Développement de chambres à fission pour l'étude des actinides avec les détecteurs 4π SCONE@NFS et TAC@n_TOF

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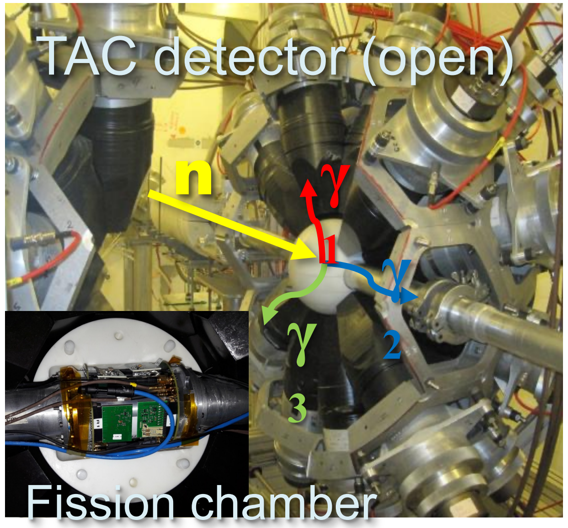
1. Fission Chamber for fission tagging

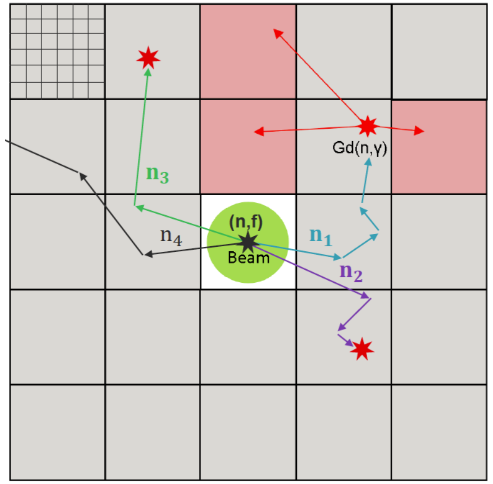
Principle of fission tagging

with the n_TOF TAC



- **Fission tagging** is necessary
  - For studying γ from fission (in coincidence mode)
  - For studying capture (in anti-coincidence mode)
- Samples are inside the fission chamber, which is inside the TAC

with SCONE



- **Fission tagging** is necessary
  - For studying n from fission (in coincidence mode)
  - For studying (n,xn) reaction (anti-coincidence)
- Samples are inside the fission chamber, which is inside SCONE

Fission Tagging development for the n_TOF TAC

TAC + FTMG-U235 [C. Guerrero et al., EPJA 48 (2012) 29]

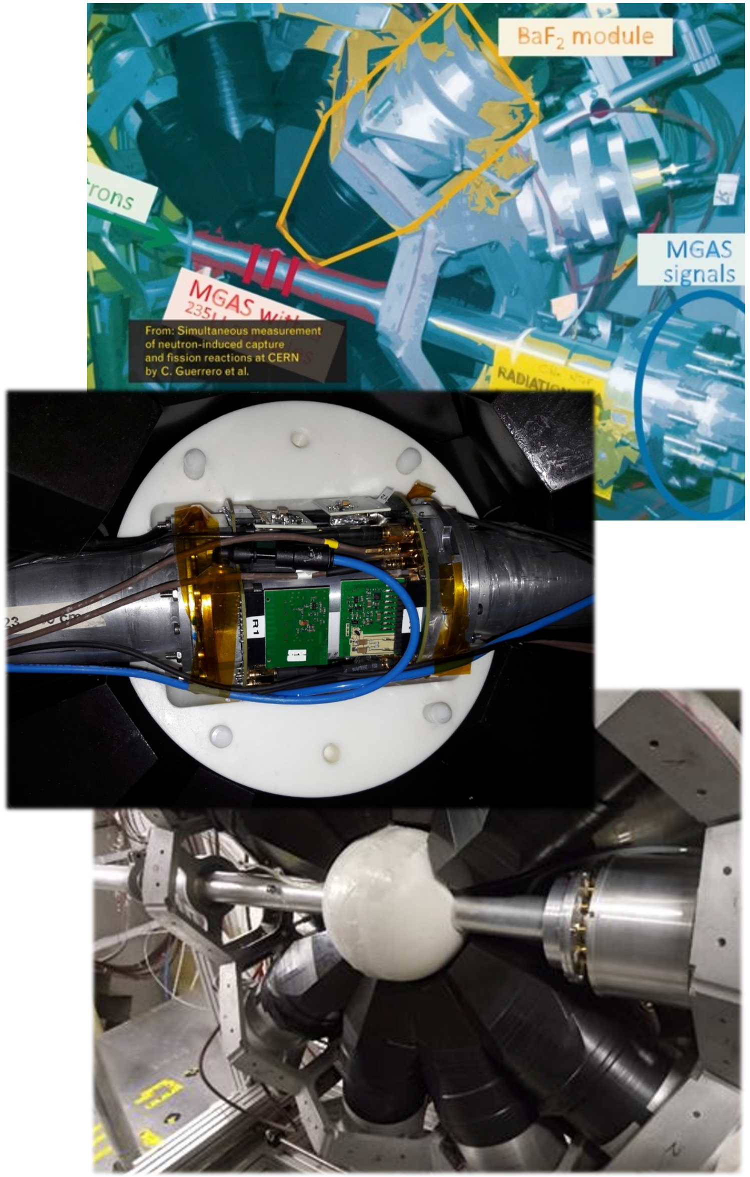
- Micromegas (MGAS) detectors / electronics outside the TAC
- Background issue due to the MGAS thick Cu mesh in the beam

TAC + FICH-U233 [M. Bacak et al., NIMA 969 (2020) 163981]

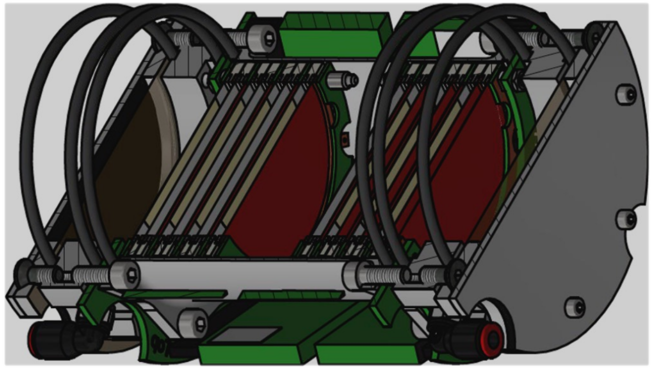
- Fission chamber
- Fast electronics plugged directly on the chamber’s PCB
- Minimum amount of material in the beam

TAC + FICH-Pu239 [J. Perkowski et al., in preparation]

- Fission chamber with minimum material in the beam
- Electronics moved off center (less background)
- Additional gas in the beam (more background)



Fission chambers for ^{233}U and ^{241}Pu



^{233}U FICH [M. Bacak, NIMA 969 (2020) 163981]

- ❑ Compact (L 12 cm x Ø 9 cm) Multi-plate chamber
  - Total of ~46 mg of ^{233}U
  - 14 samples highly enriched in 233U (JRC-Geel)
  - 14 ionization cells / Readout from 8 anodes
- ❑ Fast signals (34 ns FWHM) for high α-count rates (>1 MBq per anode)
  - Fast ionizing gas CF_4 / Fast electronics (CEA/DAM/DIF)

New ^{241}Pu FICH

- ❑ Total of ~10 mg of ^{241}Pu
- ❑ 6 to 8 samples only (more compact ⇨ less material in the beam)
- ❑ Lower gas pressure (less material in the beam ⇨ less background)
- ❑ Less massive (for reducing background from scattered neutrons)
- ❑ Improved electronics (faster signals for sustaining higher α-count rates)

Comparison/Evolution of fission chambers

	LANL-Pu241 (2010) [1]	DAM-U238 (2016) [2]	n_TOF-U233 (2016) [3]	n_TOF-Pu239 (2020) [4]	DAM-Pu239 (2023)	n_TOF-Pu241(*) (2024)
Gas and drift velocity	90%Ar-10%CH_4 6 cm/us	CF_4 11 cm/us	CF_4 11 cm/us	90%Ar-10%CF_4 12 cm/us	CF_4 11 cm/us	90%Ar-10%CF_4 12 cm/us
Gap and Voltage	12 mm 250 V/cm	2.5 mm 1300 V/cm	3 mm 1400 V/cm	4.5 mm 470 V/cm	3 mm	~3 mm
Drift time	~200 ns	23 ns	27 ns	37 ns	27 ns	~27 ns
Pressure	1.65 bars	PA + 50 mbar	1100 mbar	PA + epsilon	~100 mbar	< PA
Deposit	ø 38 mm ~200 ug/cm2 (+ VYNS film)	ø 33 mm 585 ug/cm2	ø 40 mm 264 ug/cm2	ø 20 mm 300 ug/cm2		ø 30 mm 265 ug/cm2
Cathodes	4 (SS 12.5 um)	19 (Ti 50 um)	14 (Al 10 um)	10 (Al 10 um)	8 (Al 10 um)	8 (Al 10 um)
Anodes	?	18 (Ti 50 um)	8 (Al 20 um)	10 (Al 10 um)	4 (Al 1 or 2 um)	8 (Al 10 um)
Cat. + An.		1850 um	300 um	200 um	86 um	160 um
Windows	?	Titanium (100 um)	Kapton (25 um)	Kapton (25 um)	Titanium (10 um)	Kapton (25 um)

(*) n_TOF-Pu241 with 15 mg samples (10 mg of 241Pu) of 70 MBq alpha-activity (assuming 2% 241Am)

Fission Chamber references

[1] F. Tovesson, T.S. Hill, Cross Sections for 239Pu(n,f) and 241Pu(n,f) in the Range En = 0.01 eV to 200 MeV, Nuclear Science and Engineering 165 (2010) 224; https://doi.org/10.13182/NSE09-41

[2] J. Taieb et al., A new fission chamber dedicated to Prompt Fission Neutron Spectra measurements, NIMA 833 (2016) 1–7; http://dx.doi.org/10.1016/j.nima.2016.06.137

[3] M. Bacak et al., A compact fission detector for fission-tagging neutron capture experiments with radioactive fissile isotopes, NIMA 969 (2020) 163981; https://doi.org/10.1016/j.nima.2020.163981

[4] J. Perkowski et al., Multi-section ionization fission chamber for measurement of 239Pu(n,g) reaction in fission tagging method (paper in preparation)

2.1. ^{233}U @ n_TOF (2016)

PhD Thesis of Michael Bacak

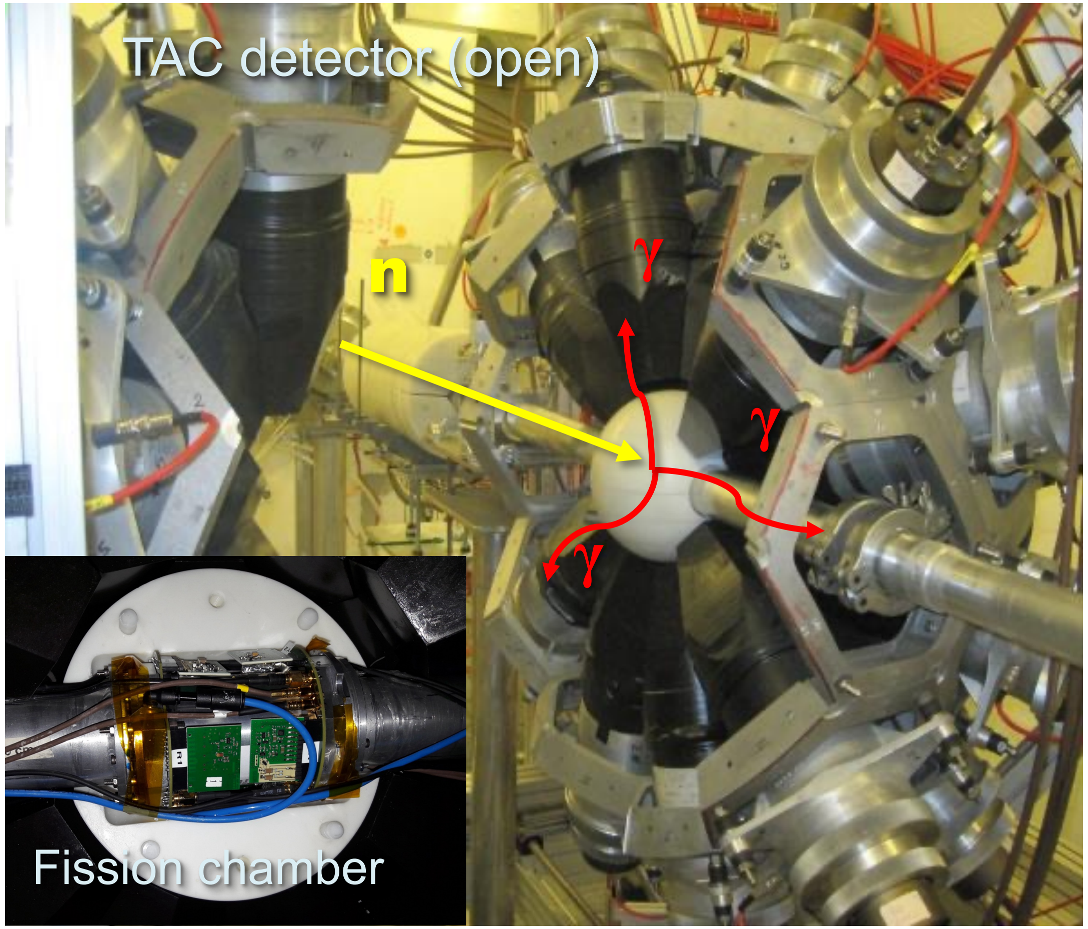
Measurement of ^{233}U @ n_TOF in 2016

Why

- U-233 capture cross section is important for reactors using the Th/U fuel cycle
- Only two measurements are available because of U-233 radioactivity and low capture cross section with respect to fission

How

- Development of a compact fission chamber to be coupled with the TAC
- Fission-tagging measurement of the U-233 capture cross section



Measurement of ^{233}U @ n_TOF in 2016

Fission Chamber
[M. Bacak, NIMA 969 (2020) 163981]

Compact (L 12 cm x Ø 9 cm)

Multi-plate chamber

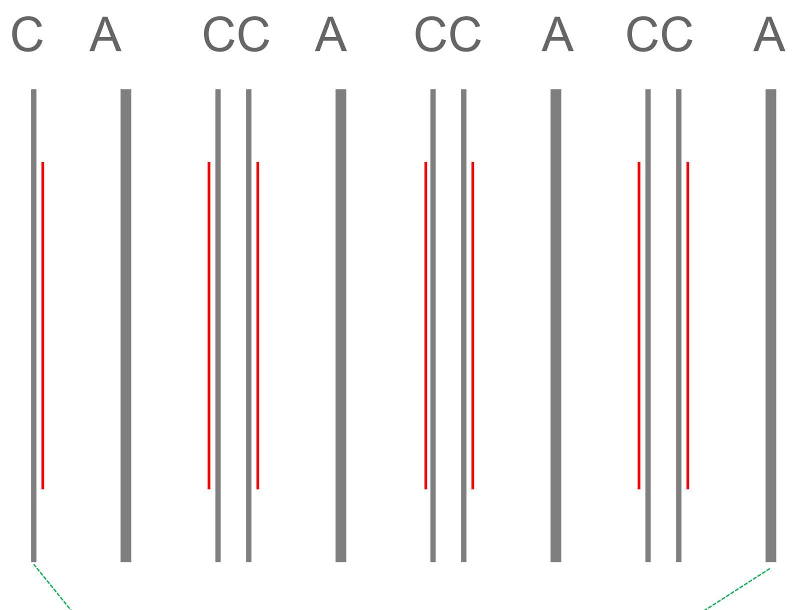
- 14 ionization cells
- Readout from 8 anodes

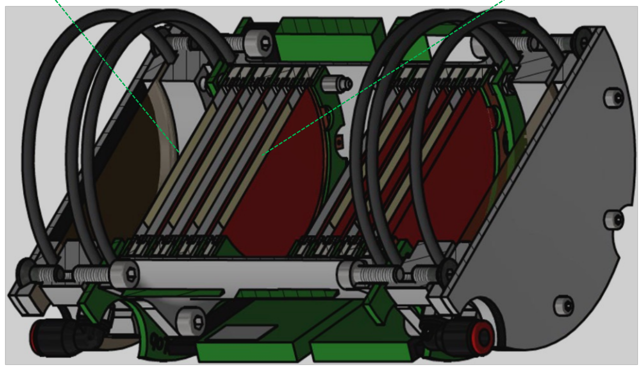
Fast signals (34 ns FWHM) for high α-count rates (>1 MBq per anode)

- Fast ionizing gas CF_4
- Fast electronics (DAM/DIF)
- Gap width: 3 mm @ 420 V

14 isotopically enriched (>99.9%) ^{233}U samples (JRC-Geel)

- Ø 4 cm
- ~46 mg ^{233}U total





Measurement of ^{233}U @ n_TOF in 2016

Data Taking

- TAC and Fission Chamber operated in coincidence
- Six weeks of measurement
- 950 TB of data

Analysis

- Multi-parametric data analysis (coincidences, anti-coincidences, crystal multiplicity, Sum energy, gamma spectra)
- Monte-Carlo simulations for various corrections (thresholds, background, efficiency)
- Still fine-tuning the analysis while finalizing the publication

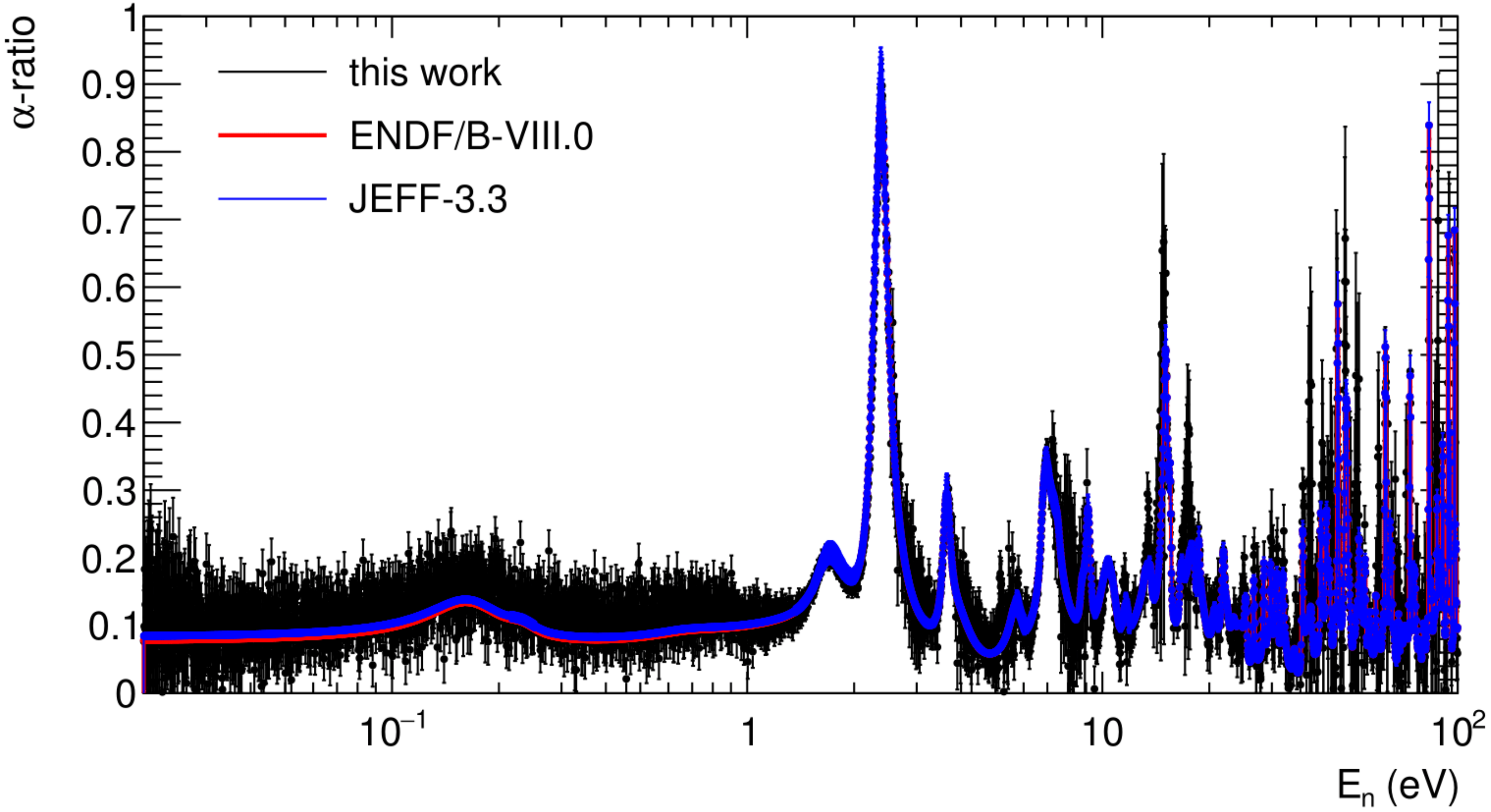
Measurement of ^{233}U @ n_TOF in 2016

Preliminary alpha-ratio
[M. Bacak, EPJ Conf 239 (2020) 01043]

- Overall consistency with evaluated libraries
- Some deviations under investigations
- Final paper still to be published

$$\alpha = \frac{\sigma_\gamma}{\sigma_f} \sim 0.1$$

$3 \leq m_{cr} \leq 7$
$2.5\ \text{MeV} \leq E_{Sum} \leq 7\ \text{MeV}$



2.2. ^{238}U @ NFS (2021)

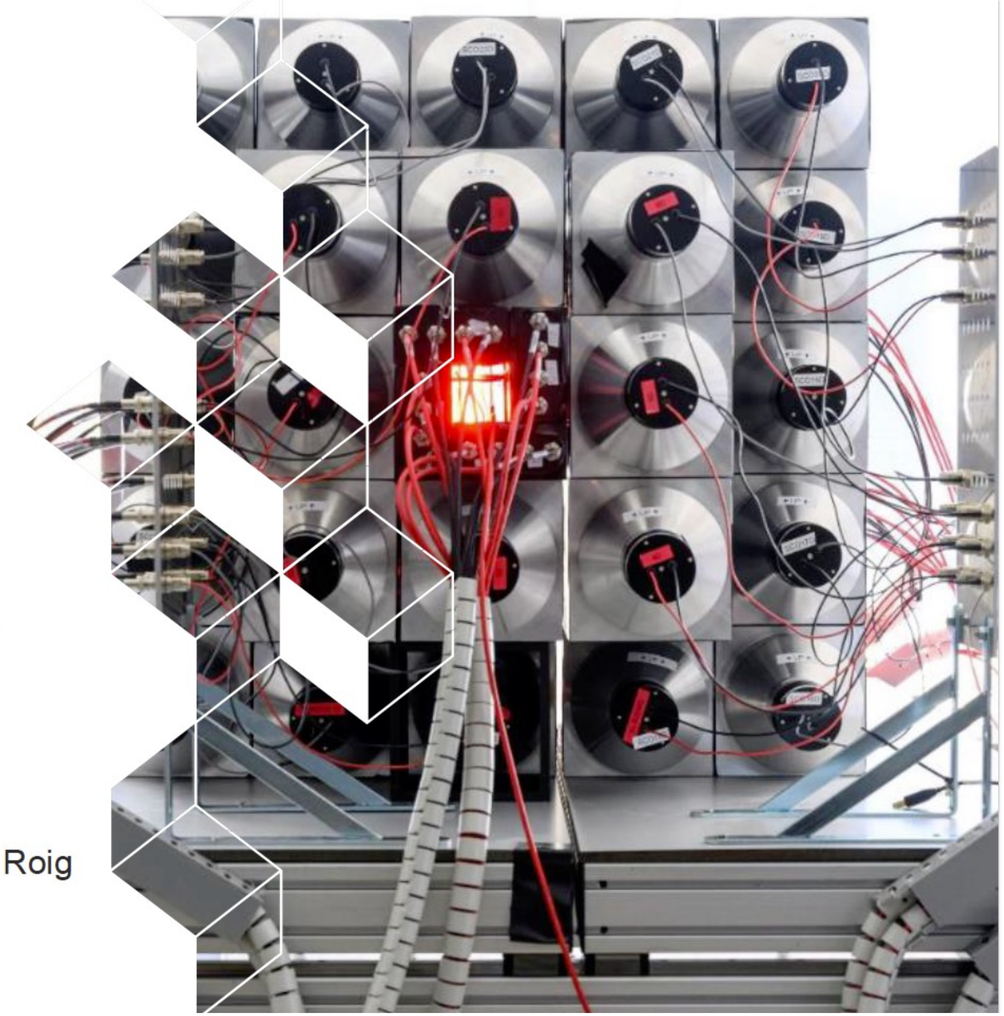
Measurement of ^{238}U @ NFS in 2021

30/11/2023

First experimental determination of the second-chance fission probability

B. Fraïsse, G. Bélier, V. Méot, L. Gaudefroy, O. Roig

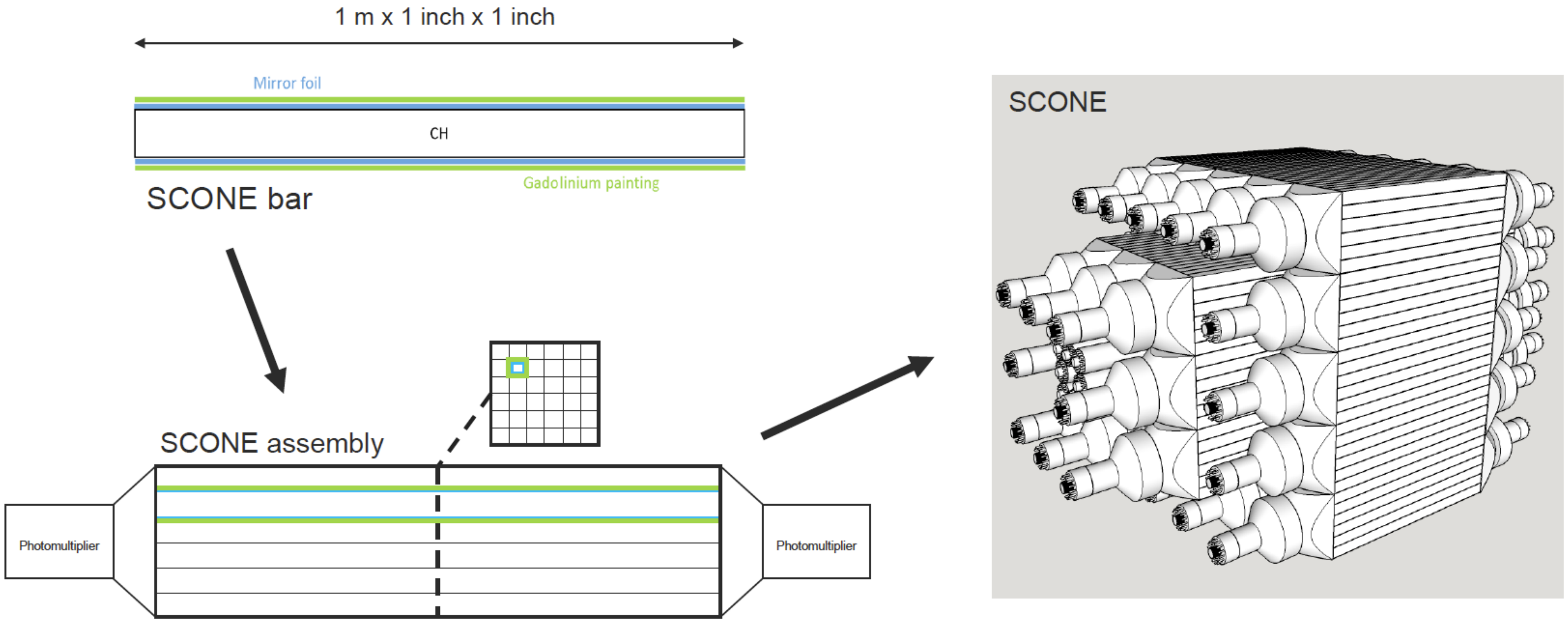
CEA, DAM Île-de-France, Bruyères-le-Châtel



B. Fraïsse et al, JEFF Meeting, OECD/NEA, November 2023, JEF/DOC-2292

Measurement of ^{238}U @ NFS in 2021

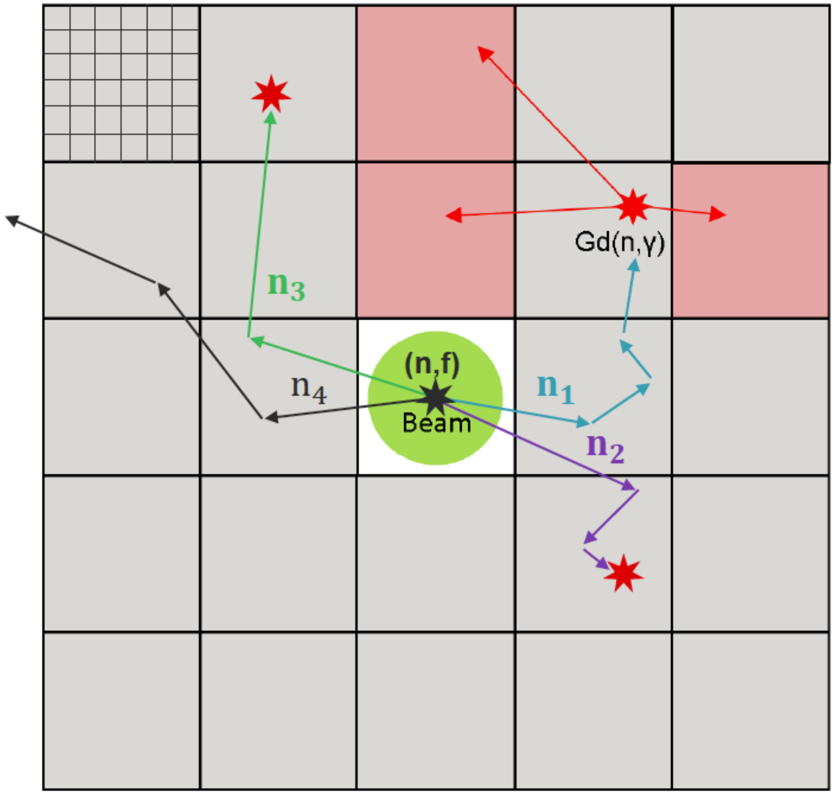
SCONE detector

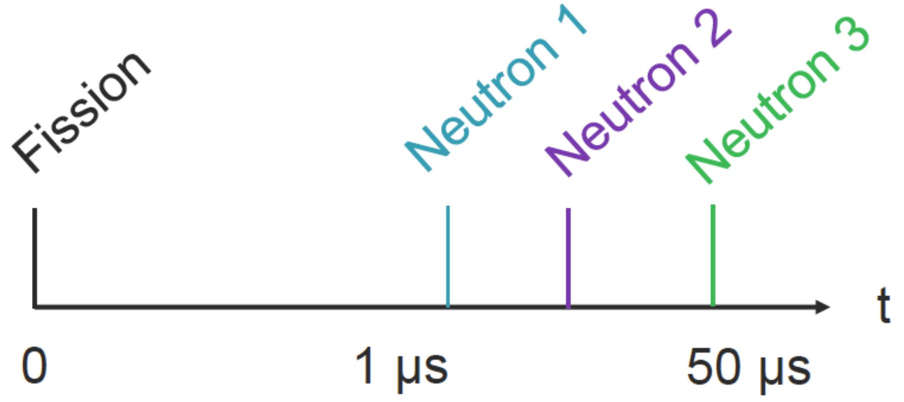


From B. Fraïsse et al, JEFF Meeting, OECD/NEA, November 2023, JEF/DOC-2292

Measurement of ^{238}U @ NFS in 2021

SCONE detector



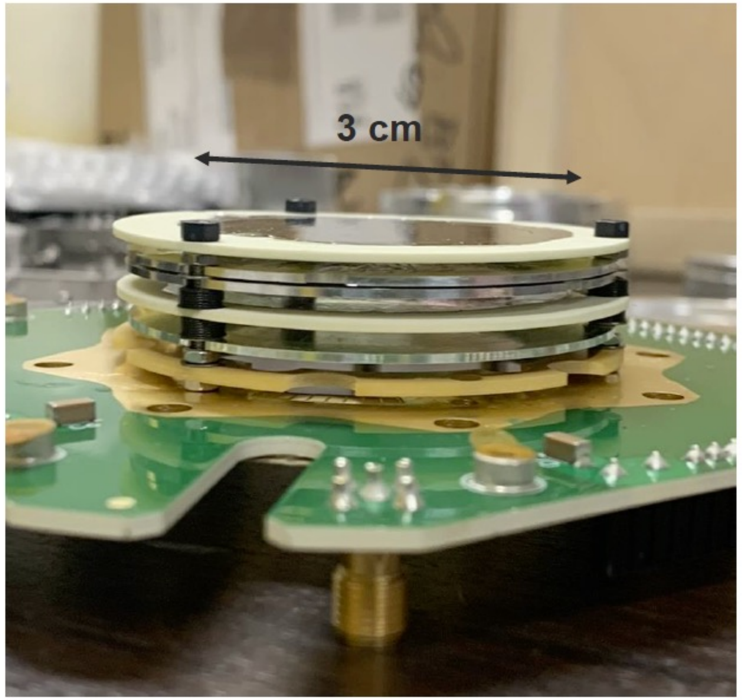


Efficiency ~ 70 %

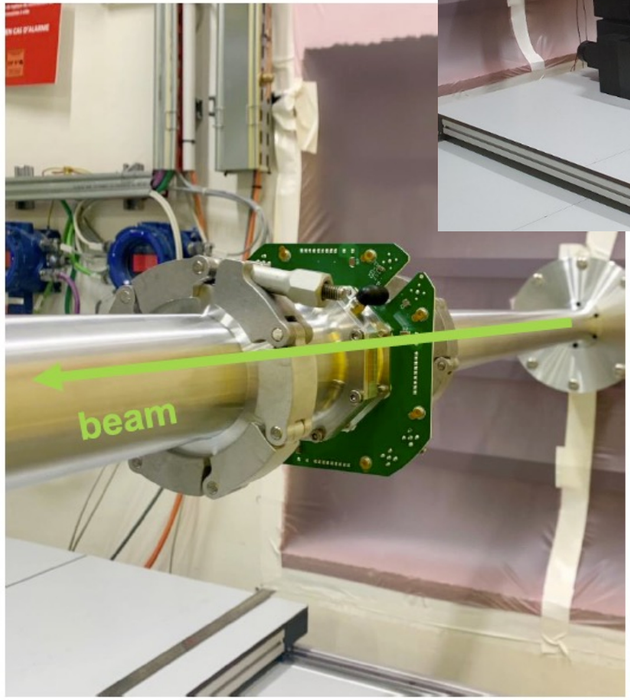
From B. Fraïsse et al, JEFF Meeting, OECD/NEA, November 2023, JEF/DOC-2292

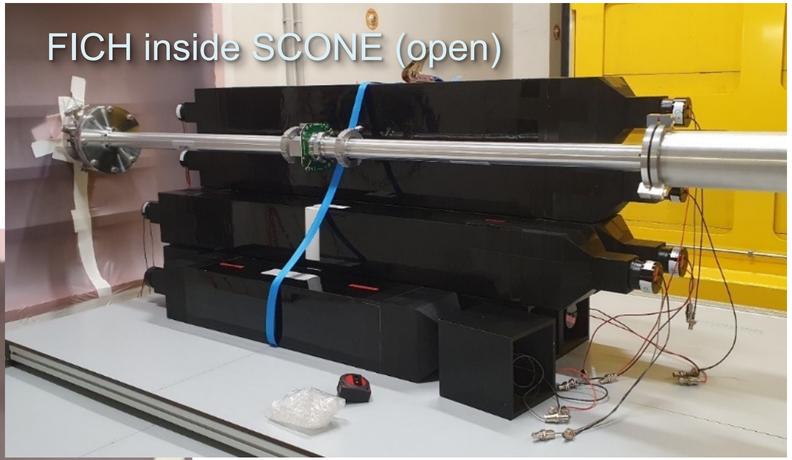
Measurement of ^{238}U @ NFS in 2021

Fission chamber



Fission chamber structure





Fission chamber in the pipe

From B. Fraïsse et al, JEFF Meeting, OECD/NEA, November 2023, JEF/DOC-2292

Measurement of ^{238}U @ NFS in 2021

Conclusion

- First experiment with SCONE at NFS (GANIL)
- First measurement of complete p(n) for uranium-238 up to 30 MeV
- Signature of second-chance fission on the standard-deviation
- Second-chance probability based on data : **constraint for evaluations ?**

From B. Fraïsse et al, JEFF Meeting, OECD/NEA, November 2023, JEF/DOC-2292

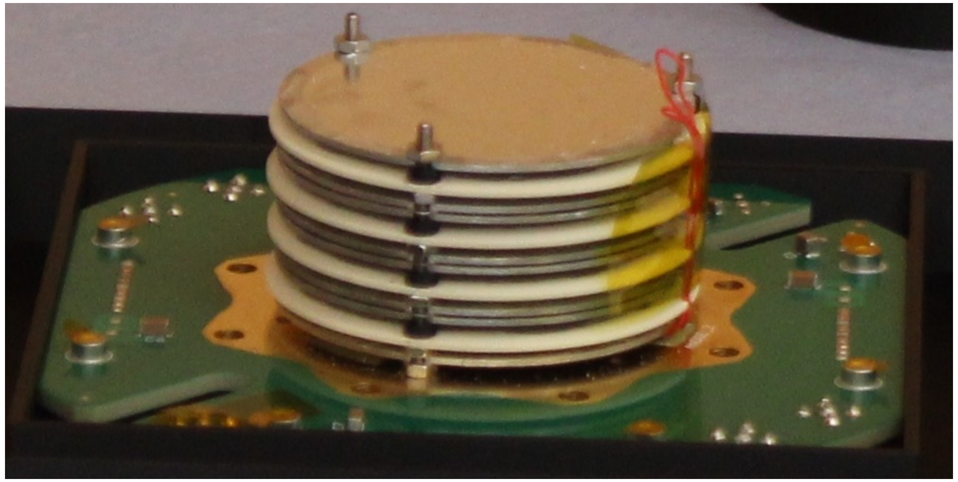
2.3. ^{239}Pu @ NFS (2023)

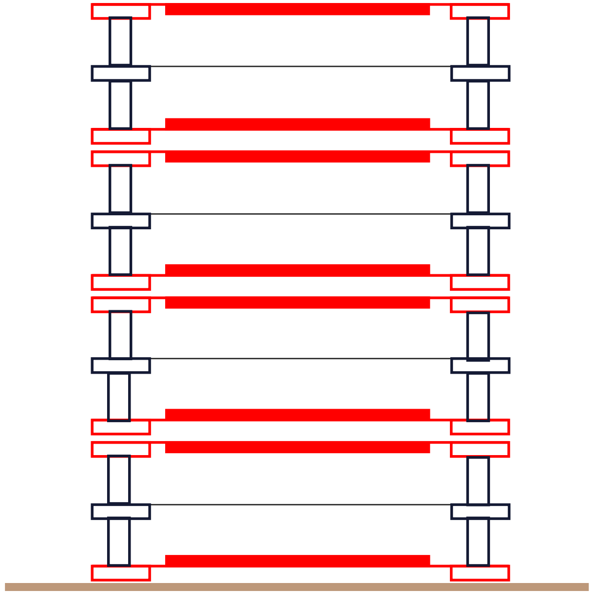
Fission chamber design

FICH DAM-Pu239 (8 deposits and 4 anodes)

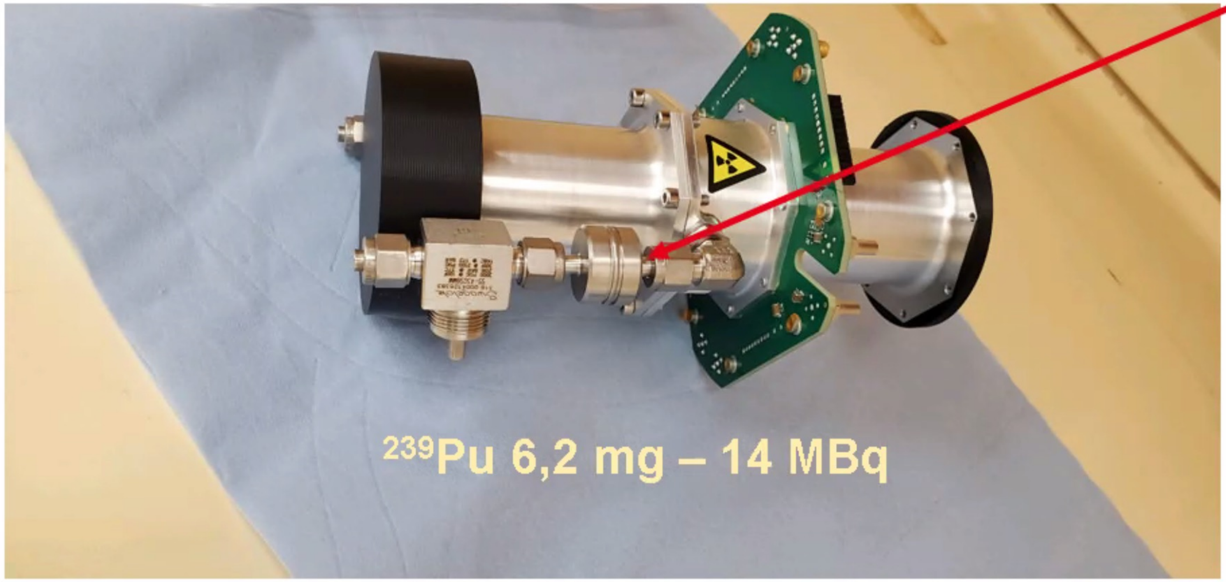
Stack of biased anodes (in black) and grounded cathodes with deposit (in red)

Gap of 3 mm => stack of 3 cm

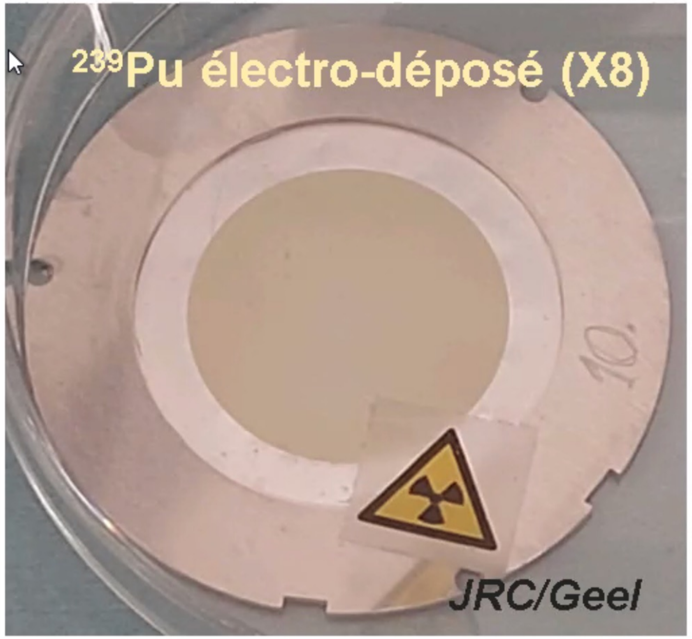




Fission chamber design

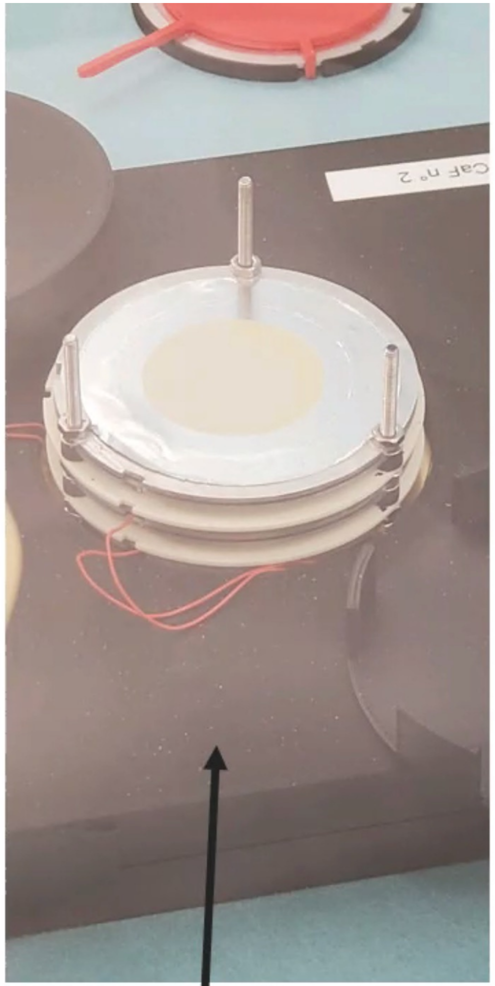


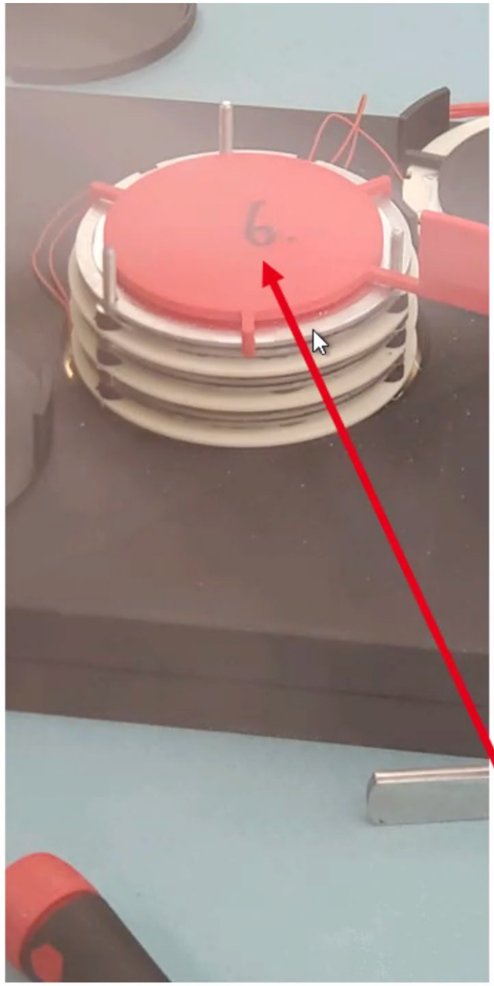
- Filtres THE métalliques
- Dépôts sur substrat Al 10 µm
- Fenêtres Ti 10 µm
- Nouvelles brides + compactes

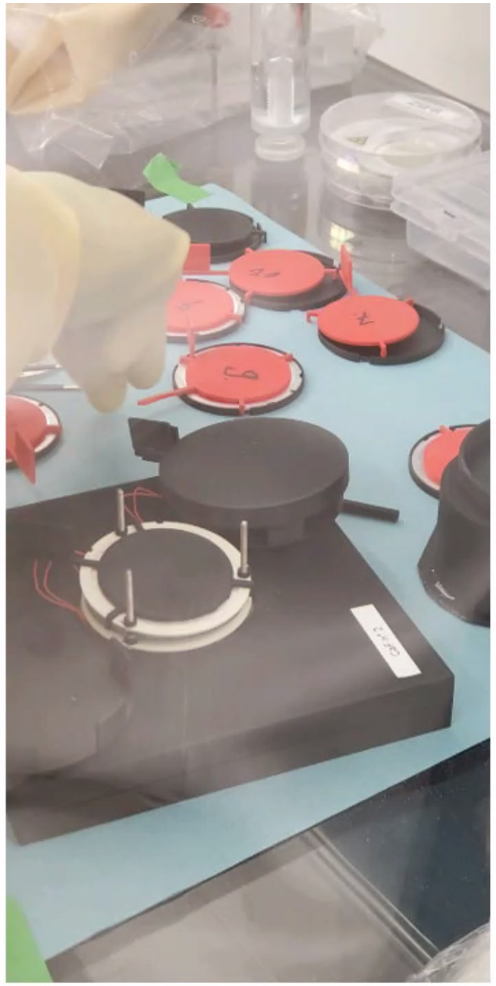


- Test de chute validé par le GANIL
- Tests de mise en pression sur la DIF

Mounting of the samples at JRC-Geel



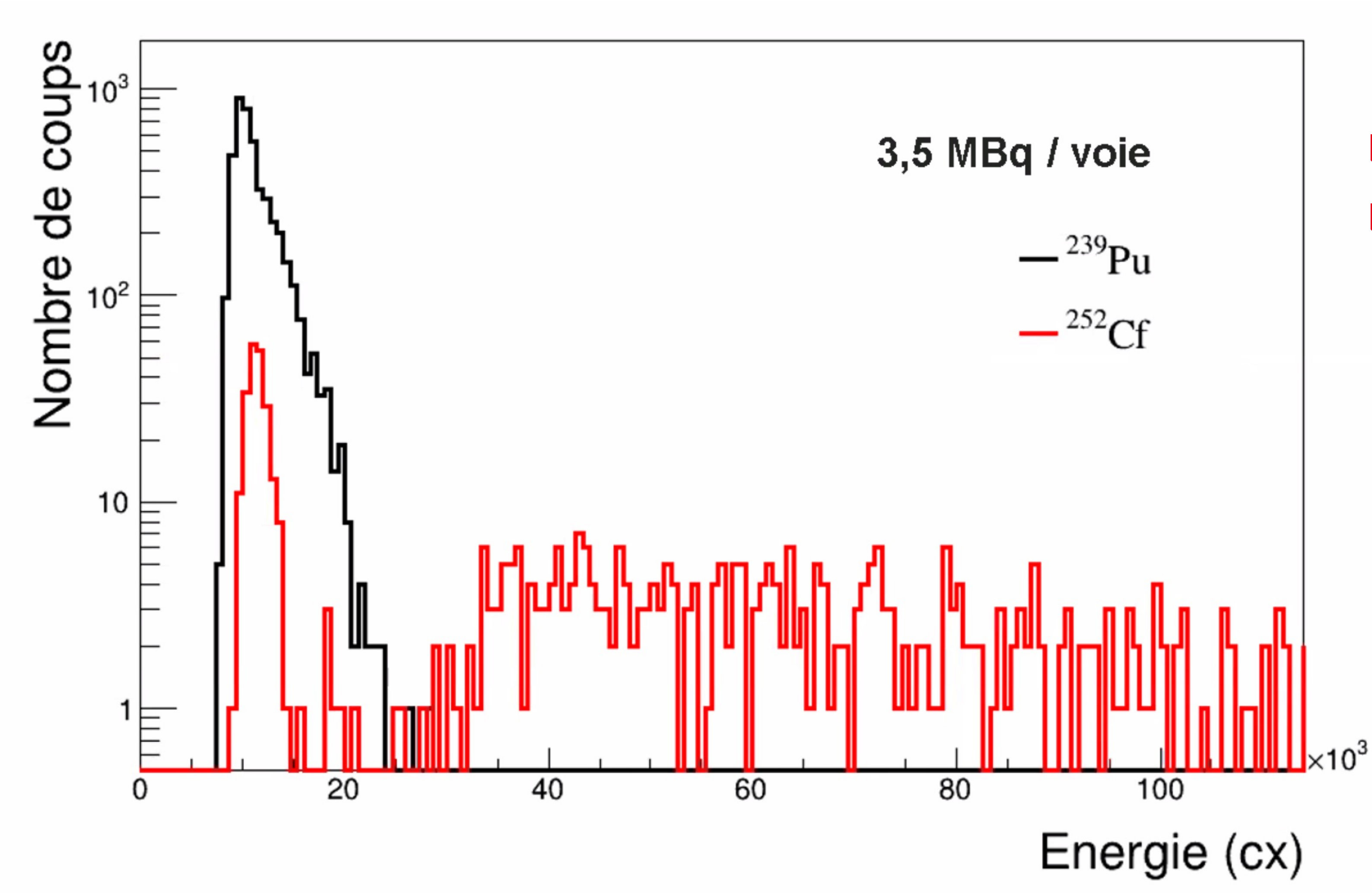




Boîtier de protection du PCB

Dispositif de manipulation d'échantillon

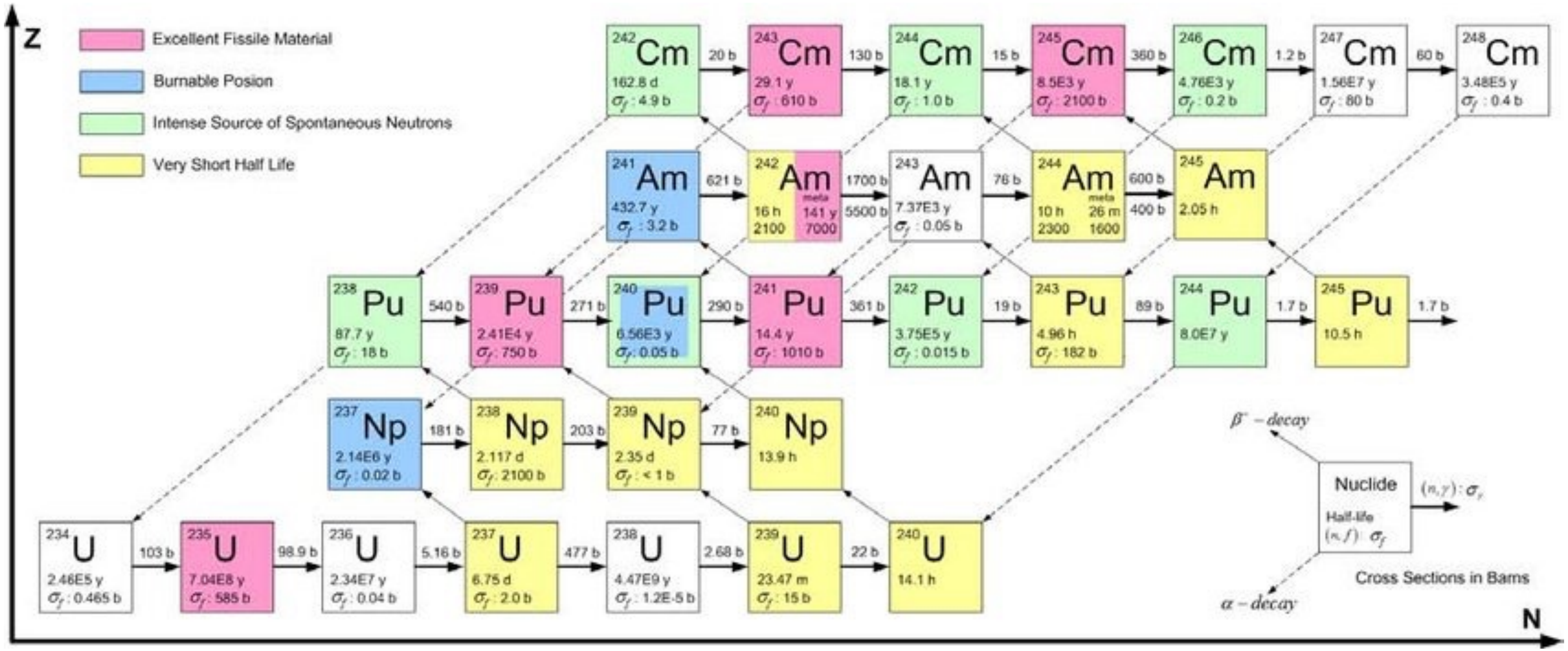
Fission chamber validation test



- Gas pressure of 800 mbar
- Good α-FF separation

2.4. ^{241}Pu @n_TOF (2025)

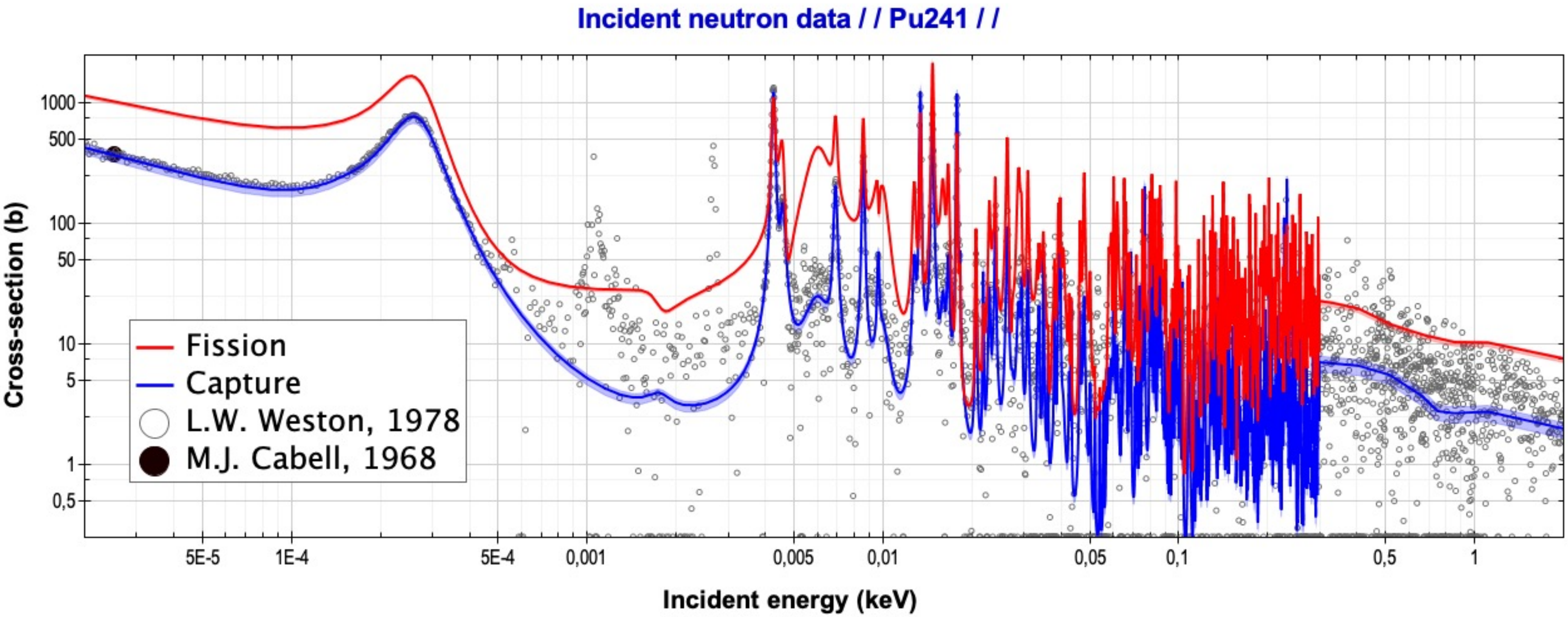
Motivations



- Plutonium is produced in reactors, ^{239}Pu and ^{241}Pu both contribute to energy production
- ^{241}Pu xs are not well known because of its short half-life (~14 years) and the ^{241}Am build-up

Capture and fission cross sections

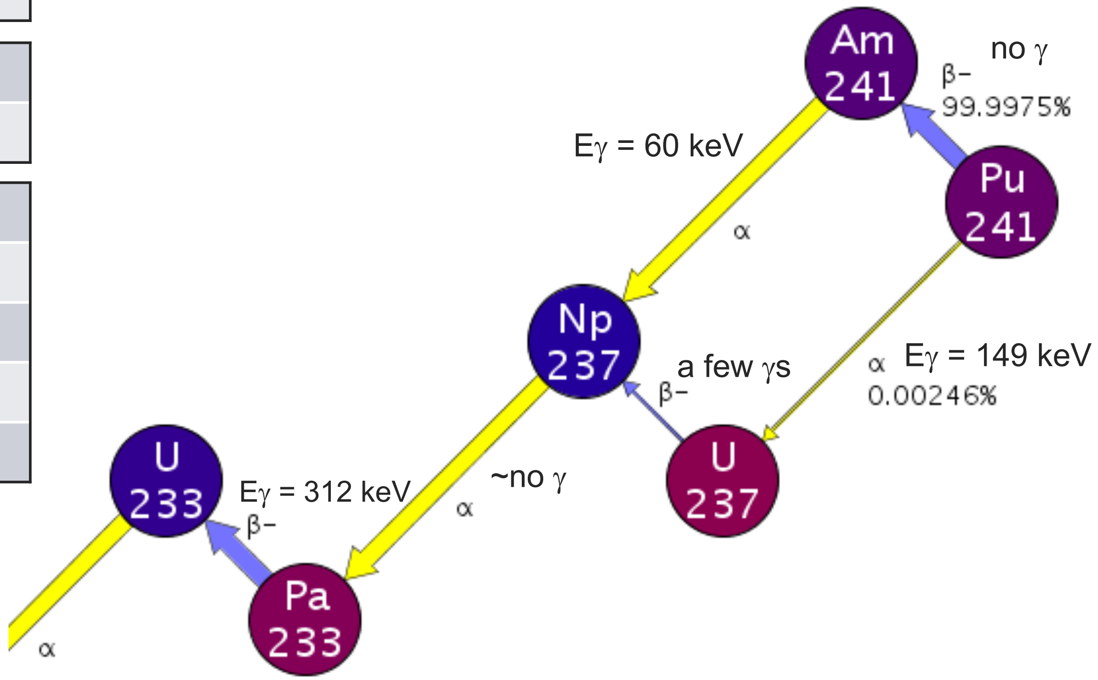
- The fission cross section of ^{241}Pu is at least three times larger than its capture cross section
- There is only one time-of-flight measurement for capture, made by Weston at ORNL in the 70's



Radioactive decay of Pu and daughters

Parent nucleus	$T_{1/2}$	Decay mode	Daughter nucleus	$T_{1/2}$
^{241}Pu	14.3 y	β- (~100%)	^{241}Am	433 y
^{241}Am	433 y	α (100%)	^{237}Np	2.1 My
^{241}Pu	14.3 y	α (<<1%)	^{237}U	6.75 d
^{237}U	6.75 d	β- (100%)	^{237}Np	2.1 My
^{237}Np	2.1 My	a (100%)	^{233}Pa	27 d
^{233}Pa	27 d	b- (100%)	^{233}U	159 ky
^{238}Pu	87.7 y	α (100%)	^{234}U	245 ky
^{239}Pu	24.1 ky	α (100%)	^{235}U	704 My
^{240}Pu	6.6 ky	α (100%)	^{236}U	23 My
^{242}Pu	373 ky	α (100%)	^{238}U	~stable
^{244}Pu	81 My	α (100%)	N/A	

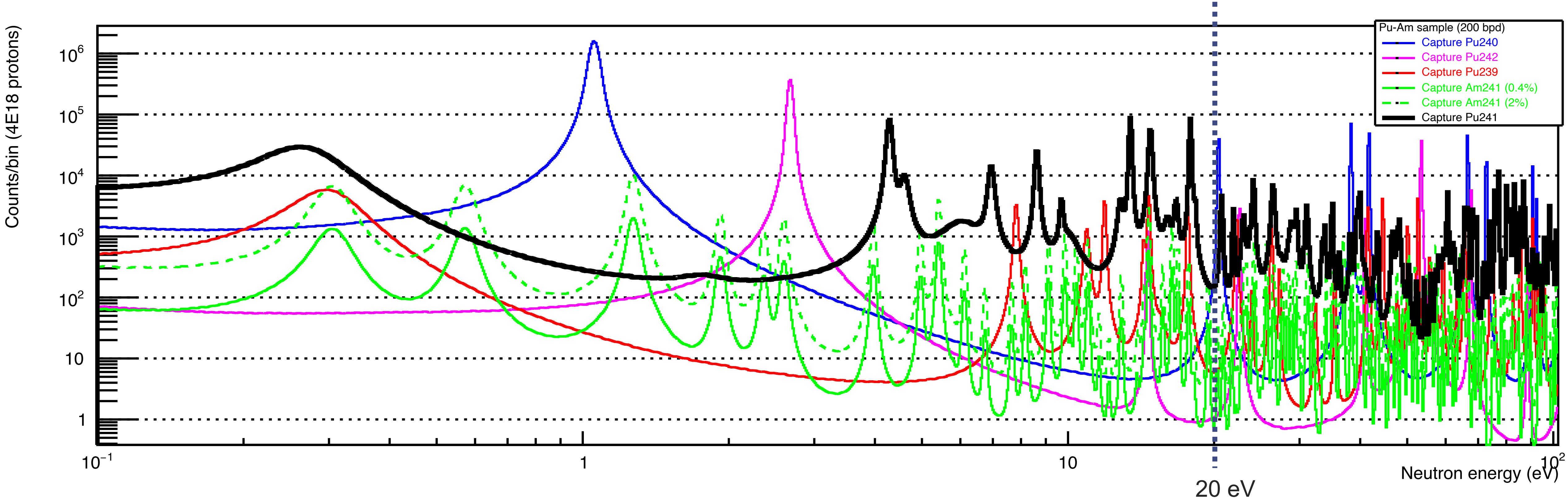
- High activity (β > 99.99% with Q ~21 keV)
- β decay populates ^{241gs}Am only (i.e., no γ)



Possible issue for capture xs interferences

Possible issue for α-background in the FICH

Simulation of capture counting rates



- Binning adapted to have a statistical precision better than ~3% (~10^3 counts/bin)
- E < ~20 eV ⟹ 200 bpd (bins per decade) allow for good resolution and statistics

Alpha-activity from a 15 mg sample (~10 mg of ^{241}Pu)

Composition calculated in 2025 four months after the last Am purification

	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241	Total
Atom %	**0.07%**	4.3%	**16.6%**	65.3%	12.4%	**~1%**	100%
α-activity (MBq)	**6.5**	1.5	**21**	0.9	0.3	**~20**	~50

- Significant contributions from ^{238}Pu, ^{240}Pu, and especially ^{241}Am
- Total contribution from Pu isotopes is 30 MBq (in 4π)
- ^{241}Am build-up would add between 20 MBq (1% build-up) and 40 MBq (2% build-up)
- The new FICH should be able to handle such an α-background, i.e. up to ~5 MBq/channel (see tests at 14 MBq/channel in Laurent et al., NIMA 990 (2021) 164966)

Deposits of ~10 mg of ^{241}Pu (15 mg of Pu)

Assuming a maximum activity in 4π of 70 MBq (30 MBq from Pu + 40 MBq from 2% ^{241}Am)

- 1 anode for every deposit (on the cathode), i.e. alpha-activity in 2π only
- deposits of diameter 40 mm or 30 mm

		4 anodes	6 anodes	8 anodes	10 anodes
α-activity/channel		8.8 MBq	5.8 MBq	4.4 MBq	3.5 MBq
Deposit thickness	ø = 40 mm	298 μg/cm²	200 μg/cm²	150 μg/cm²	
	ø = 30 mm		354 μg/cm²	265 μg/cm²	212 μg/cm²

=> 8 deposits of ø 30 mm should be ok (8 channels is the maximum with the "FICH-U3" design)

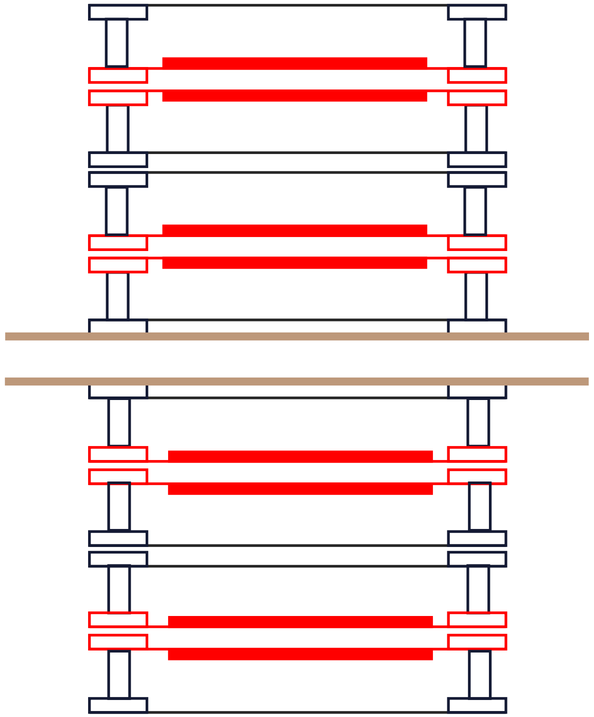
Fission chamber preliminary design

FICH n_TOF-Pu241 (8 deposits and 8 anodes)

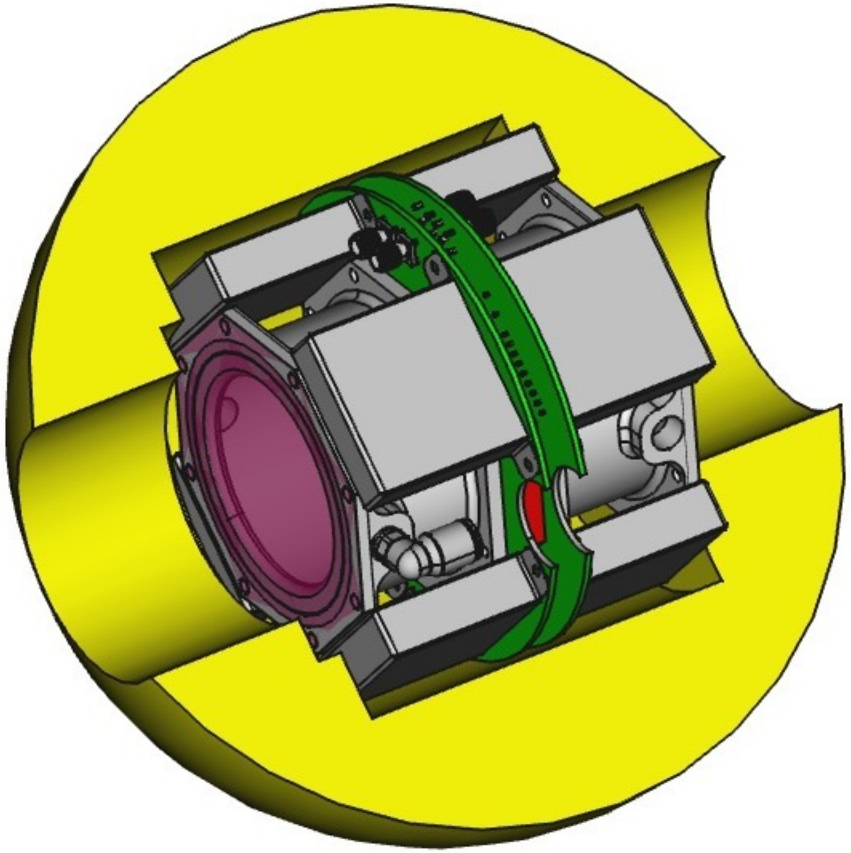
Stack of biased anodes (in black)
and grounded cathodes with deposits (in red)

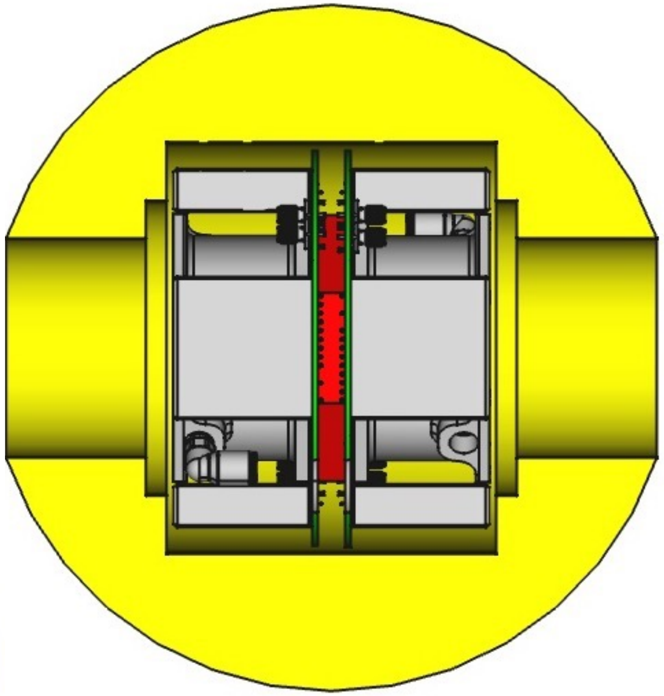
Gap of 3 mm
=> two symmetric half-stacks of 19 mm

NB: the length of the chamber is constrained
by the size of a preamp (which is ~37 mm)

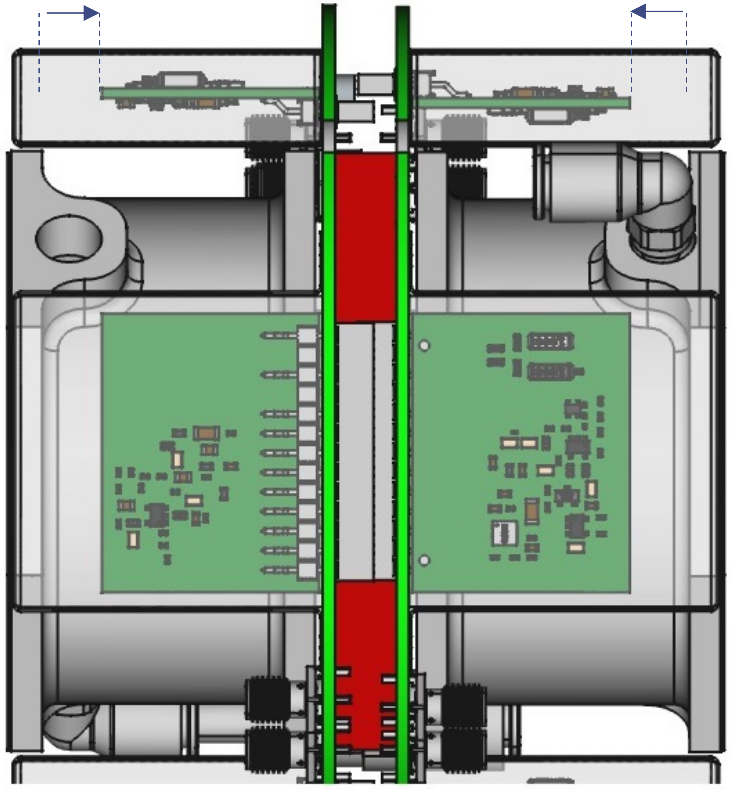


Fission chamber preliminary design





Compact configuration (-7 mm) with bottom connections for PA



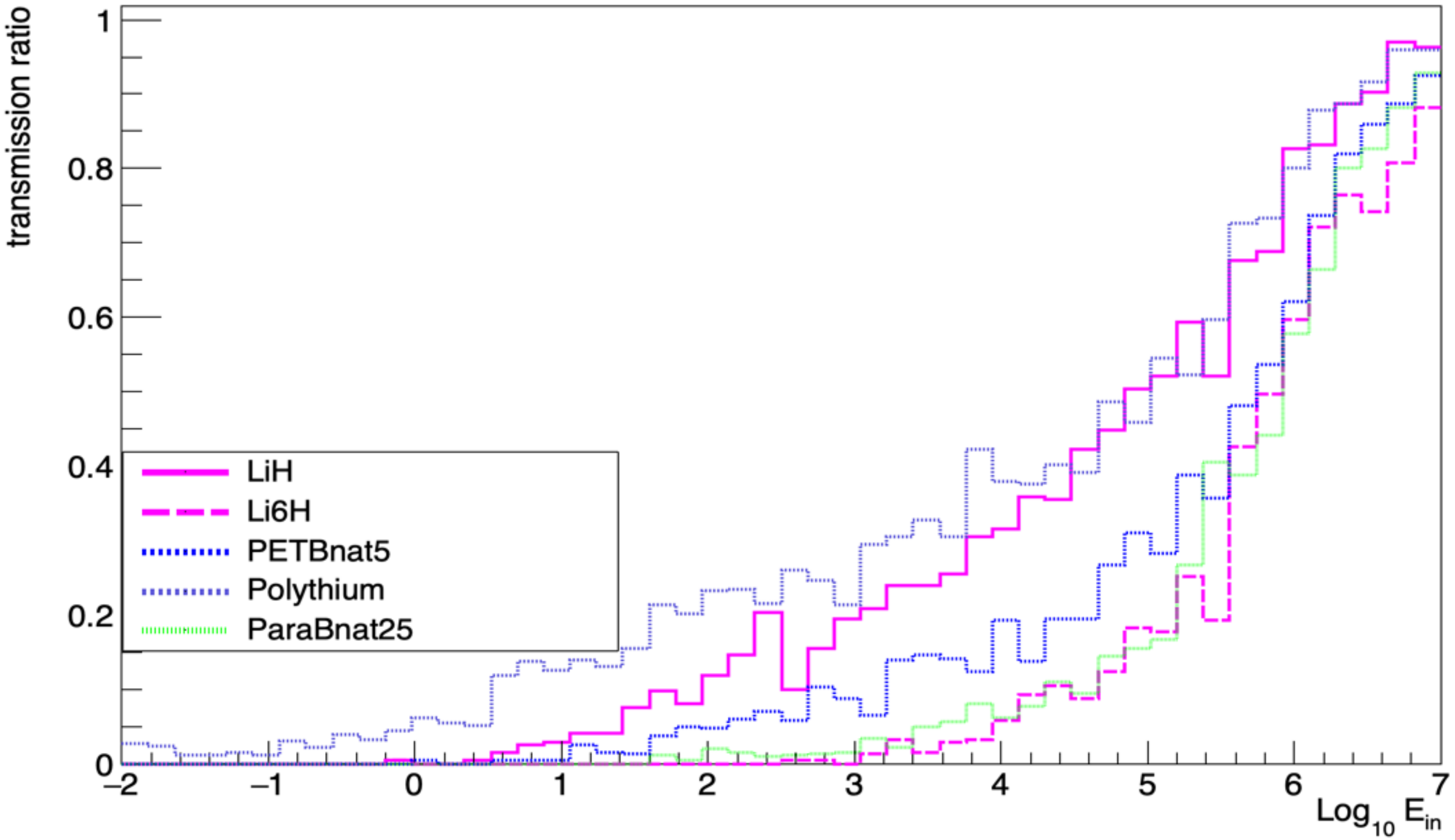
Absorber material preliminary study

The absorber of the TAC should:

- moderate/absorb all scattered (and fission) neutrons
- be transparent to gamma
- not produce any problematic secondary gammas

Typical materials:
LiH, Polyethylene (PE) with Li or B

Geant4 simulation shows that 4 cm B-PE with 5% B-nat is very efficient



3. Summary

Summary

Collaboration between DRF, DAM, DES, LP2i Bordeaux, IRSN (+JRC-Geel)

- Development of Tagging Fission Chambers for high counting rates
  - Compact FICH-U233 for TAC based on DAM design for high counting rate
  - More compact FICH-U238 and FICH-Pu239 for SCONE based on FICH-U233
  - FICH-Pu241 for TAC will be based on the latest developments

- Measurements making use of these developments
  - @n_TOF: ^{233}U in 2016 (and ^{241}Pu in 2025)
  - @NFS: ^{238}U in 2021 (and ^{239}Pu…)

Thank you for your attention