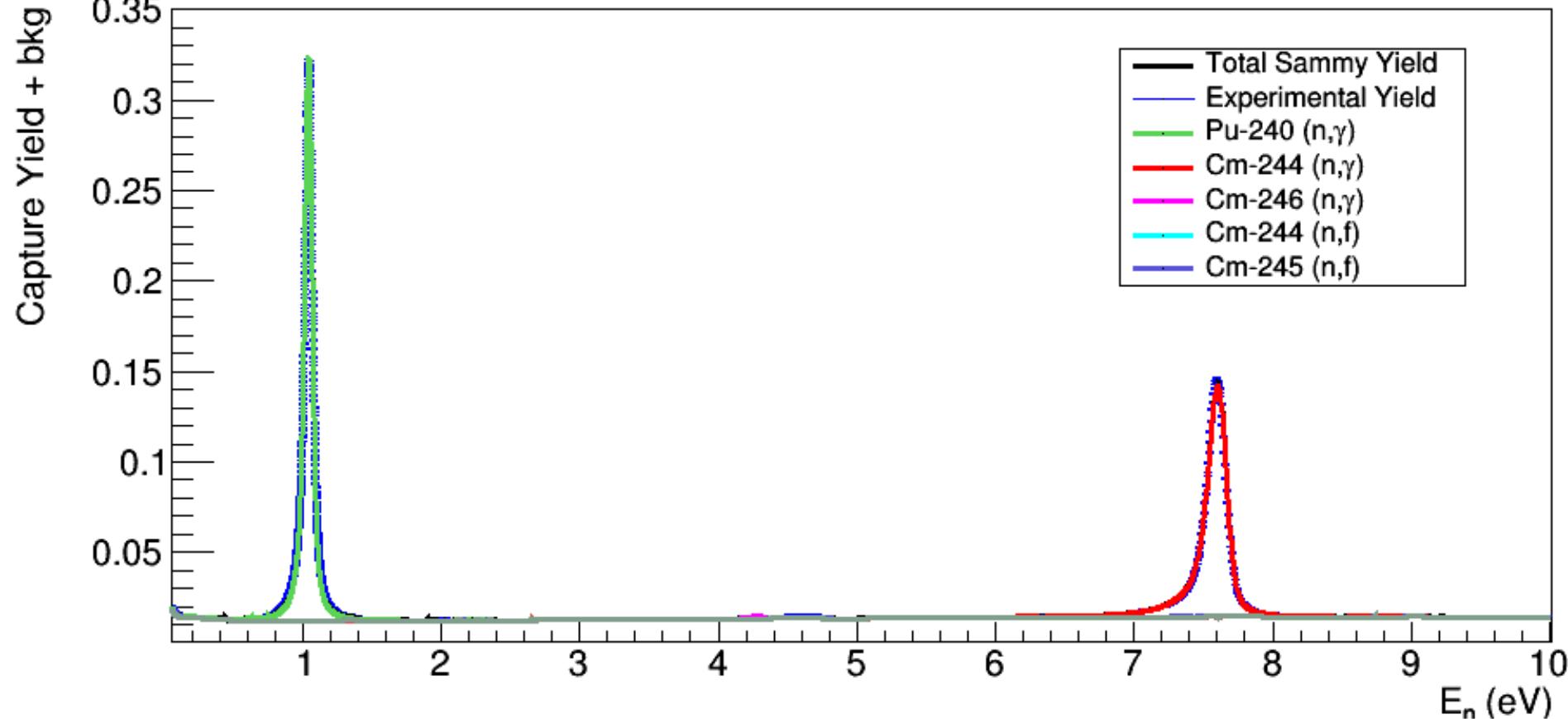


Preliminary capture yields



JEFF 3.3 C.S. in ^{244}Cm and
 ^{246}Cm (1eV to 1keV) is
taken from JENDL 4.0

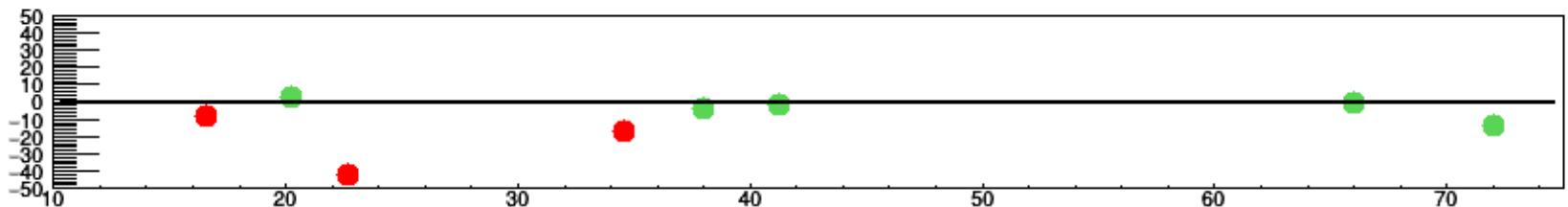
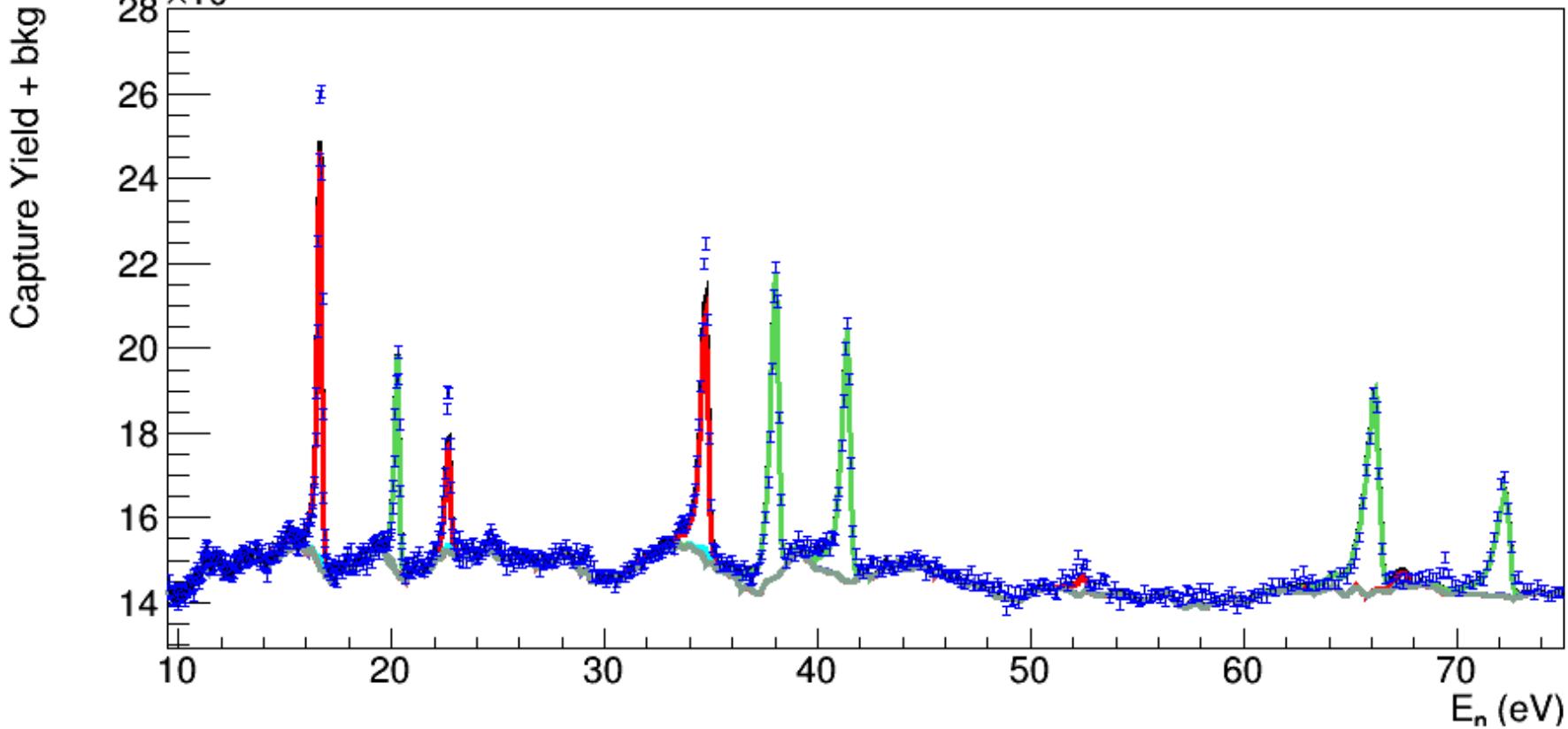
Capture Yield+bkg ^{244}Cm Bins per decade 3000



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Capture Yield+bkg ^{244}Cm Bins per decade 1000

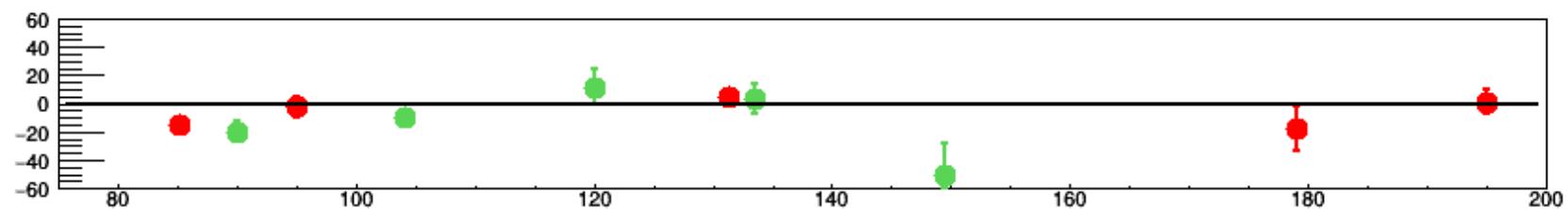
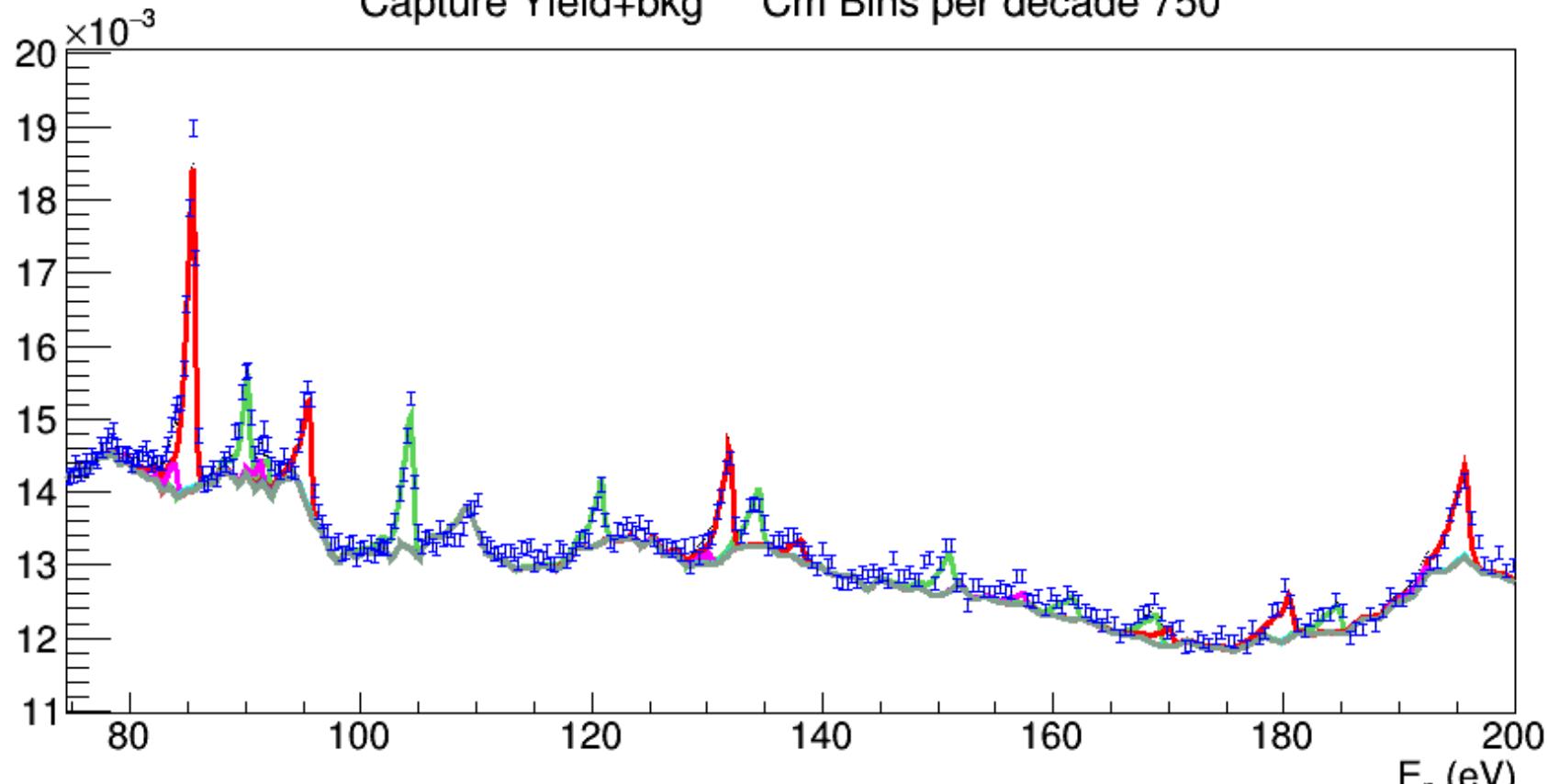


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Capture Yield+bkg ^{244}Cm Bins per decade 750

Capture Yield + bkg

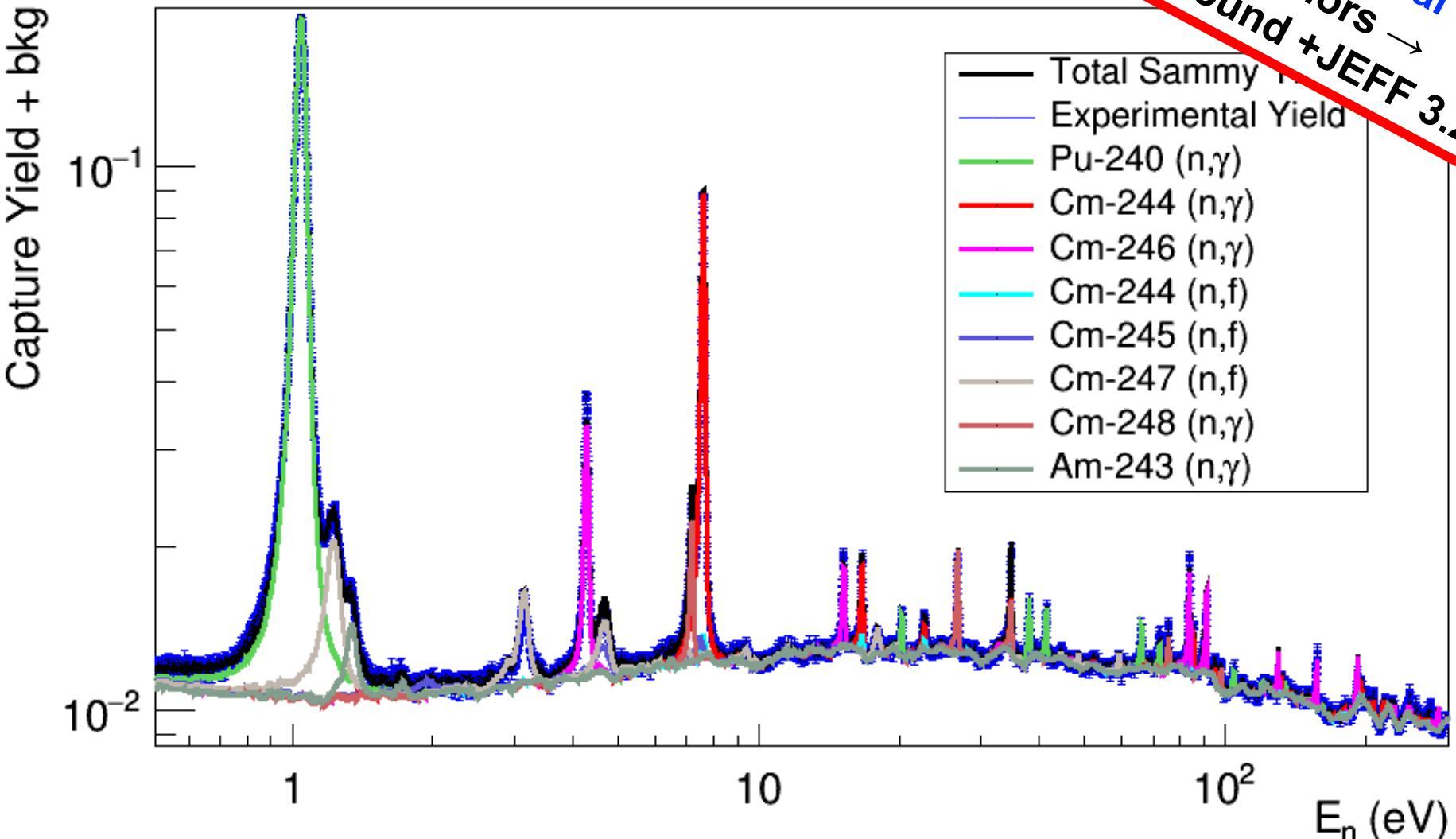


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Capture Yield+bkg ^{246}Cm Bins per day

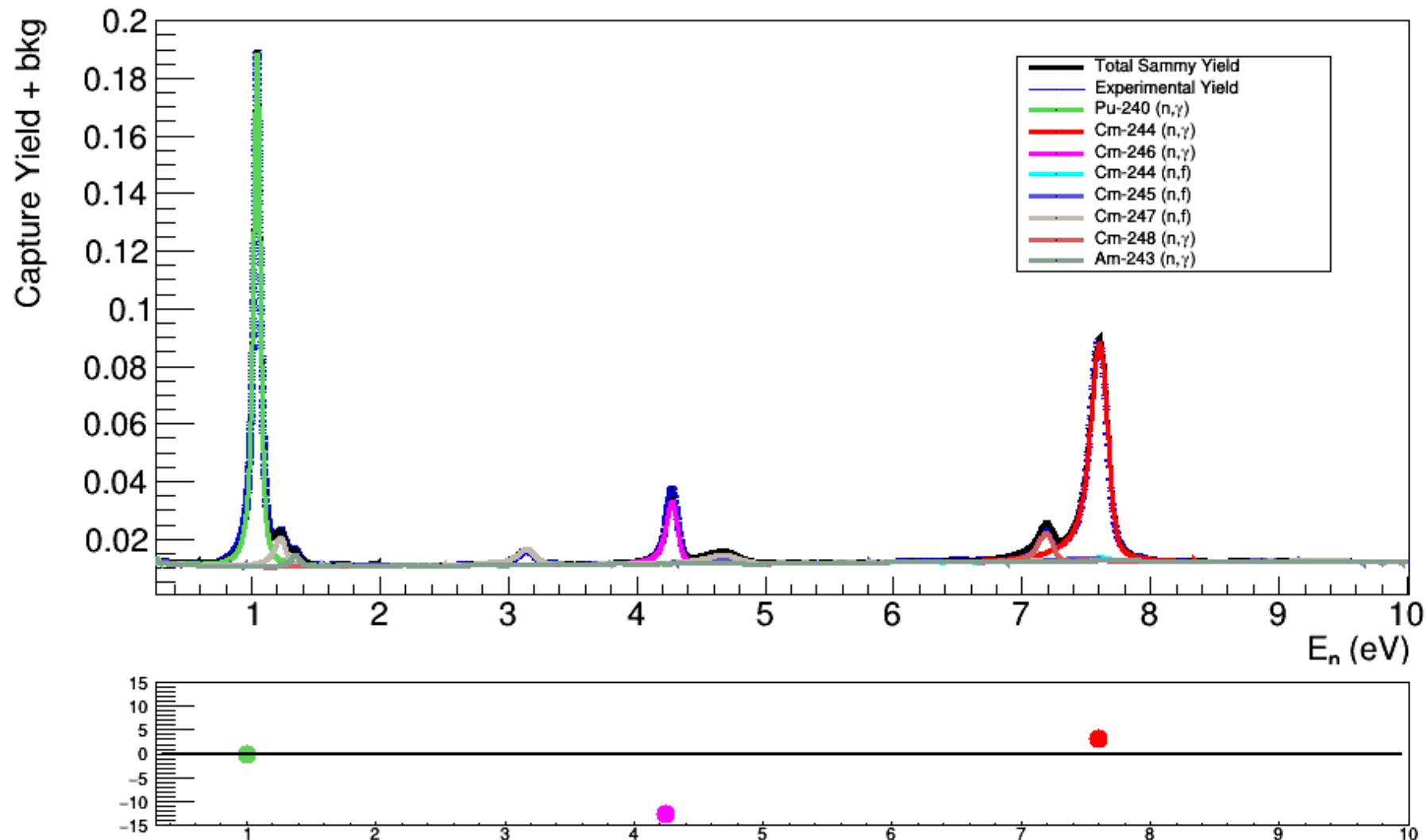
*Blue → Experimental Yield
Other colors →
Background + JEFF 3.2*



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Capture Yield+bkg ^{246}Cm Bins per decade 3000

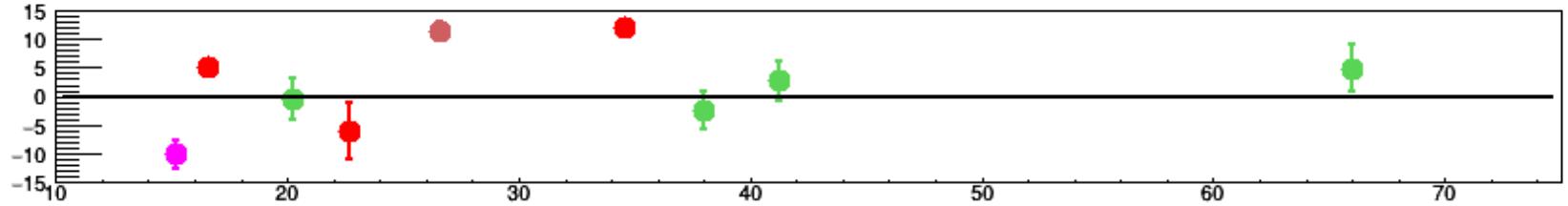
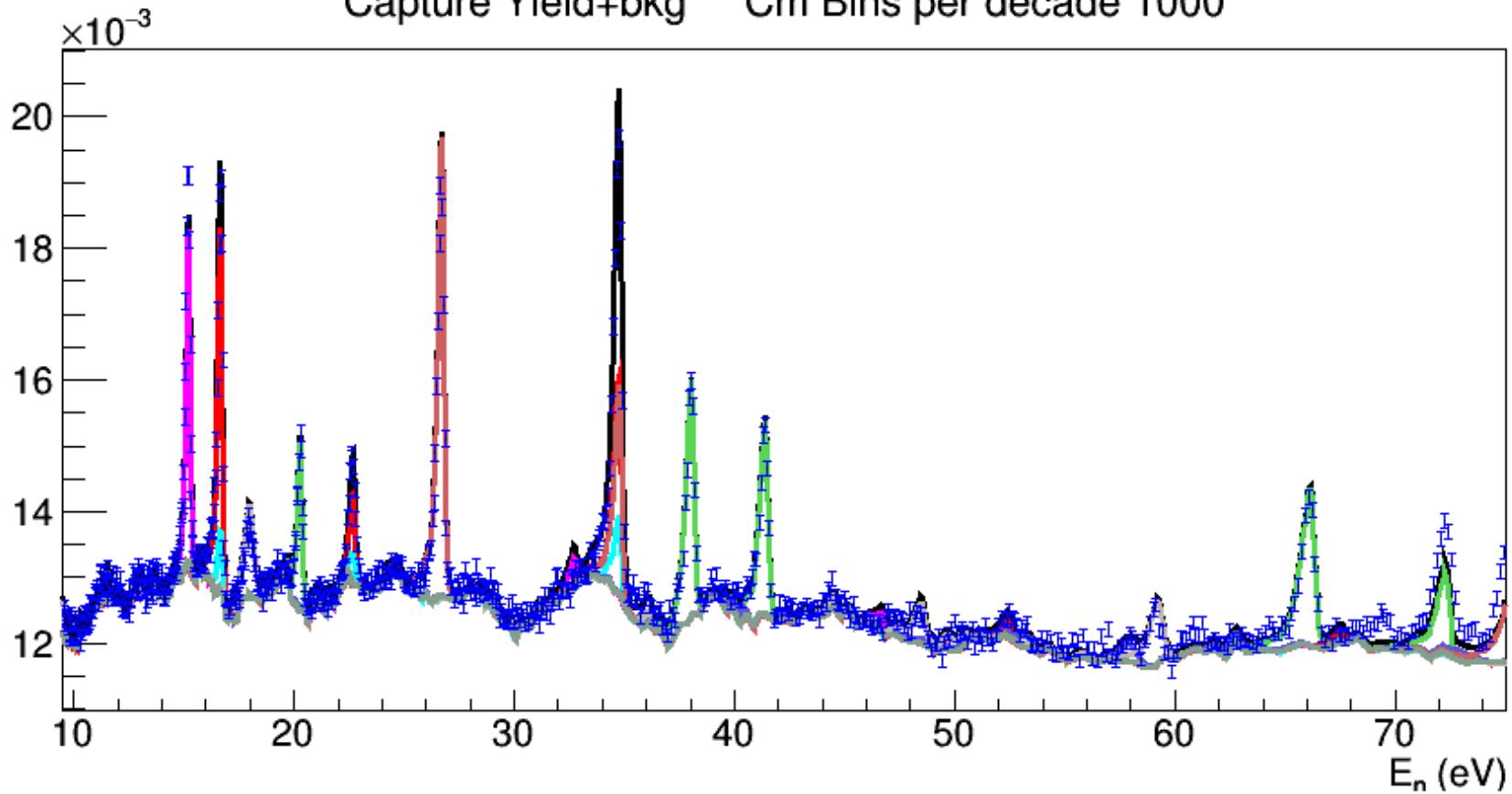


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Capture Yield+bkg ^{246}Cm Bins per decade 1000

Capture Yield + bkg

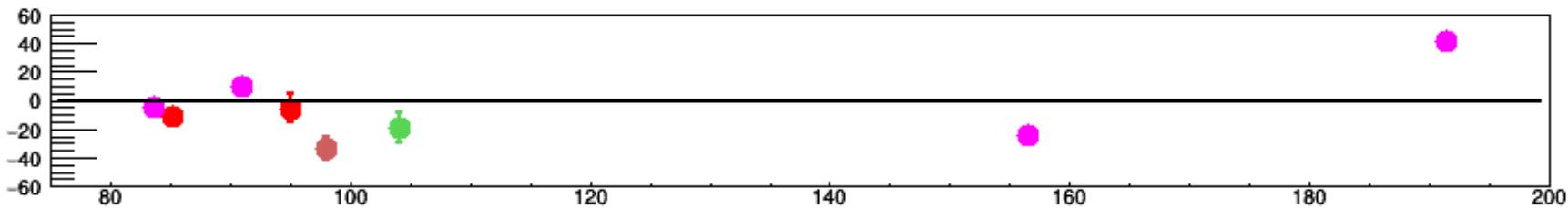
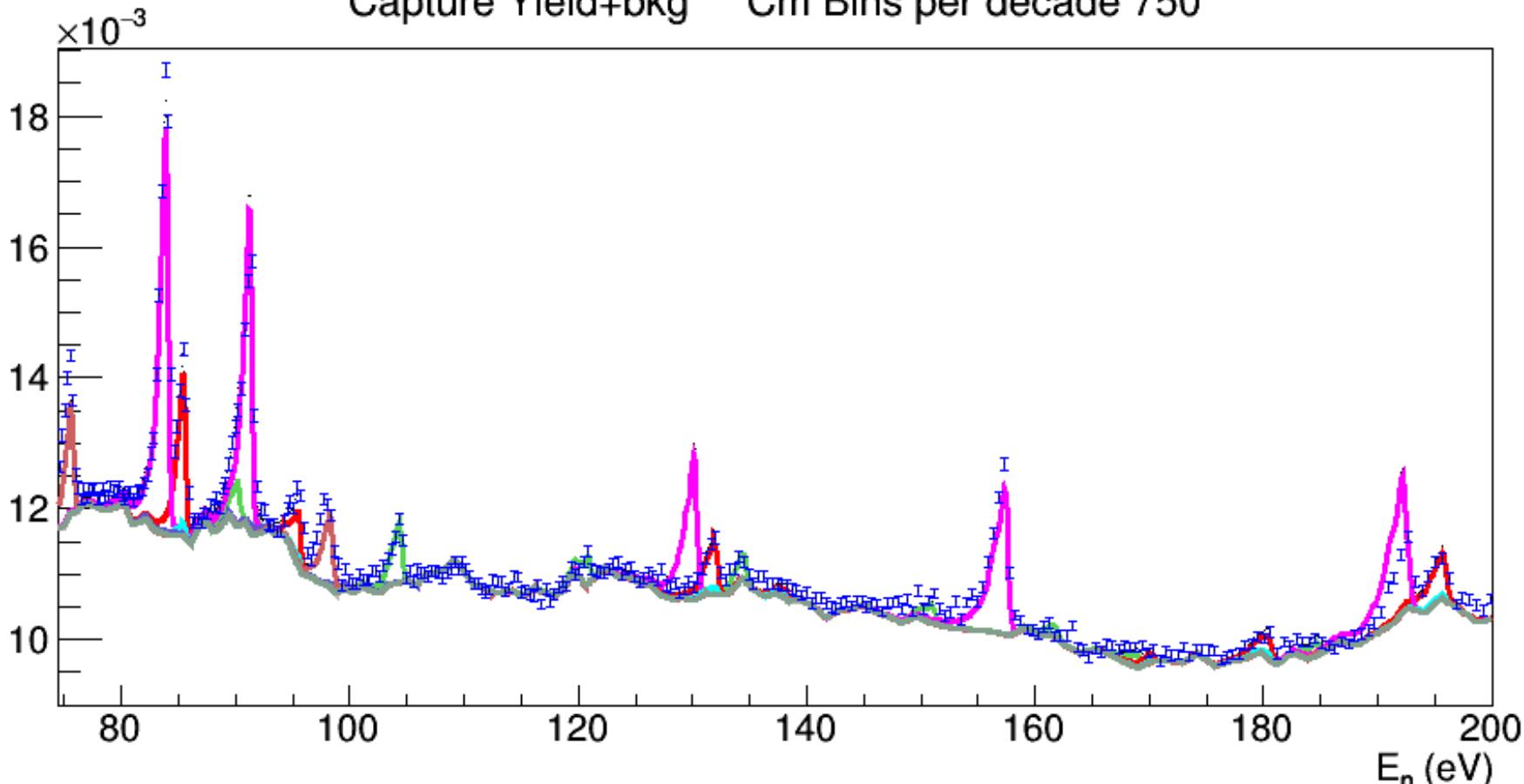


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Capture Yield+bkg ^{246}Cm Bins per decade 750

Capture Yield + bkg



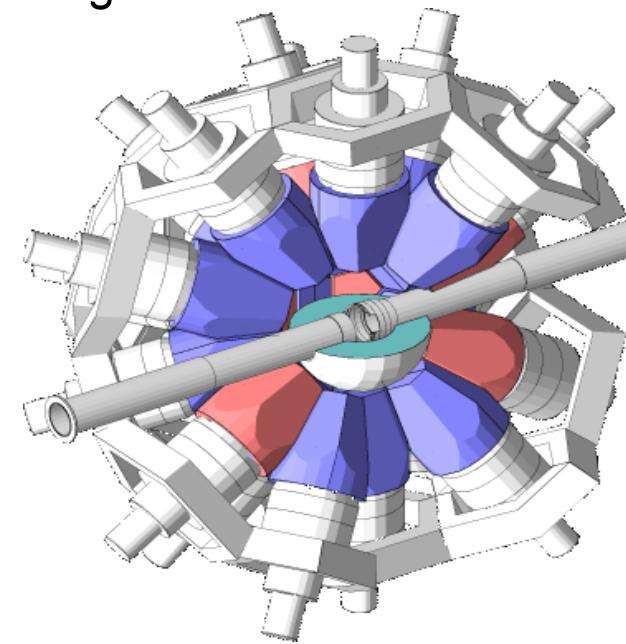
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TAC Setup EAR1 (~185m)

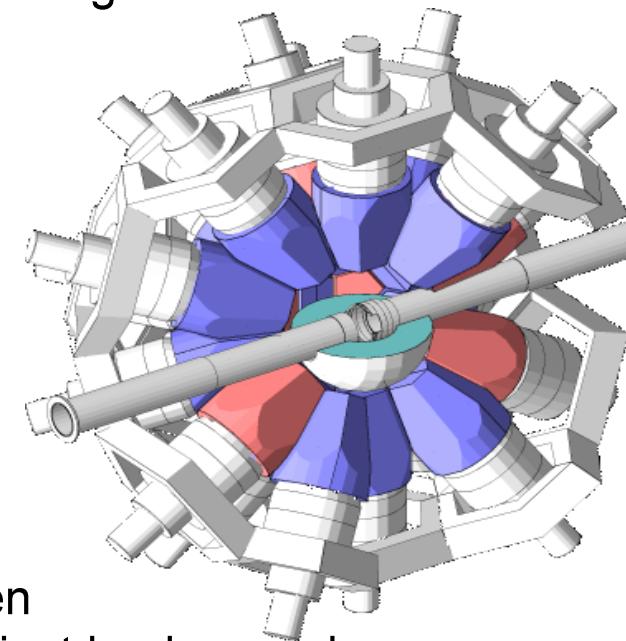
- TAC(Total Absorption Calorimeter).
 - Sphere of 40 BaF₂ crystals, 95% solid angle.
 - Detecting almost all the gammas in the cascade.



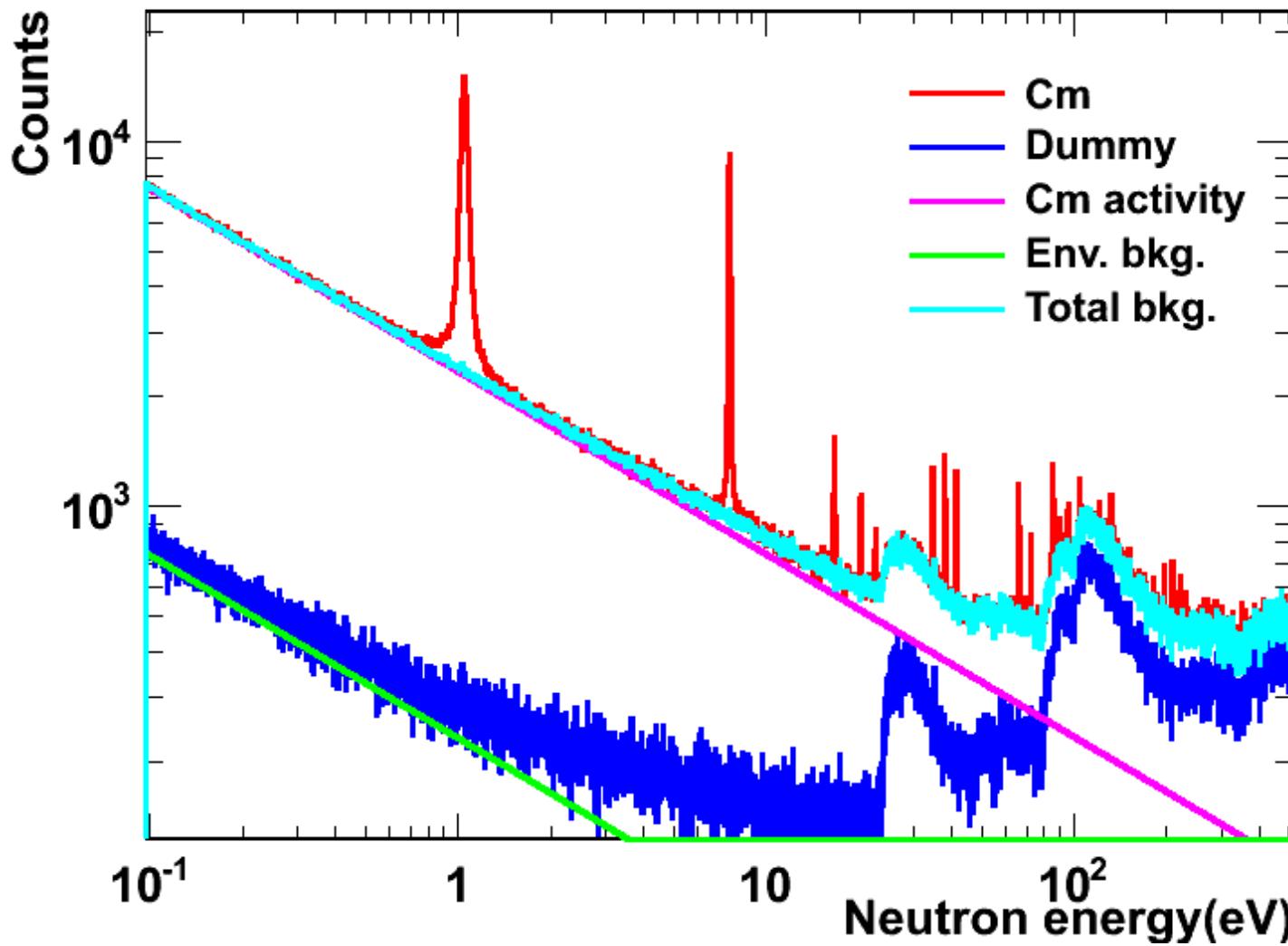


TAC Setup EAR1 (~185m)

- **TAC(Total Absorption Calorimeter).**
 - Sphere of 40 BaF₂ crystals, 95% solid angle.
 - Detecting almost all the gammas in the cascade.
- **Experiment**
 - 2 weeks.
 - $5 \cdot 10^{17}$ protons of 20 GeV/c.
- **Analysis**
 - Coincidence between the 40 detectors. Reject background.
 - Montecarlo simulations with Geant4.



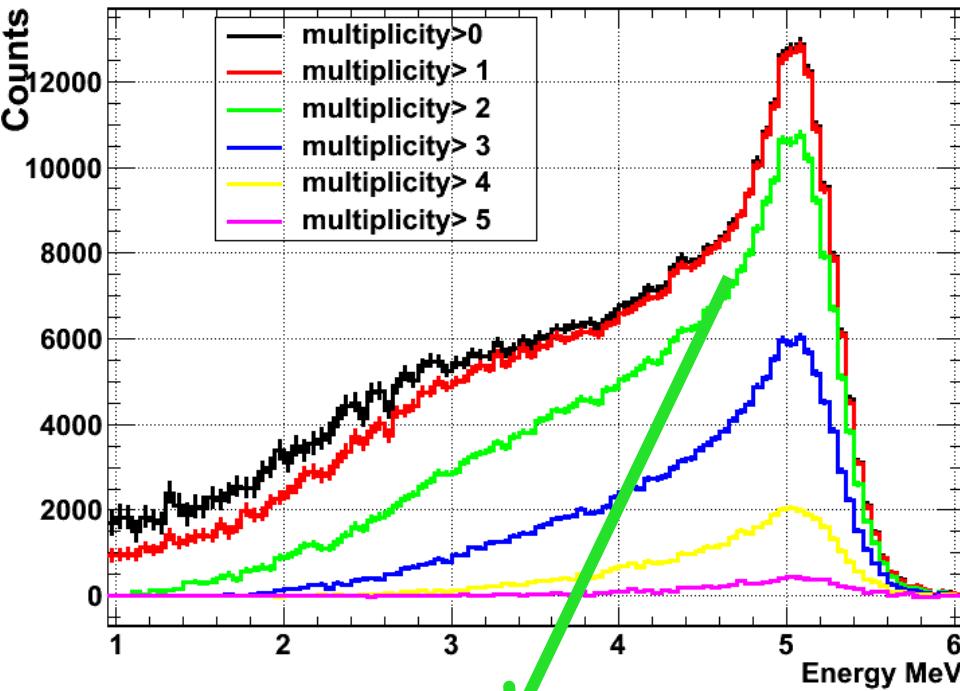
^{244}Cm Experimental **results EAR1 TAC**



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First Resonance Pu²⁴⁰, Neutron Energy 1 to 1.1 eV



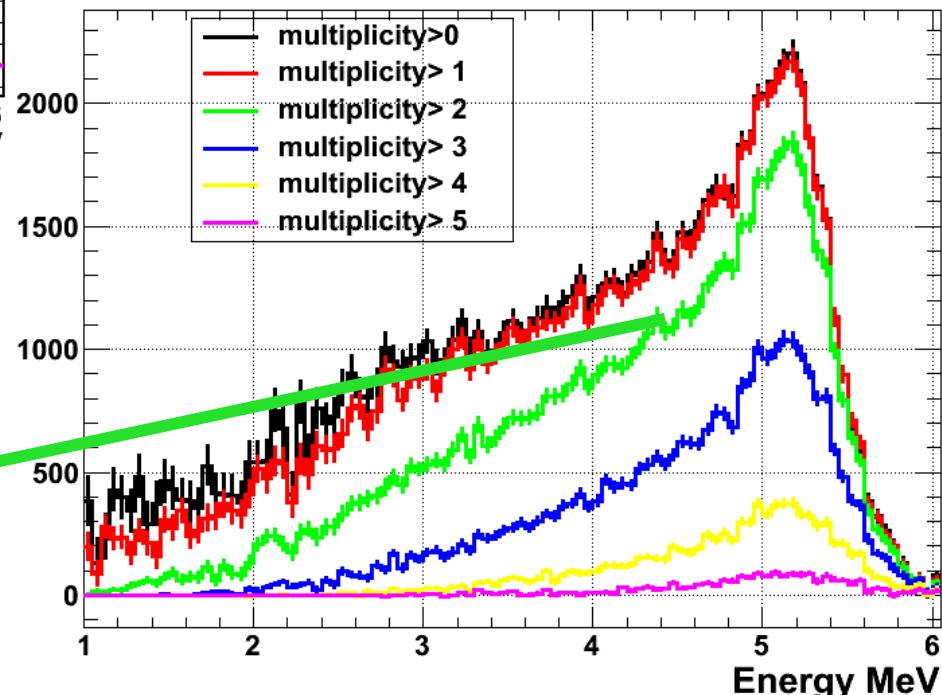
~300000 counts

~60000 counts

EM cascades TAC

- Background subtracted
- Different multiplicities
- Normalization

First Resonance Cm²⁴⁴, Neutron Energy 7.5 to 7.7 eV



Summary and conclusions

- Only 2 previous difficult measurements of ^{244}Cm and ^{246}Cm .
- ~1 mg samples with high activity (~1 GBq) provided by JAEA.
- Radiative capture cross section of ^{244}Cm and ^{246}Cm were successfully measured at n_TOF in both areas with complementary set ups:
 - EAR2 with C_6D_6 (TED).
 - EAR1 with TAC.
- Preliminary results are promising and we expect to reach the proposed goals with a precise analysis.

BACKUP SLIDES



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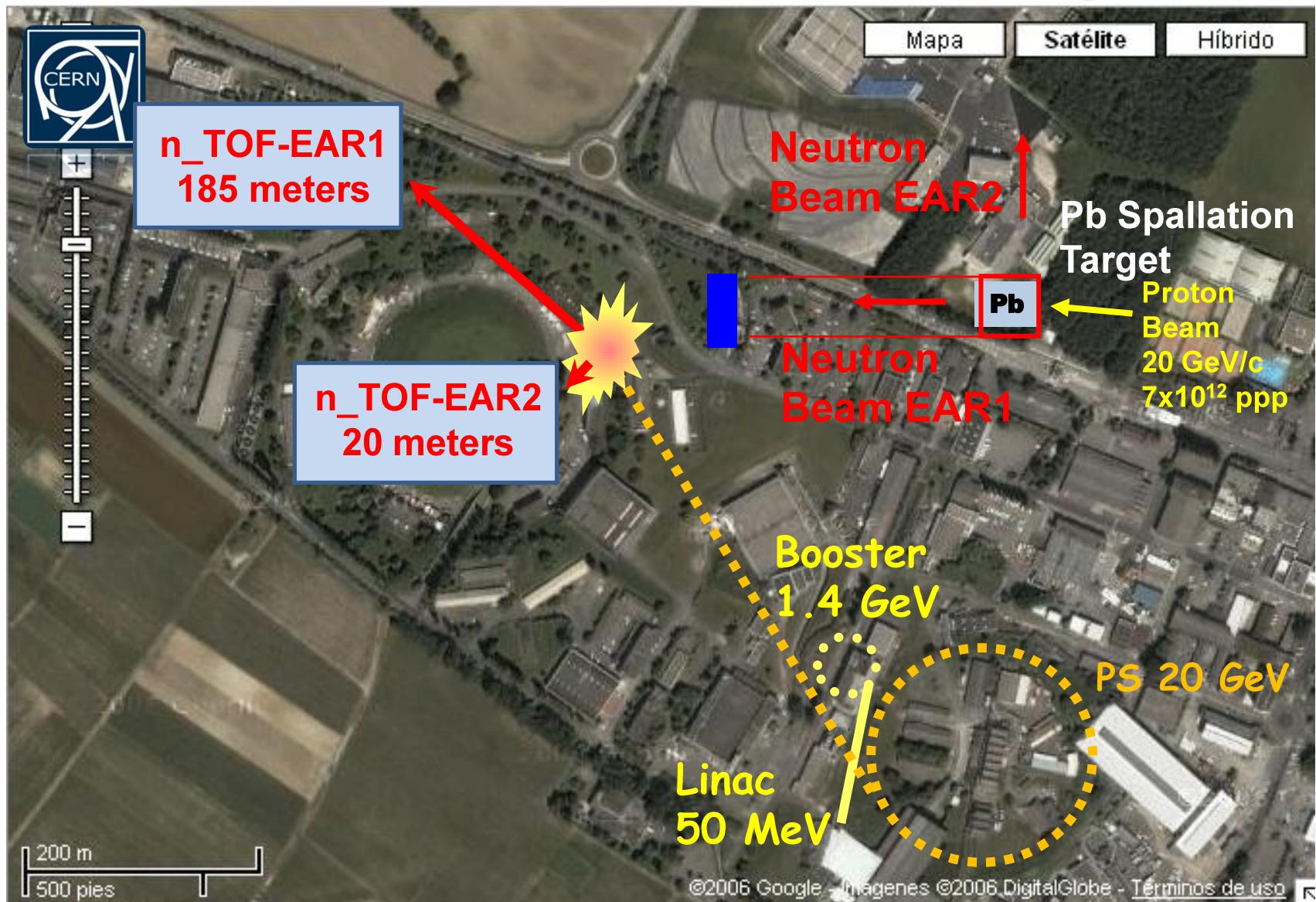
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Motivation

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 (6/2+) EC, α	Cf246 35.7 h 0+ EC, α ,sf,...	Cf247 3.11 h (7/2+) EC, α	Cf248 333.5 d 0+ EC, α ,sf	Cf249 351 y 9/2- EC, α ,sf	Cf250 13.08 y 0+ EC, 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,-	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+ EC, 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+ EC, α ,sf	Cm245 8500 y EC, α ,sf	Cm246 4730 y EC, α ,sf	Cm247 1.56E+7 y EC, α ,sf	Cm248 3.4E+5 y EC, 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 0+ EC, α ,sf	Am244 0.1 h 0- EC, β -	Am245 2.05 h (5/2+) EC, β -	Am246 39 m (7-) EC, β -	Am247 23.0 m (5/2) EC, β -
Pu237 45.2 d 7/2- EC, α	Pu238 87.7 y 0+ EC, α ,sf	Pu239 24110 y 1/ α ,sf	Pu240 6563 y 1/ α ,sf	Pu241 14.35 y EC, β , α ,sf,...	Pu242 3.733E+5 y EC, β , α ,sf,...	Pu243 4.916 h EC, β , α ,sf,...	Pu244 8.08E+7 y 0+ EC, β , α ,sf,...	Pu245 10.5 h (9/2-) EC, β -	Pu246 10.84 d 0+ EC, β -
Np236 1.54E5 y (6-)* EC, β , α ,sf,...	Np237 2.144E+6 y 5/2+ α ,sf	Np238 2.117 d 2+ β -	Np239 2.155 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 2.29 m (7-) β -	
U235 7.038E+8 y 7/2- α , ²⁰ Ne,sf,... 0.7200	U236 2.342E7 y 0+ EC, α ,sf	U237 6.75 d 1/2+ β -	U238 4.468E+9 y 0+ EC, β , α ,sf,... 99.2745	U239 3.35 m 0+ β -	U240 14.1 h 0+ β -	U241 EC, β -	U242 16.8 m 0+ β -		



The n_TOF Facility at CERN: a Google™ view



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Courtesy of J.Lerendegui U.S.

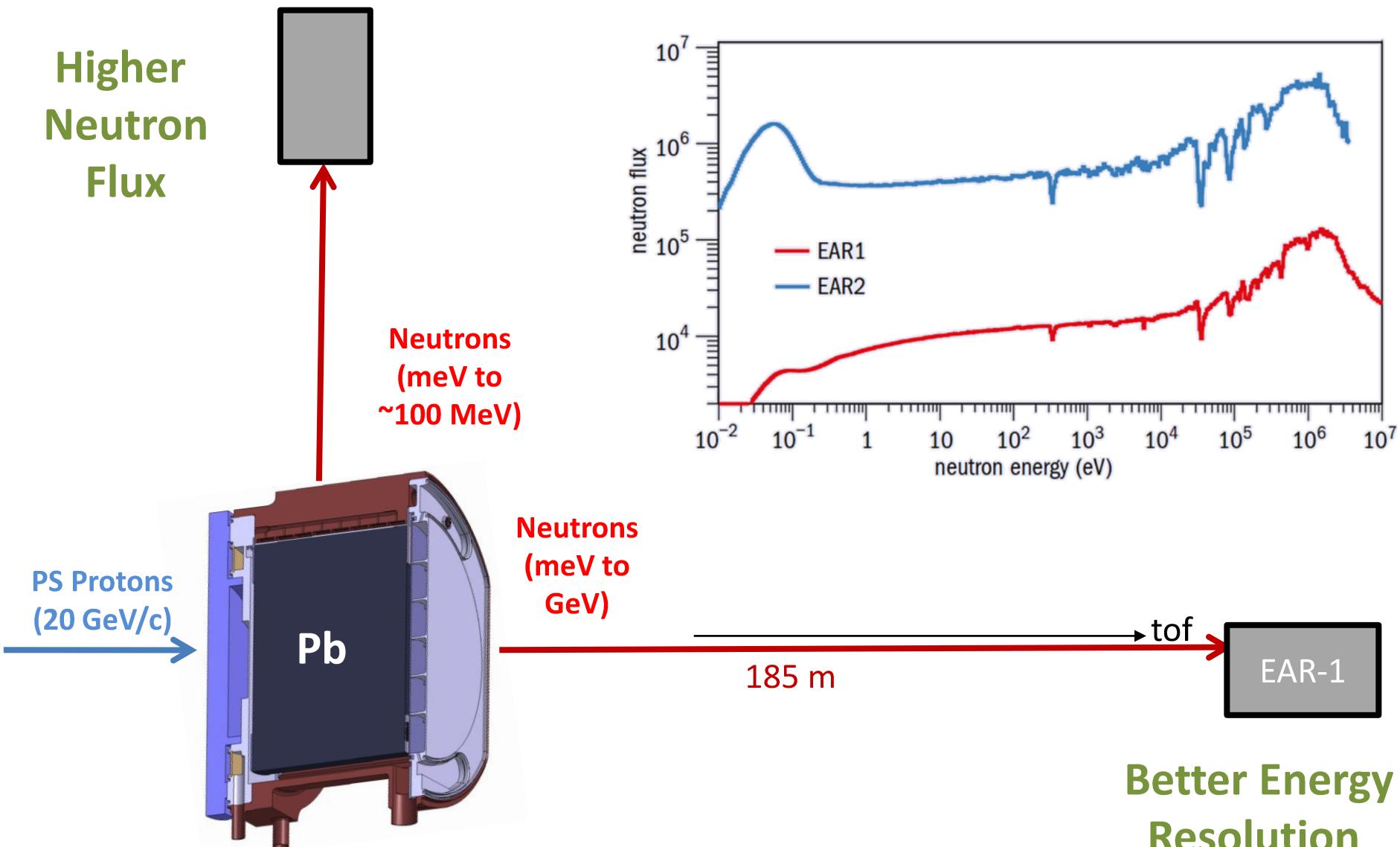


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The n_TOF lead spallation target



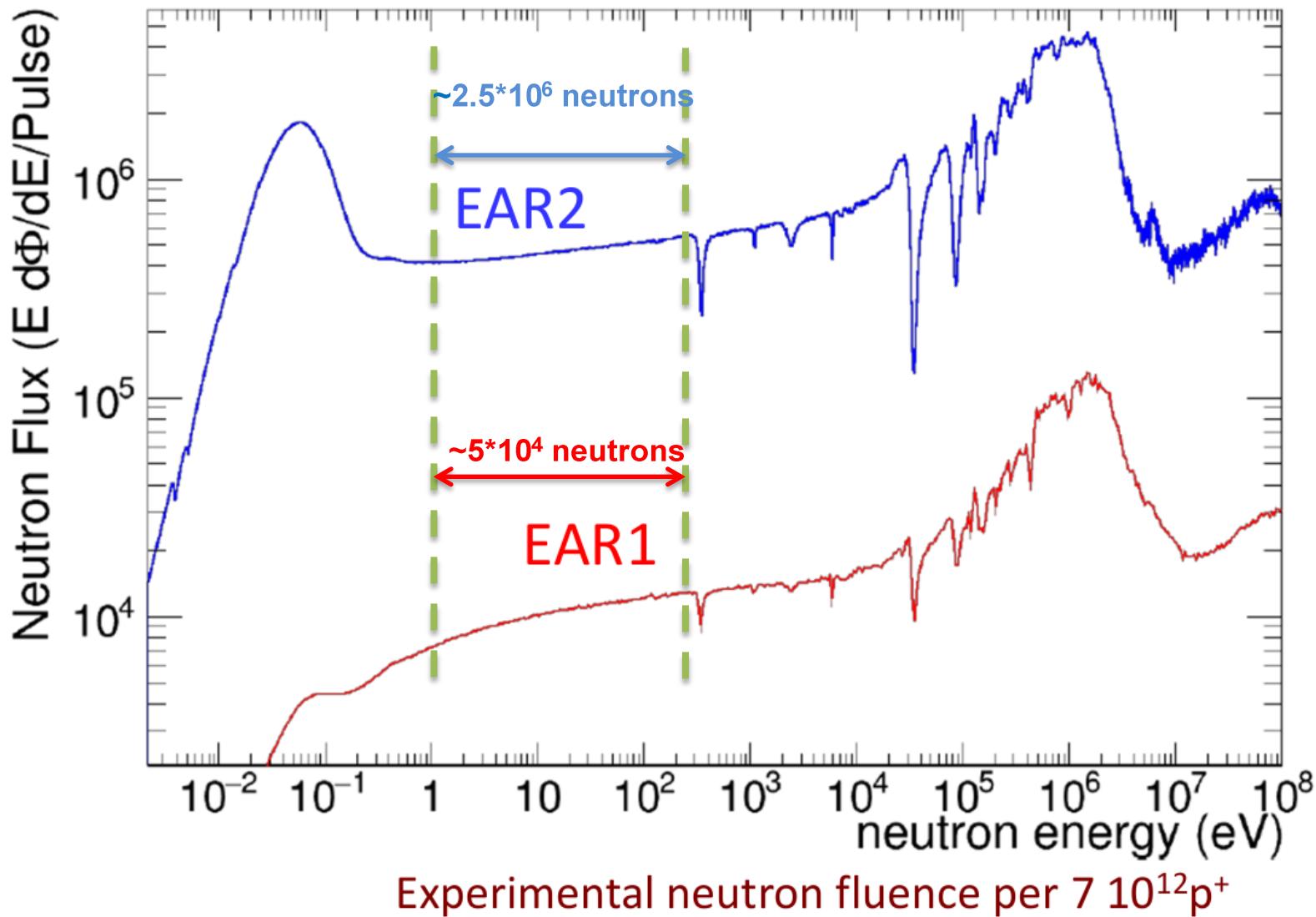
Better Energy
Resolution



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Neutrons per Pulse at n TOF



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Neutron Capture measurement techniques

$$E_c = S_n + E_n$$

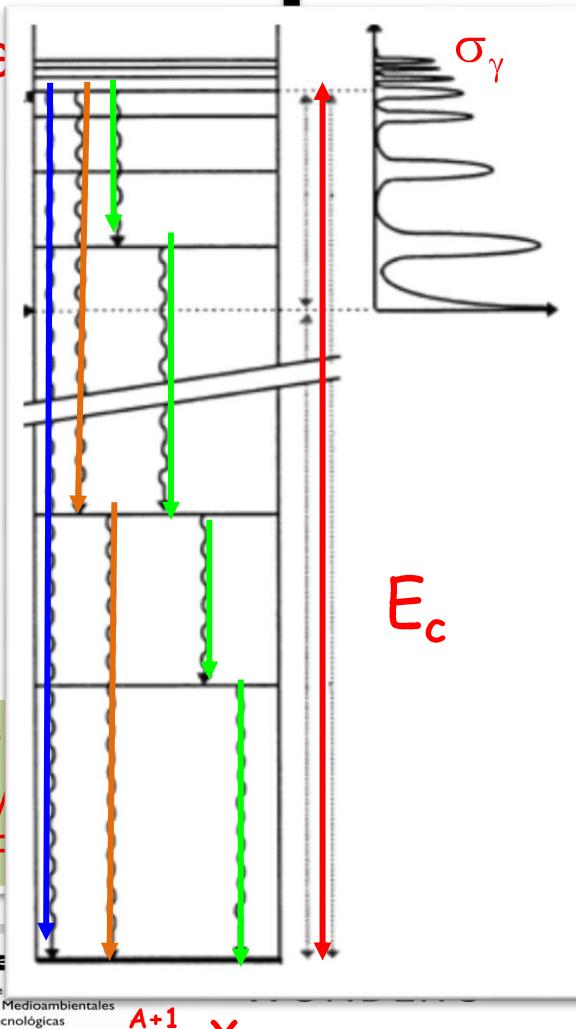
TED
(Total Energy Detector)

Total efficiency depends on E_c and not on the decay path

$$\sum_{i=1}^n \epsilon_{\gamma i}$$

$$\sum_{i=1}^n \gamma_i$$

The proportionality between efficiency and γ -ray energy obtained by software(WF)

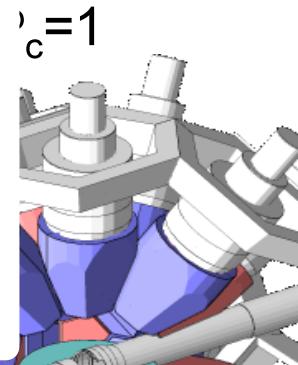


TAC
(Absorption Calorimeter)

basic and solid angle efficiencies are large
 $e : 4\pi$ Detector

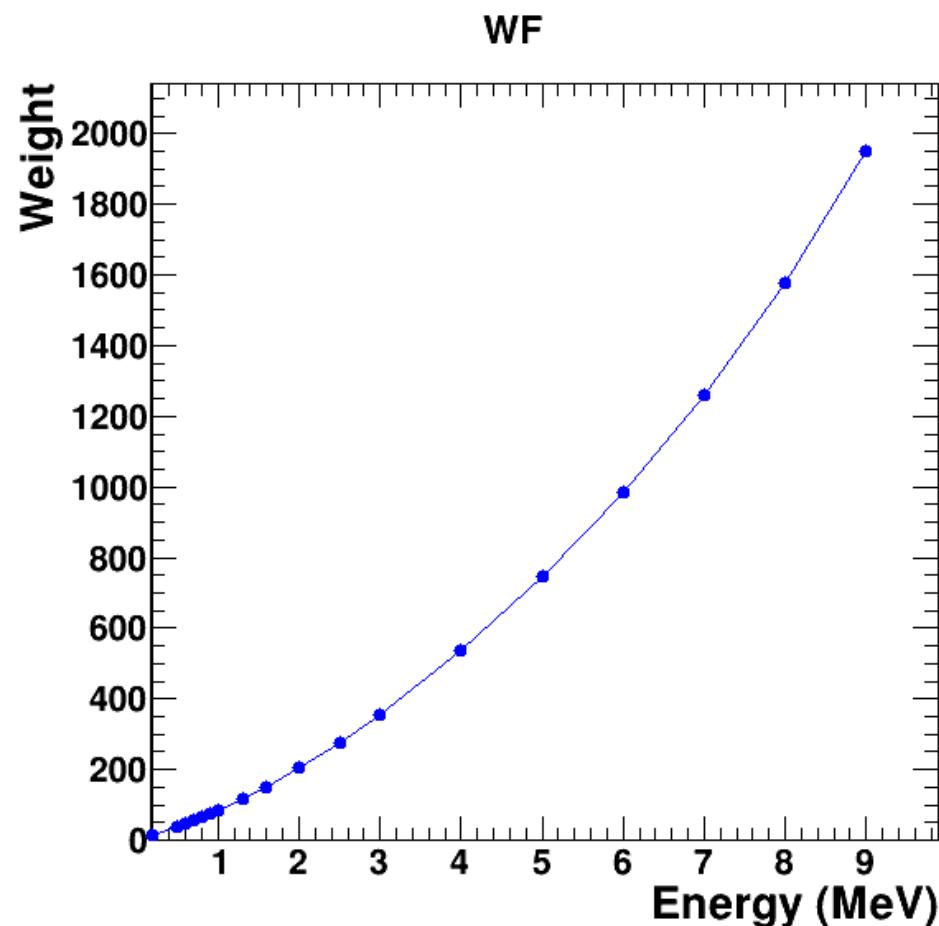
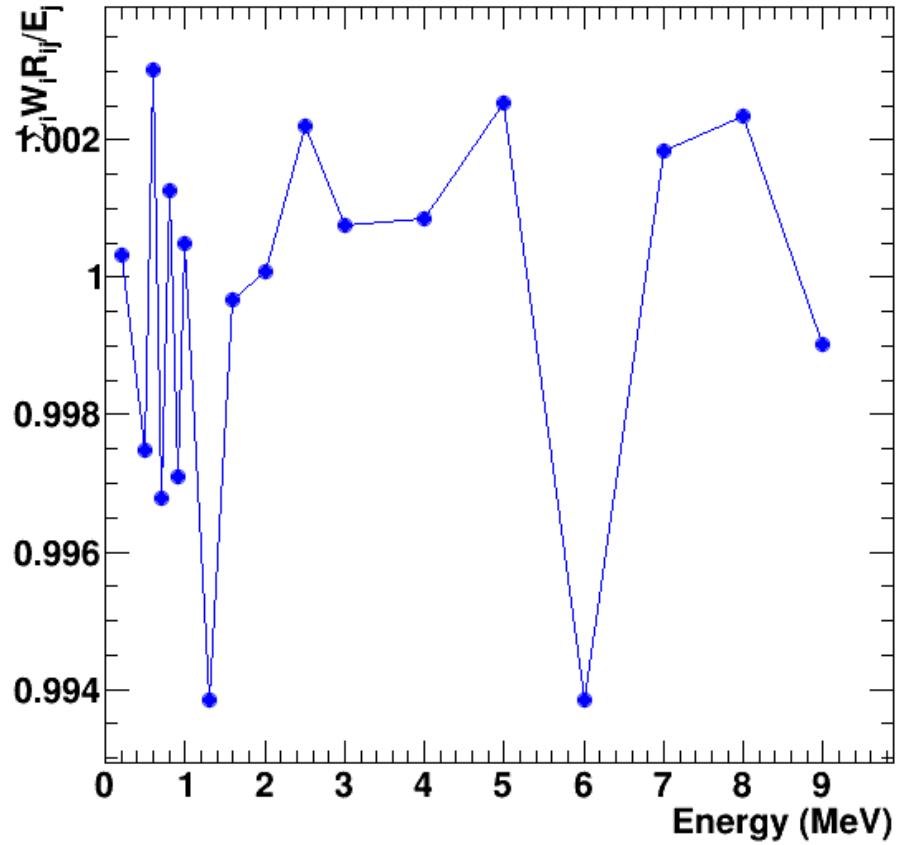
the gammas are ideally detected.

$$\epsilon_c = \epsilon_p = 1$$



Preliminary WF Det1

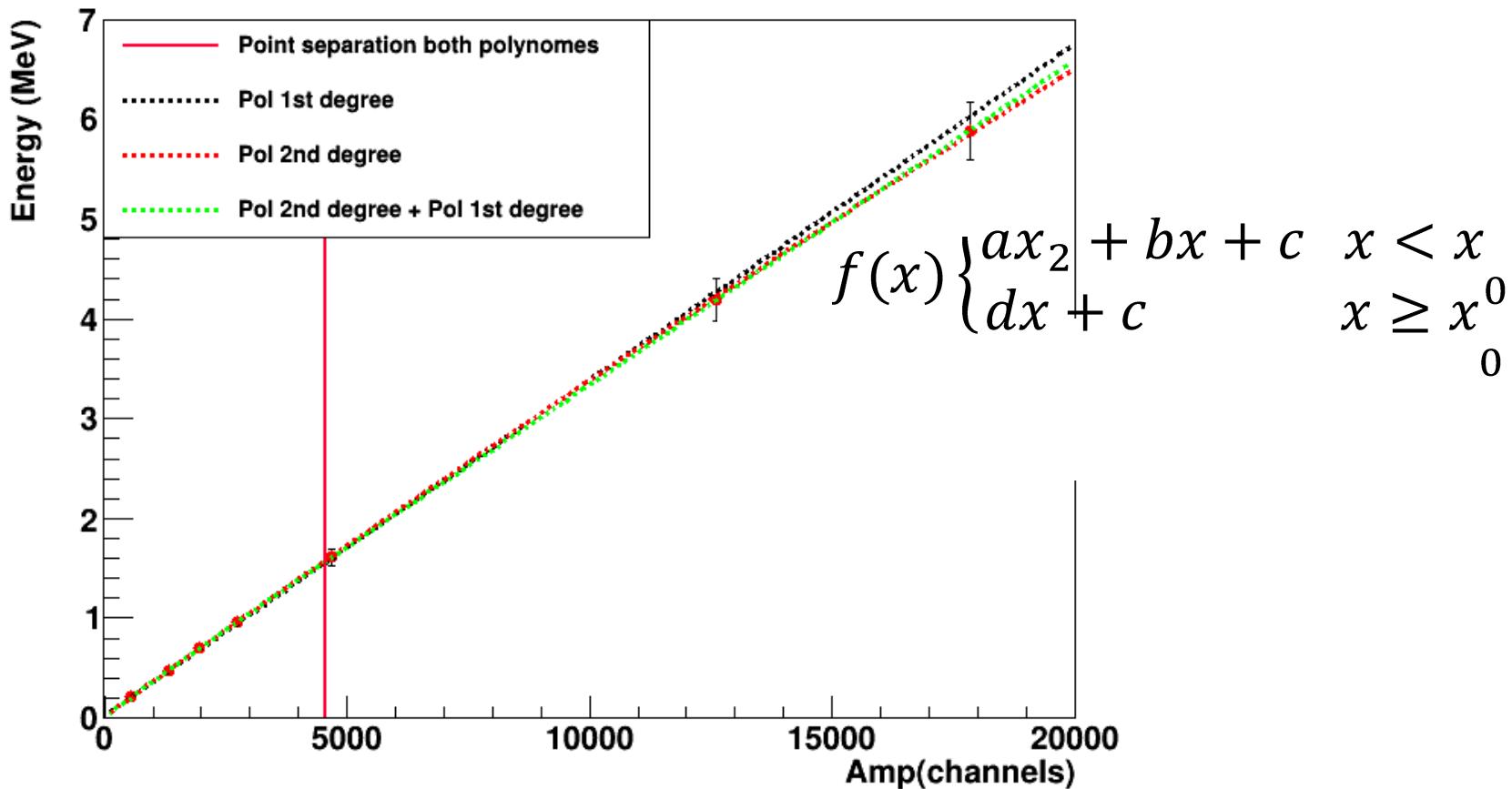
$$\sum_i W_i R_{ij} / E_j$$



Energy calibrations

- 6 sources used($^{133}\text{Ba}, ^{137}\text{Cs}, ^{60}\text{Co}, ^{88}\text{Y}, \text{AmBe}, \text{CmC}$)
- Fitting MC simulation with χ^2

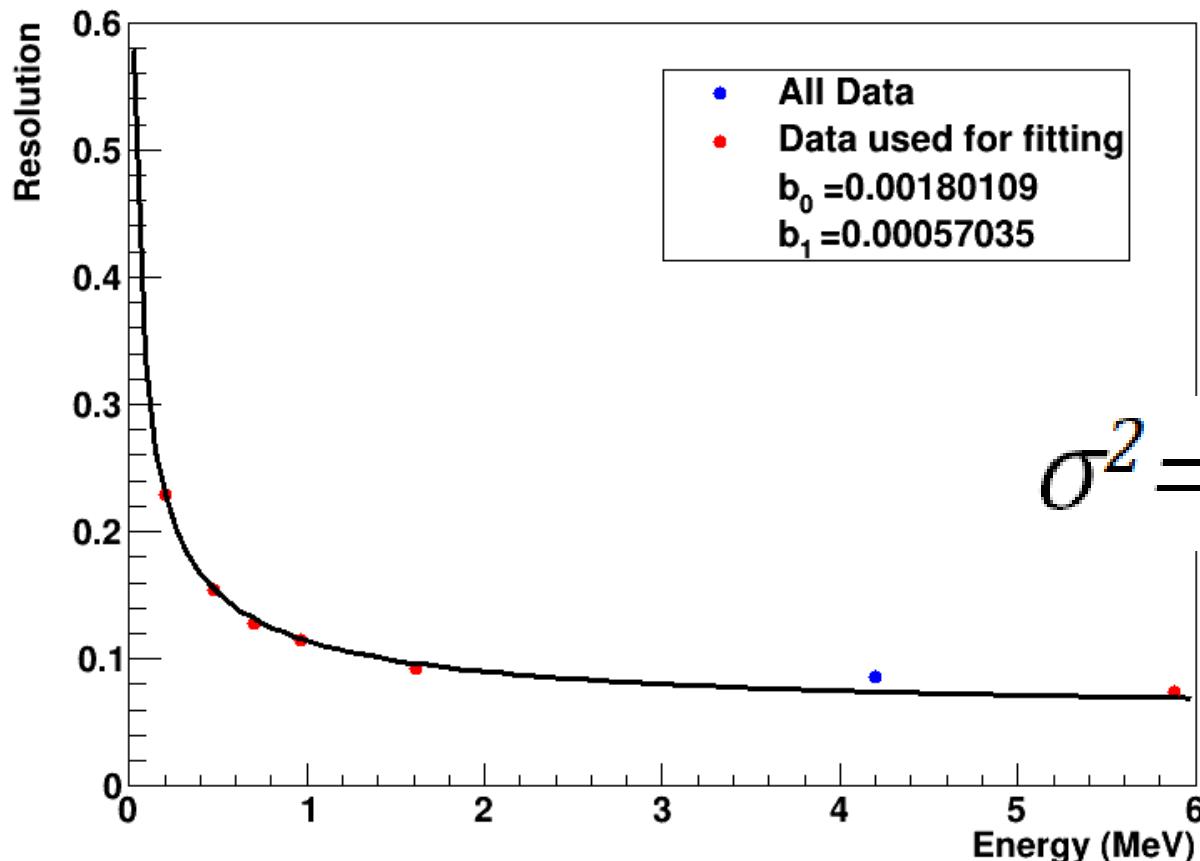
Calibration Detector 1



Energy resolution

- 6 sources used(^{133}Ba , ^{137}Cs , ^{60}Co , ^{88}Y , Ambe, CmC)
- Fitting MC simulation with χ^2 method varying gain and

results01/2/Data2Det1.txt

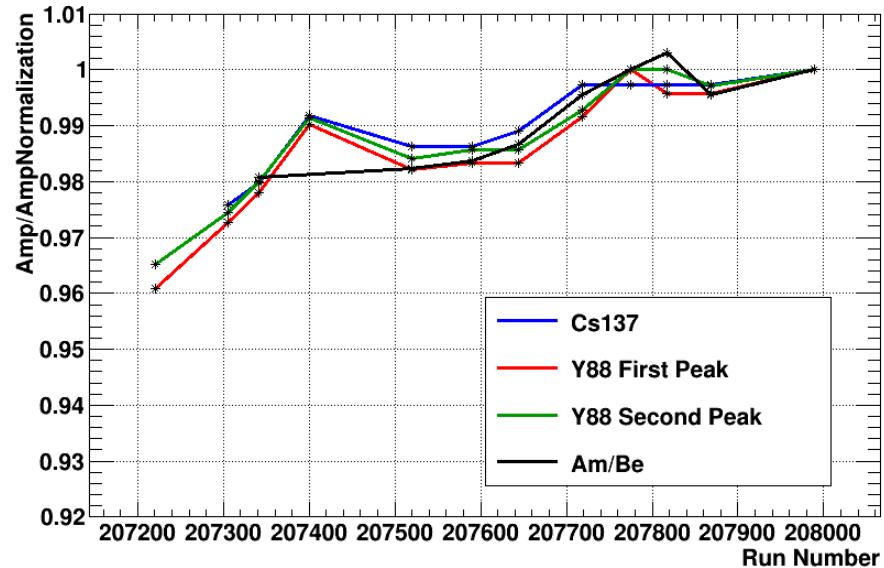


$$\sigma^2 = b_0 E + b_1 E^2$$

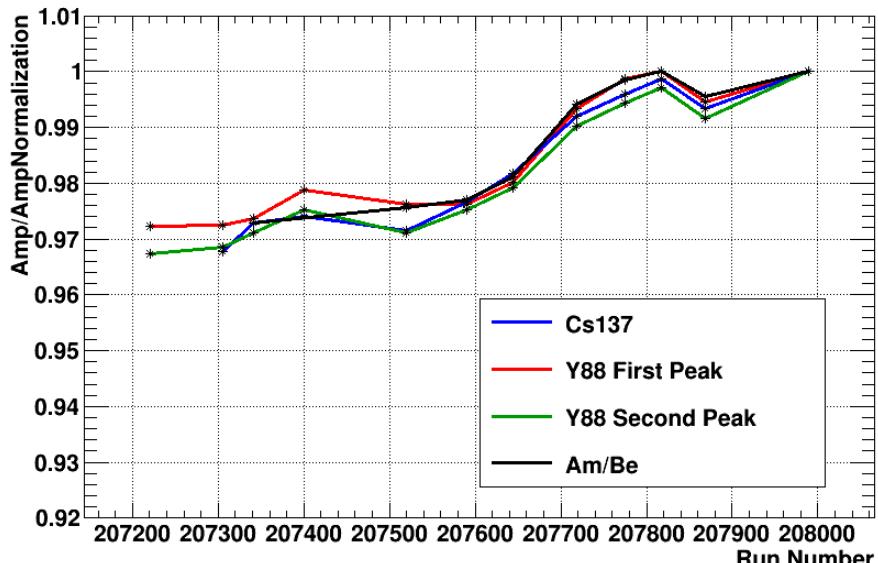
Gain Drifts

- 10 calibration measurements
- Source always in the same position.

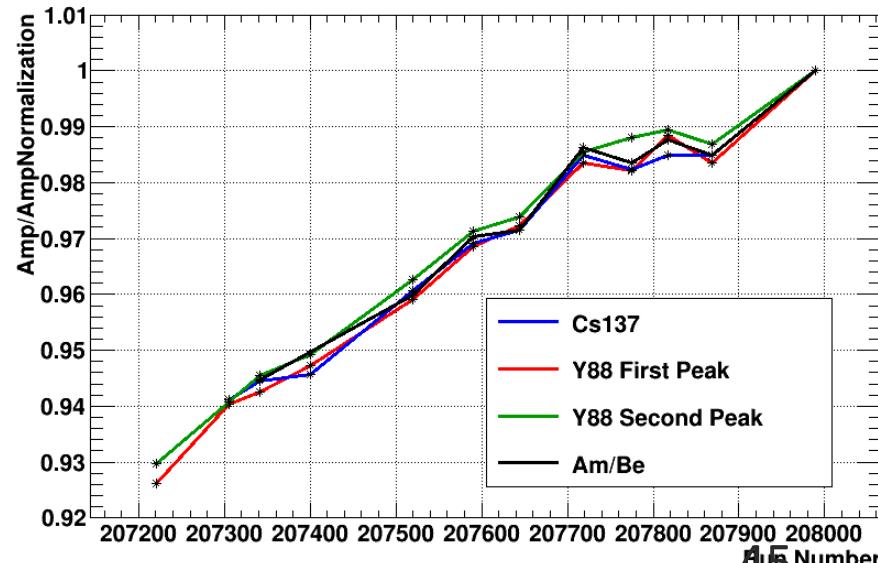
Gain Drifts for Det1



Gain Drifts for Det3

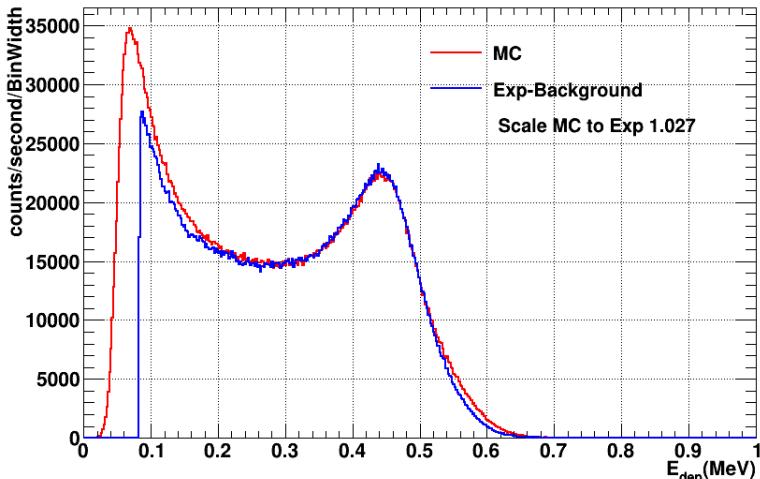


Gain Drifts for Det4

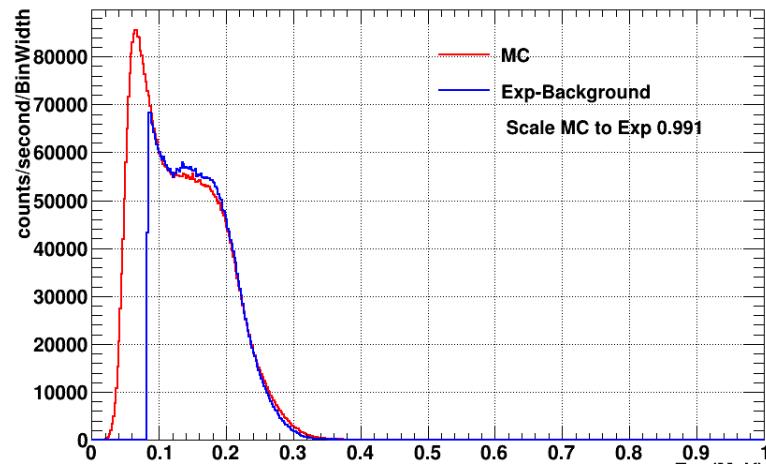


MC Simulations Source Response

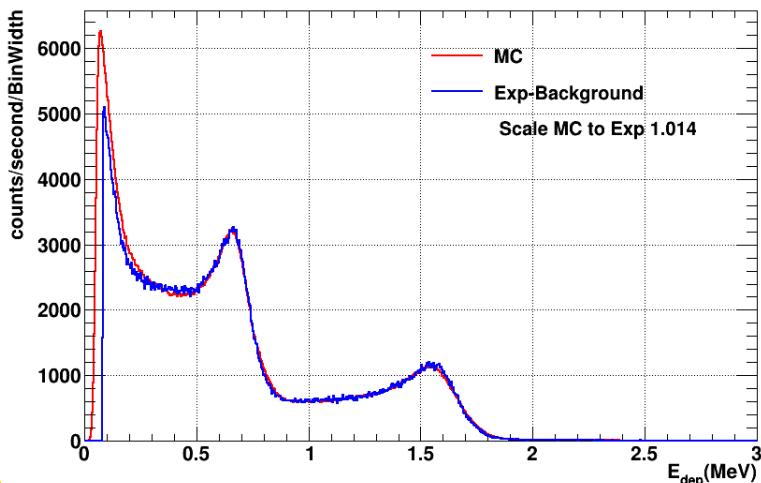
Cs Det1



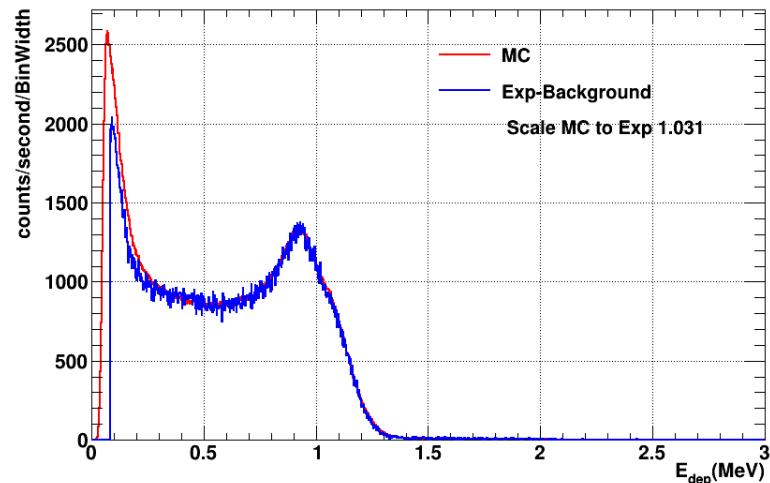
Ba Det1



Y_12 Det1

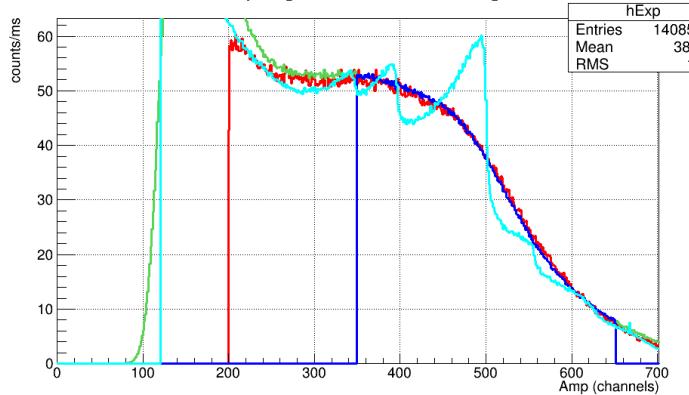


Co Det1

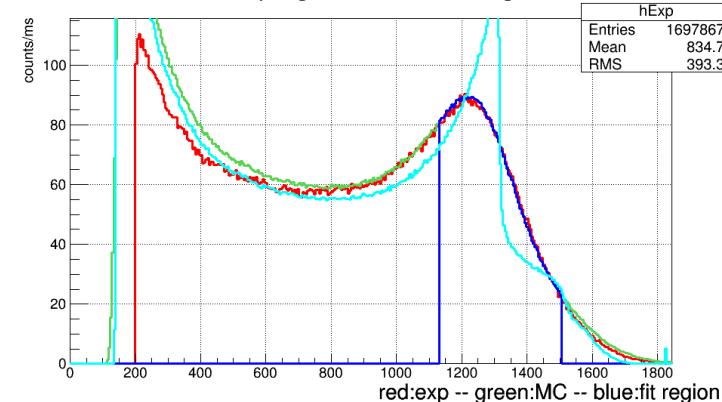


CALIBRATIONS

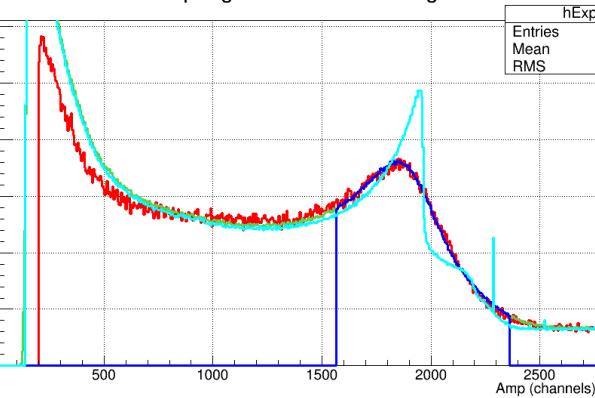
red:exp -- green:MC -- blue:fit region



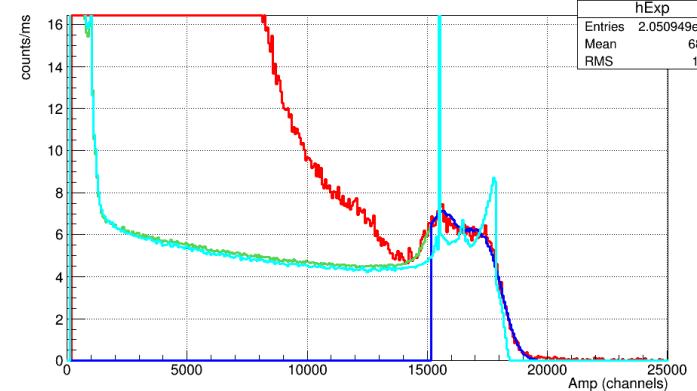
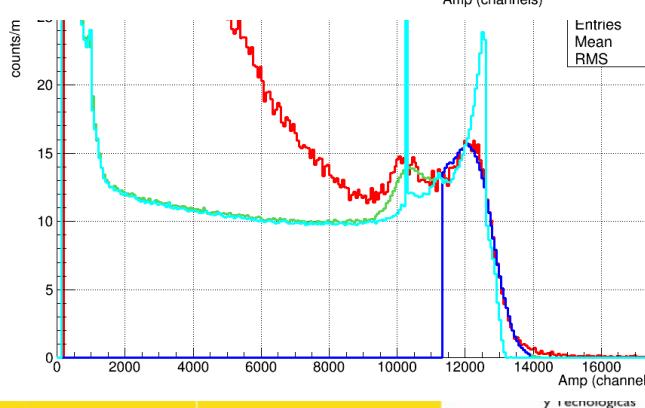
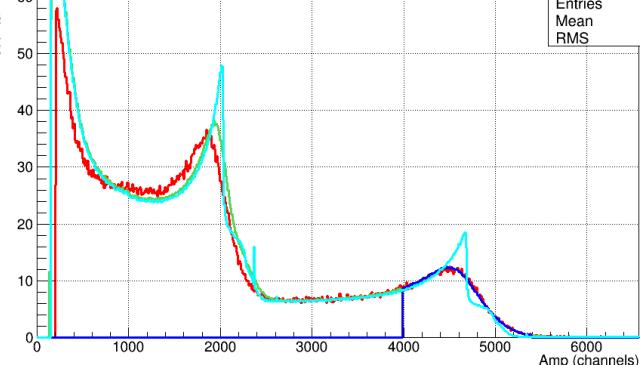
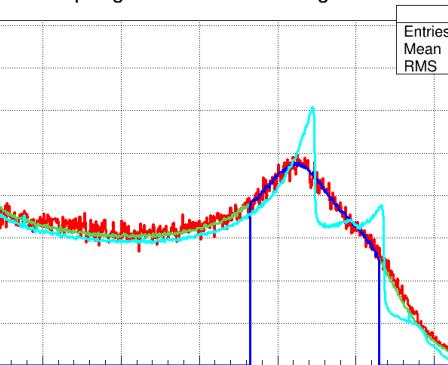
red:exp -- green:MC -- blue:fit region



red:exp -- green:MC -- blue:fit region



red:exp -- green:MC -- blue:fit region

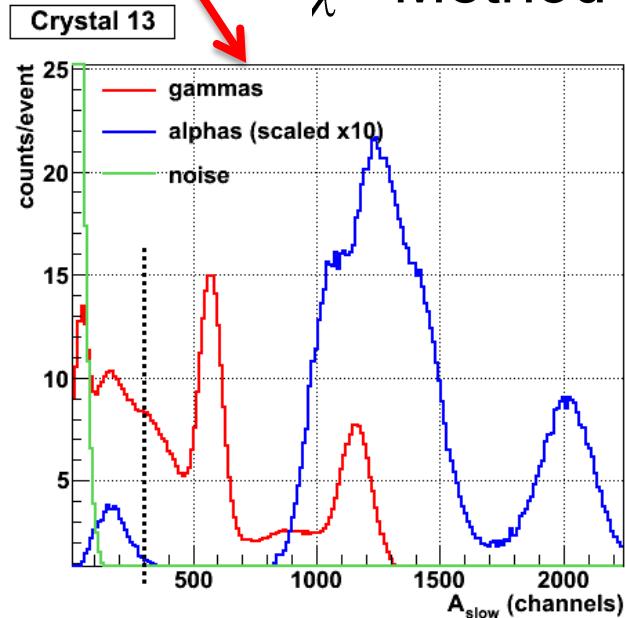


Alpha/gamma discrimination and energy and time calibration

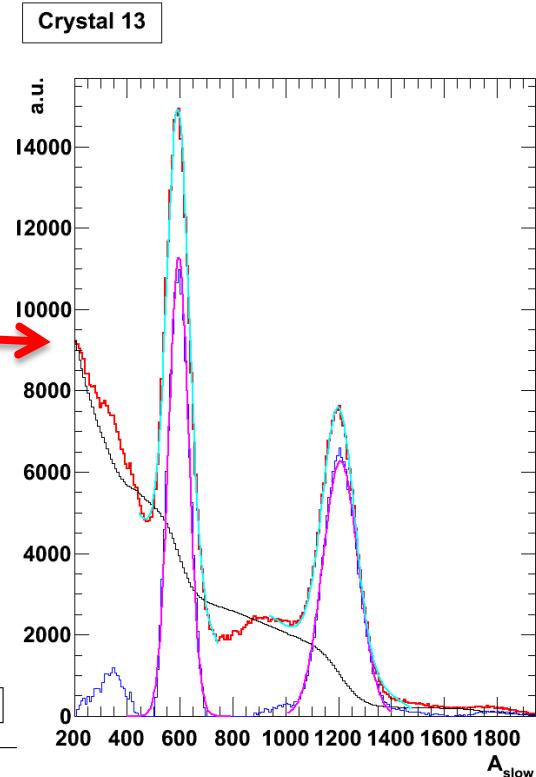
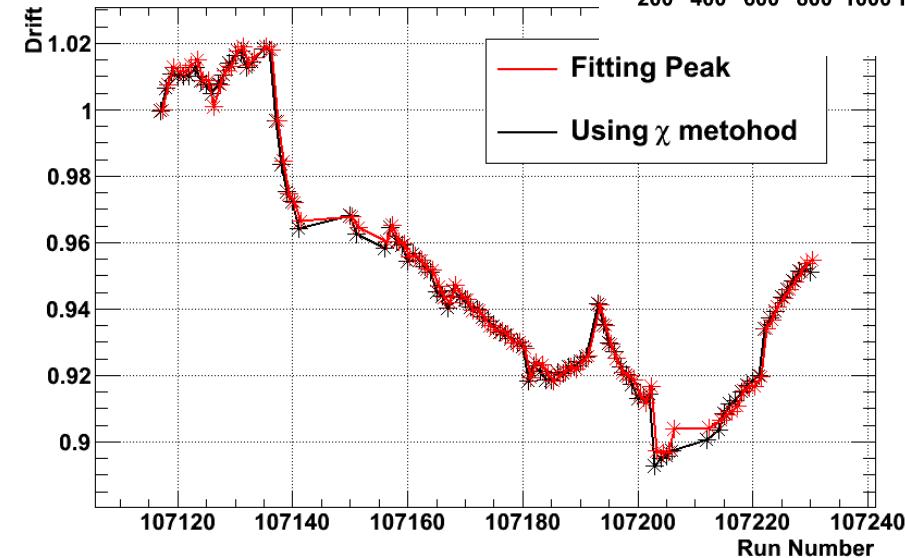
- Alpha discrimination
- Calibrations, Gaussian fitting
- Gain drift correction. Using alpha spectra

Two techniques:

- Gaussian fit of last peak
- χ^2 -Method using reference run



Gain Drift Correction Alphas Detector 11



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