Cross section measurements of minor actinides a the n_TOF-Ph2 experiment at CERN

Daniel Cano Ott on behalf of the n_TOF collaboration

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Nuclear data for the transmutation of nuclear waste

Transmutation of the Minor Actinides by (n,f) and (n,γ) in new nuclear systems, thus reducing:

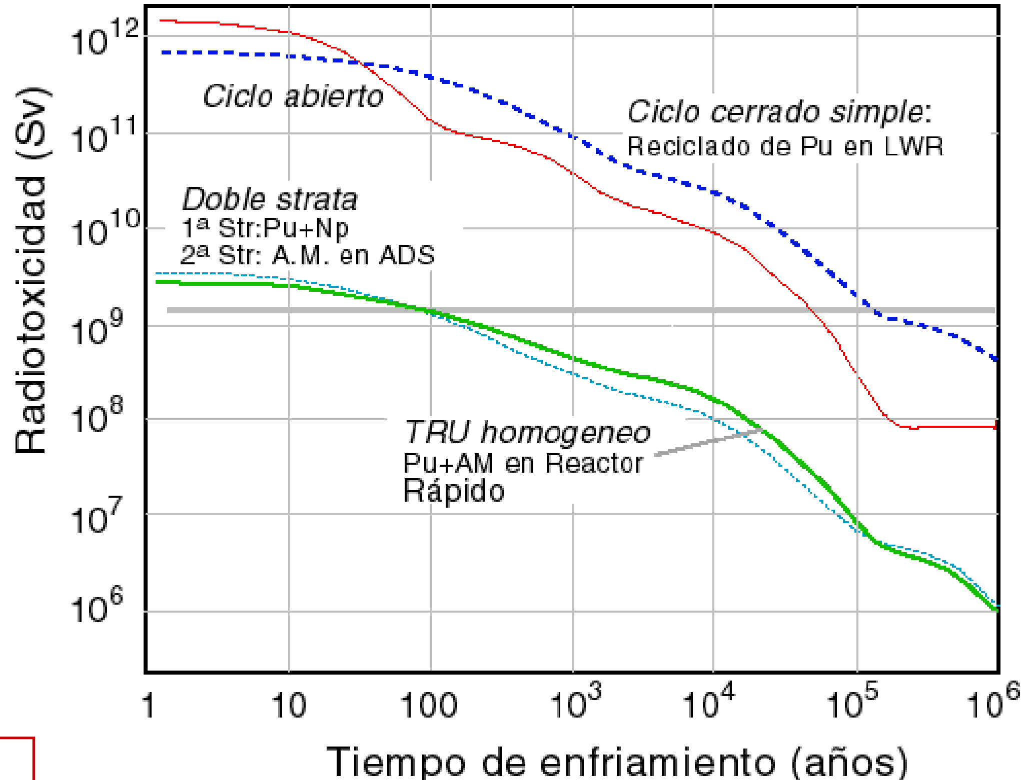
- radiotoxicity inventory 1/100
- cooling time 1/1000

↓

Computational design tools

↓

Need of accurate& reliable **NUCLEAR DATA**

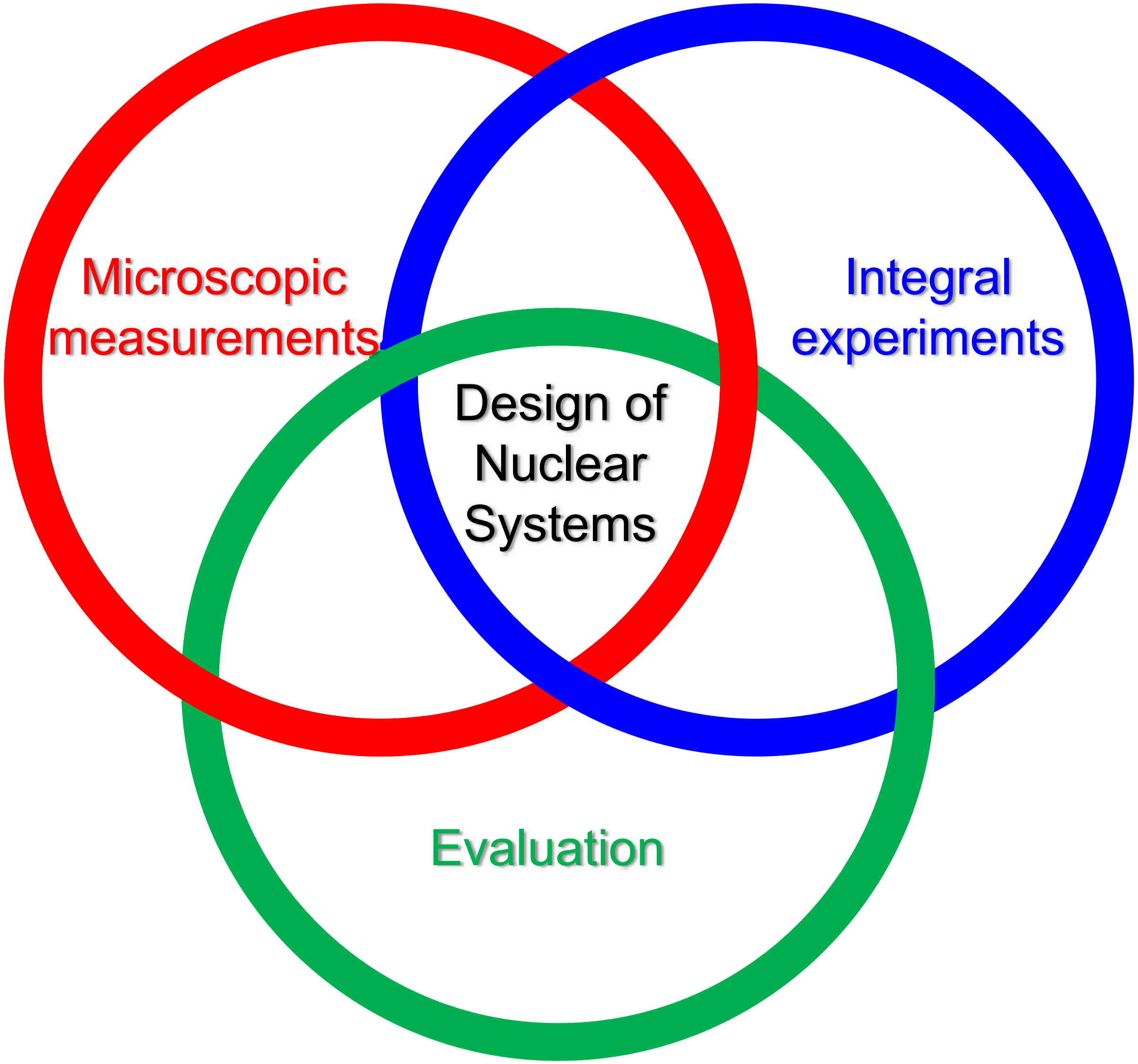


Nuclear data: compilation of experimental and/evaluated data describing nuclear properties.

- **Differential and average reaction cross sections**
- Particle emission probabilities and energies (γ, e-, α…)
- Nuclear structure data (half lives, isomers…)

Needs from various fields and applications:

- **Transmutation of Nuclear Waste**
- **Nuclear reactor design and safety assessment**
- Hadron therapy
- Dosimetry (space, aircrafts, linacs)
- Shielding design: accelerators, power plants, hospitals…


Microscopic
measurements
Integral
experiments
Design of
Nuclear
Systems
Evaluation

Are the nuclear data accurate enough?

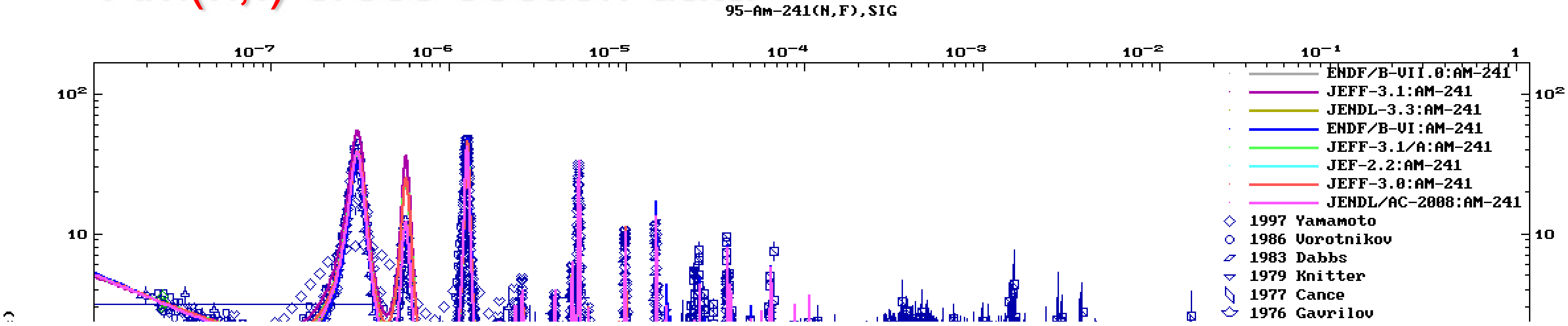
How accurately can we answer to the question:

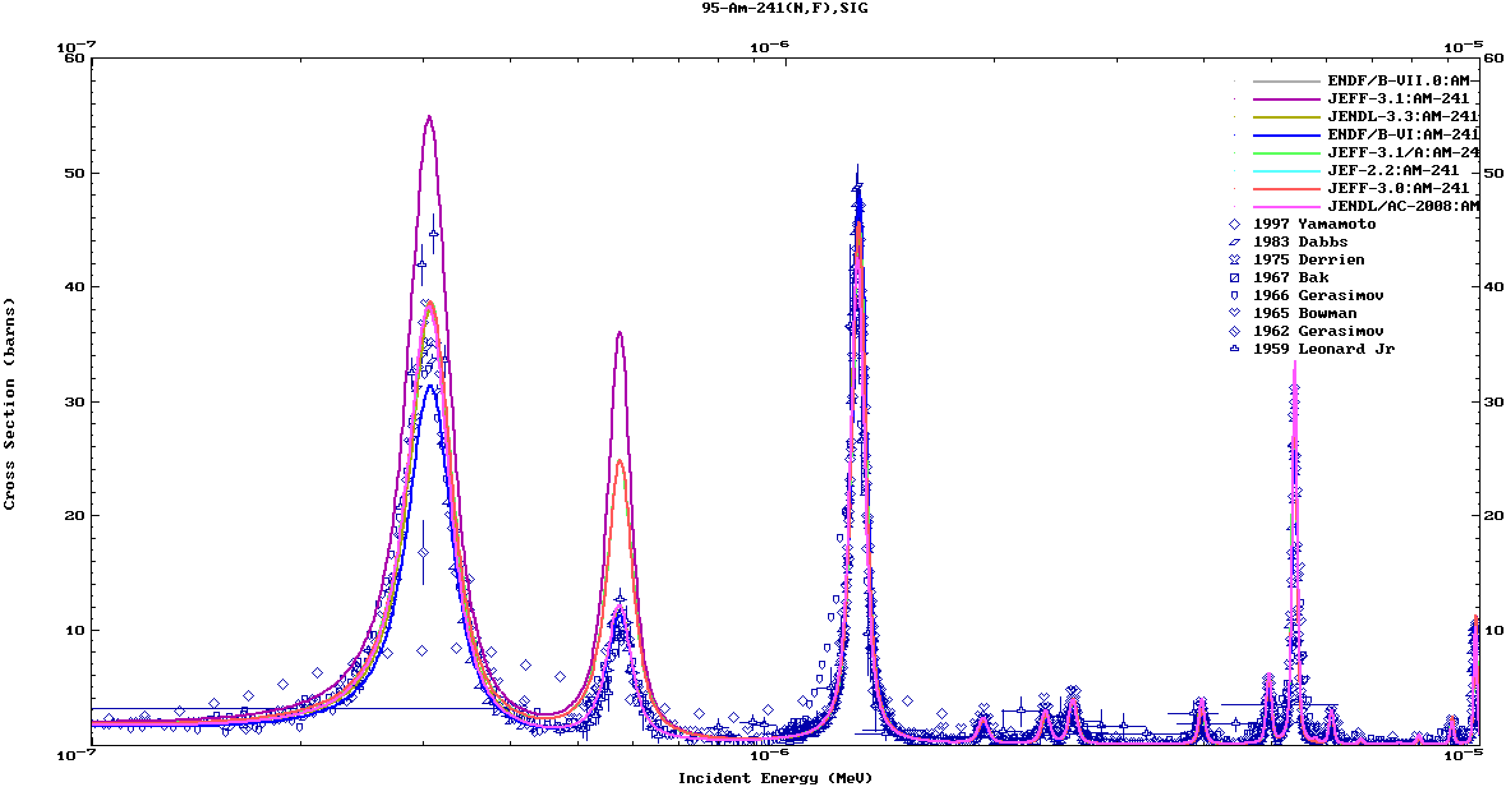
What is transmutation rate of ^{241,243}Am by fission inside an Accelerator Driven System or a fast reactor?

Lets have a look at the evaluated cross section databases:

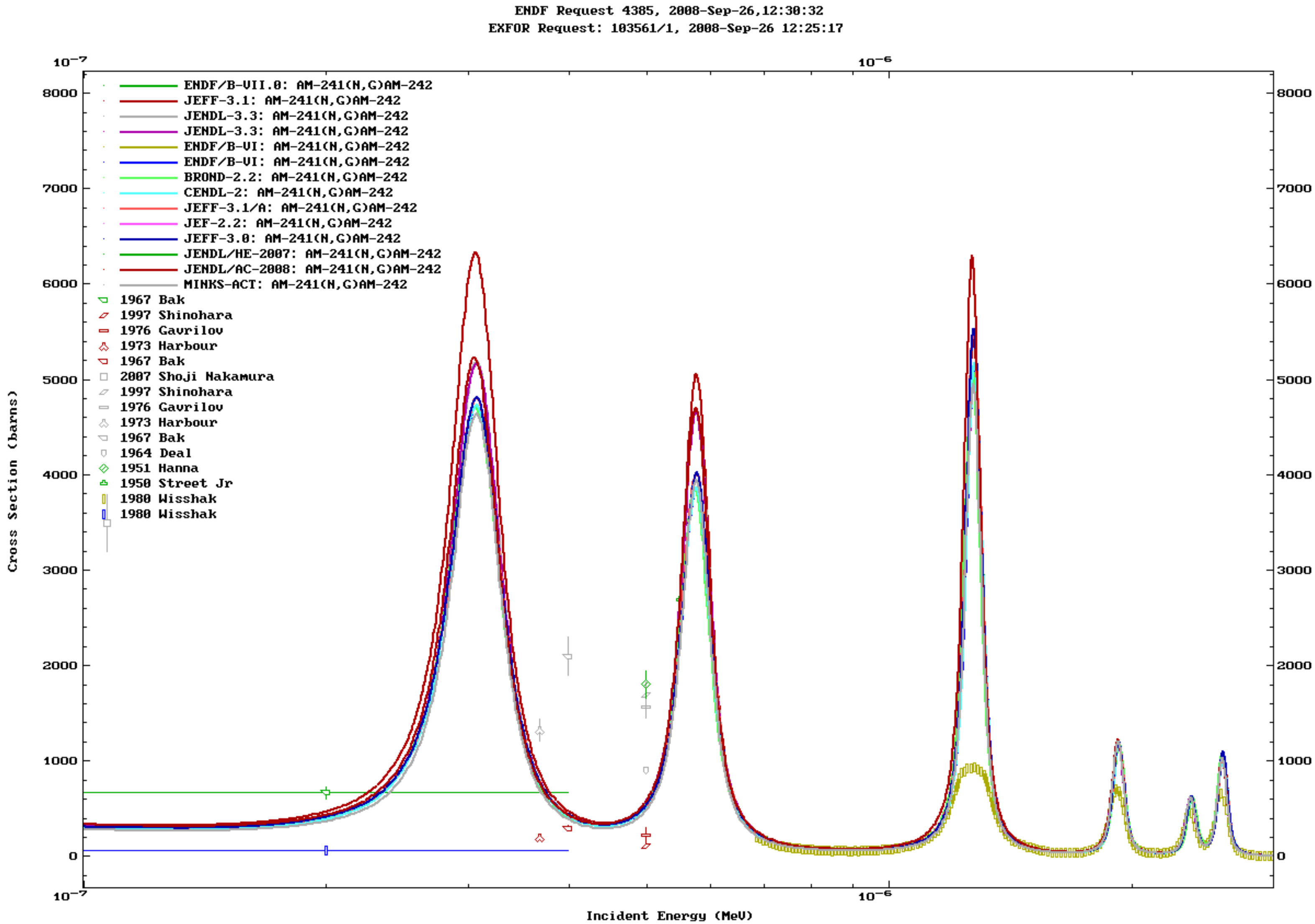
- ENDF
- JENDL
- JEFF
- BROND, CENDL,…

^{241}Am(n,f) cross section data

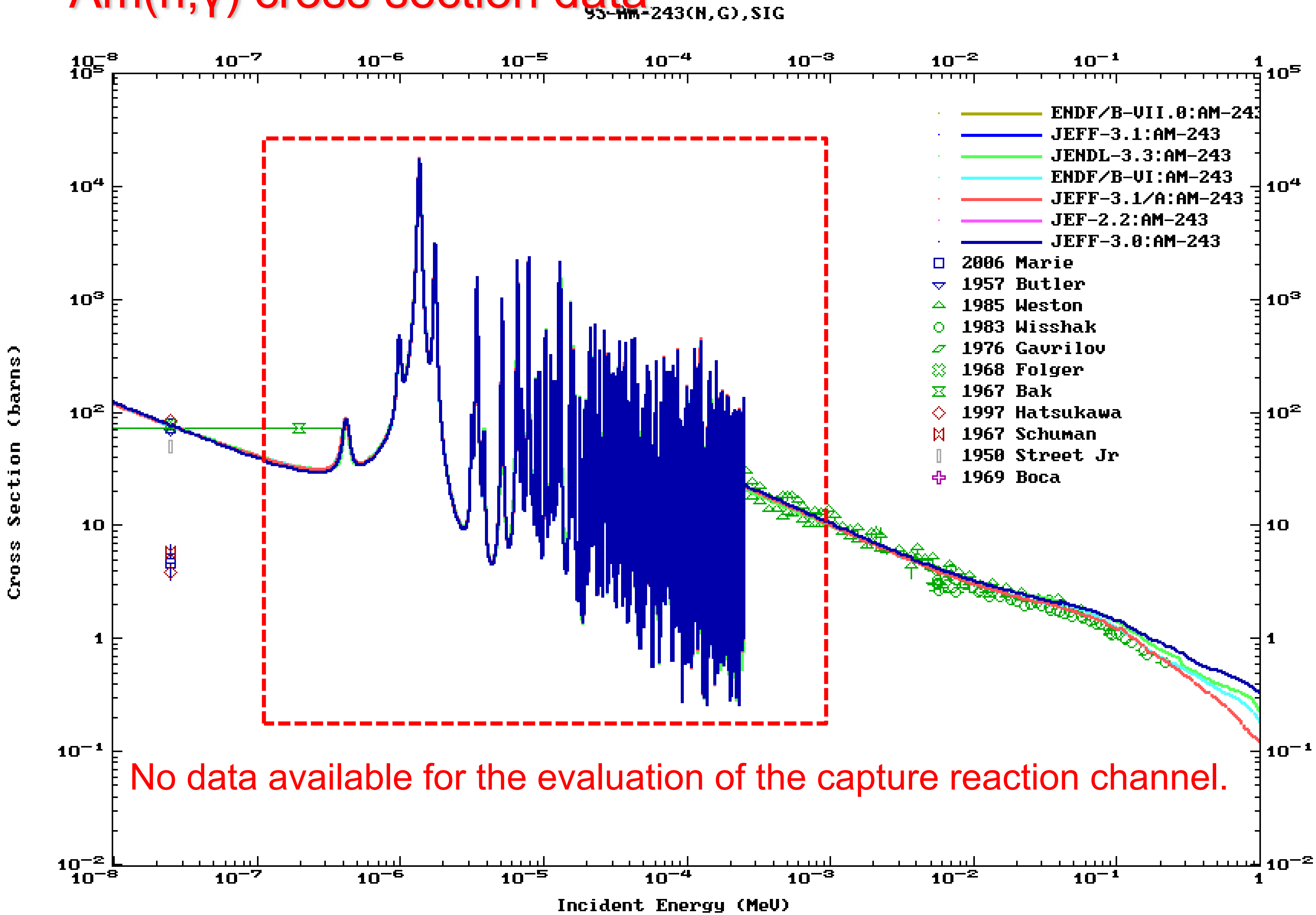




$^{241}Am(n,\gamma)$ cross section data



$^{243}Am(n,\gamma)$ cross section data



The reality about Minor Actinide cross sections

- In many cases there is insufficient accuracy (>10%).
- The systematic uncertainties are sometimes larger than indicated.
- There exist significant differences between evaluated data libraries.
- Data for some isotopes/reaction channels are missing.
- Scarce or non-existent covariance data, sometimes "guessed" in absence of experimental information.

Which isotopes, reaction channels and energy ranges are prioritary in the different scenarios?

Result of the available (and ongoing) sensitivity analyses and list of recommedations.

ABTR, SFR, EFR, GFR, LFR, ADS: Uncertainty Reduction Requirements Needed to Meet Integral Parameter Target Accuracies

Isotope	Cross-Section	Energy Range	Uncertainty (%) Initial	Uncertainty (%) Required
U238	σ_{capt}	24.8 - 9.12 keV	9.4	1.8
		9.12 - 2.03 keV	3.1	1.8
U238	σ_{inel}	19.6 - 6.07 MeV	29.3	9.0
		6.07 - 2.23 MeV	19.8	2.0
		2.23 - 1.35 MeV	20.6	2.1
		1.35 - 0.498 MeV	11.6	2.3
		498 - 183 keV	4.2	3.8
		183 - 67.4 keV	11.0	4.2
Pu239	σ_{capt}	1.35 - 0.498 MeV	18.2	6.6
		498 - 183 keV	11.6	4.4
		183 - 67.4 keV	9.0	4.0
		67.4 - 24.8 keV	10.1	4.2
		24.8 - 9.12 keV	7.4	3.8
		9.12 - 2.03 keV	15.5	3.2
Pu240	σ_{fiss}	6.07 - 2.23 MeV	4.8	2.9
		2.23 - 1.35 MeV	5.7	2.6
		1.35 - 0.498 MeV	5.8	1.6
		498 - 183 keV	3.9	3.7
		2.03 - 0.454 keV	21.6	11.8
Pu241	σ_{fiss}	6.07 - 2.23 MeV	14.2	5.0
		2.23 - 1.35 MeV	21.3	3.9
		1.35 - 0.498 MeV	16.6	2.1
		498 - 183 keV	13.5	1.7
		183 - 67.4 keV	19.9	1.7
		67.4 - 24.8 keV	8.7	1.9
		24.8 - 9.12 keV	11.3	2.0
		9.12 - 2.03 keV	10.4	2.1
		2.03 - 0.454 keV	12.7	2.7
		454 - 22.6 eV	19.4	5.4

Isotope	Cross-Section	Energy Range	Uncertainty (%) Initial	Uncertainty (%) Required
Am242m	σ_{fiss}	1.35 - 0.498 MeV	23.4	21.4
		498 - 183 keV	16.5	6.3
		183 - 67.4 keV	16.6	4.7
		67.4 - 24.8 keV	16.6	4.8
		24.8 - 9.12 keV	14.4	5.6
		2.04 - 0.454 keV	11.8	5.9
Am243	σ_{fiss}	6.07 - 2.23 MeV	11.0	2.3
		2.23 - 1.35 MeV	6.0	1.9
		1.35 - 0.498 MeV	9.2	1.7
Am243	σ_{inel}	6.07 - 2.23 MeV	17.9	4.9
		2.23 - 1.35 MeV	35.3	3.9
		1.35 - 0.498 MeV	42.2	2.3
		498 - 183 keV	41.0	3.7
		183 - 67.4 keV	79.5	3.7
		67.4 - 24.8 keV	80.8	12.4
Cm244	σ_{fiss}	6.07 - 2.23 MeV	31.3	3.0
		2.23 - 1.35 MeV	43.8	2.6
		1.35 - 0.498 MeV	50.0	1.5
		498 - 183 keV	36.5	4.0
		183 - 67.4 keV	47.6	7.3

2007 Symposium on Nuclear Data
November 29 - 30, 2007
RICOTTI Convention Center, Tokai, Ibaraki, Japan

by Aliberti, Oct. 2007

CANDIDE, 10 April 2008, Ai

List of new entries

ID	Target	Reaction	Quantity	Energy range		Acc	Sec.E/Angle
432	95-AM-241	(n,g),	(n,tot), SIG	thermal	eV	5%	
433	95-AM-243	(n,f)	n, spectrum	Eth	10 MeV	10%	
434	96-CM-244	(n,f)	n, spectrum	Eth	10 MeV	10%	
435	92-U-238	(n,n')	SIG	67.4 keV	19.6 MeV	2%	Emis spec.
436	94-PU-238	(n,f)	SIG	9.12 keV	6.07 MeV	3%	
437	94-PU-238	(n,f)	nu	67.4 keV	1.35 MeV	2.50%	
438	95-AM-241	(n,f)	SIG	183 keV	19.6 MeV	1.20%	
439	95-AM-242	(n,f)	SIG	9.12 keV	1.35 MeV	4.70%	
440	95-AM-243	(n,n')	SIG	24.8 keV	6.07 MeV	2.30%	Emis spec.
441	96-CM-242	(n,f)	SIG	67.4 keV	6.07 MeV	32%	
444	96-CM-244	(n,f)	SIG	67.4 keV	6.07 MeV	1.50%	
445	83-BI-209	(n,n')	SIG	0.498 MeV	2.23 MeV	2.80%	Emis spec.
446	96-CM-245	(n,f)	SIG	0.454 keV	6.07 MeV	2.90%	
447	5-B-10	(n,a)	SIG	0.498 MeV	2.23 MeV	2.70%	
448	11-NA-23	(n,n')	SIG	0.498 MeV	1.35 MeV	10.50%	Emis spec.
449	6-C-12	(n,g)	SIG	0.54 eV	4 eV	5%	
450	6-C-12	(n,g)	SIG	6.07 MeV	19.6 MeV	7.10%	
451	94-PU-239	(n,g)	SIG	0.1 eV	0.54 eV	0.90%	
452	94-PU-241	(n,g)	SIG	0.1 eV	0.54 eV	2.40%	
453	8-O-16	(n,g)	SIG	2.23 MeV	19.6 MeV	9.90%	
454	26-FE-56	(n,n')	SIG	0.498 MeV	19.6 MeV	1.50%	Emis spec.
455	94-PU-241	(n,f)	SIG	Thermal	6.07 MeV	1.50%	

HPRL-JEFDOC1235

Where can these cross sections be measured?

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Available neutron time of flight facilities worldwide

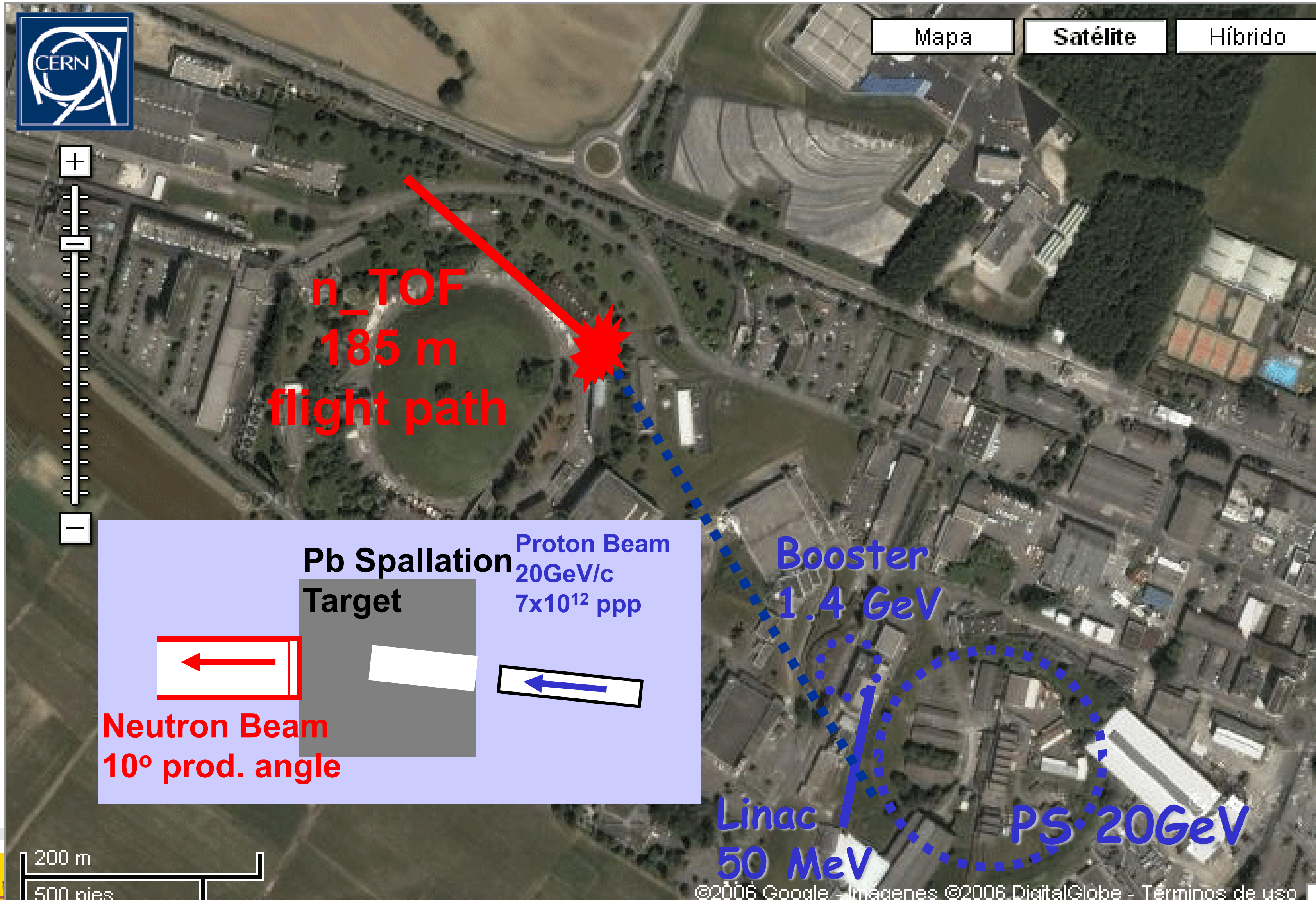
I. Photoproduction neutron sources
(GELINA – Europe, ORELA & RPI – USA) + reactors (KURRI - Japan…):

- Good energy resolution.
- Low intensity per accelerator pulse / high repetition rate -> **low duty cycle**.
- **Need of "massive" samples (several 100 mg) -> STABLE ISOTOPES**

II. Spallation neutron sources
(LANSCE-LANL - USA and n_TOF @ CERN - Europe)

- Intrinsically worse energy resolution.
- High intensity at even low repetition rates -> **high duty cycle, favorable reaction rate to decay rate ratios, even for high intrinsic activities (1 GBq).**
- **Samples can have masses as low as 1 mg.**

(A Google-view of) The n_TOF facility at CERN



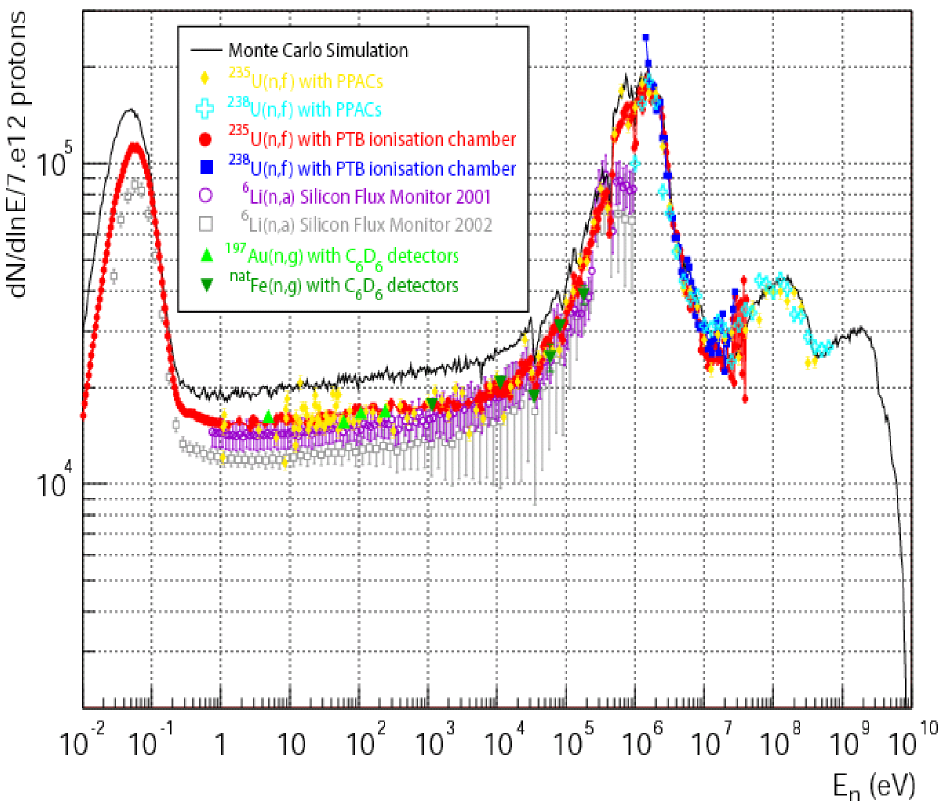
The n_TOF collaboration

U.Abbondanno[14], G.Aerts[7], H.Álvarez[24], F.Alvarez-Velarde[20], S.Andriamonje[7], J.Andrzejewski[33], P.Assimakopoulos[9], L.Audouin[5], G.Badurek[1], P.Baumann[6], F. Bečvář [31], J.Benlliure[24], E.Berthoumieux[7], F.Calviño[25],D.Cano-Ott[20],R.Capote[23],A.Carrillo de Albornoz[30],P.Cennini[4], V.Chepel1[7], E.Chiaveri[4], N.Colonna1[3], G.Cortes[25], D.Cortina[24], A.Couture[29], J.Cox[29], S.David[5], R.Dolfini[15], C.Domingo-Pardo[21], W.Dridi[7], I.Duran[24], M.Embid-Segura[20], L.Ferrant[5], A.Ferrari[4], R.Ferreira-Marques[17], L.Fitzpatrick[4], H.Frais-Koelbl[3], K.Fujii[13], W.Furman[18], C.Guerrero[20], I.Goncalves[30], R.Gallino[36], E.Gonzalez-Romero[20], A.Goverdovski[19], F.Gramegna[12], E.Griesmayer[3], F.Gunsing[7], B.Haas[32], R.Haight[27], M.Heil[8], A.Herrera-Martinez[4], M.Igashira[37], S.Isaev[5], E.Jericha[1], Y.Kadi[4], F.Käppeler[8], D.Karamanis[9], D.Karadimos[9], M.Kerveno[6], V.Ketlerov[19], P.Koehler[28], V.Konovalov[18], E.Kossionides[39], M.Krtička[31], C.Lamboudis[10], H.Leeb[1], A.Lindote[17], I.Lopes[17], M.Lozano[23], S.Lukic[6], J.Marganiec[33], L.Marques[30], S.Marrone[13], P.Mastinu[12], A.Mengoni[4], P.M.Milazzo[14], C.Moreau[14], M.Mosconi[8], F.Neves[17], H.Oberhummer[1], S.O'Brien[29], M.Oshima[38], J.Pancin[7], C.Papachristodoulou[9], C.Papadopoulos[40], C.Paradela[24], N.Patronis[9], A.Pavlik[2], P.Pavlopoulos[34], L.Perrot[7], R.Plag[8], A.Plompen[16], A.Plukis[7], A.Poch[25], C.Pretel[25], J.Quesada[23], T.Rauscher[26], R.Reifarth[27], M.Rosetti1[1], C.Rubbia1[5], G.Rudolf[6], P.Rullhusen[16], J.Salgado[30], L.Sarchiapone[4], C.Stephan[5], G.Tagliente[13], J.L.Tain[21], L.Tassan-Got[5], L.Tavora[30], R.Terlizzi[13], G.Vannini[35], P.Vaz[30], A.Ventura[11], D.Villamarin[20], M.C.Vincente[20], V.Vlachoudis[4], R.Vlastou[40], F.Voss[8], H.Wendler[4], M.Wiescher[29], K.Wisshak[8]

Funded by the EC 5th Framework programme, CERN and National Funding Agencies.

n_TOF beam characteristics

The n_TOF facility was built and commissioned in a period of 2 years and provides unique features for measuring capture and fission cross sections of unstable (and also stable) isotpes.

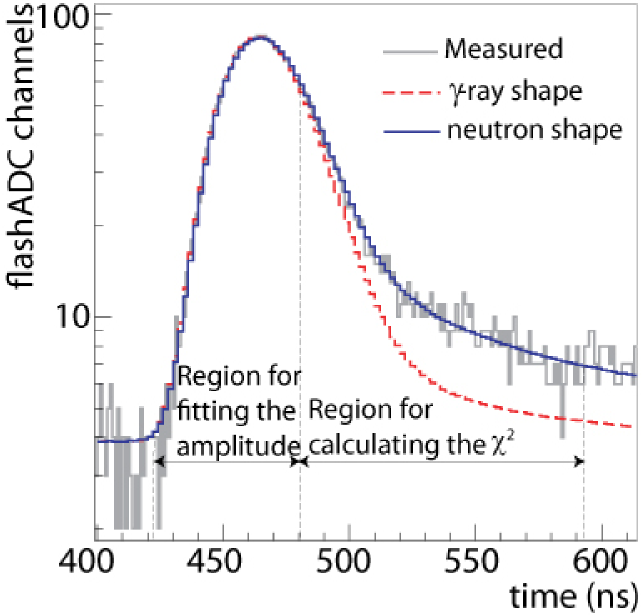


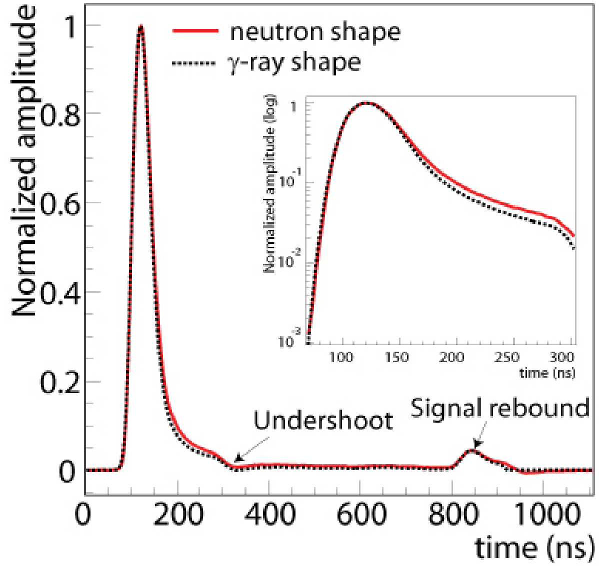
•White neutron beam: 0.1 eV up to 1 GeV
•$\Delta E/E \sim 1 \cdot 10^{-4}$ in the RRR

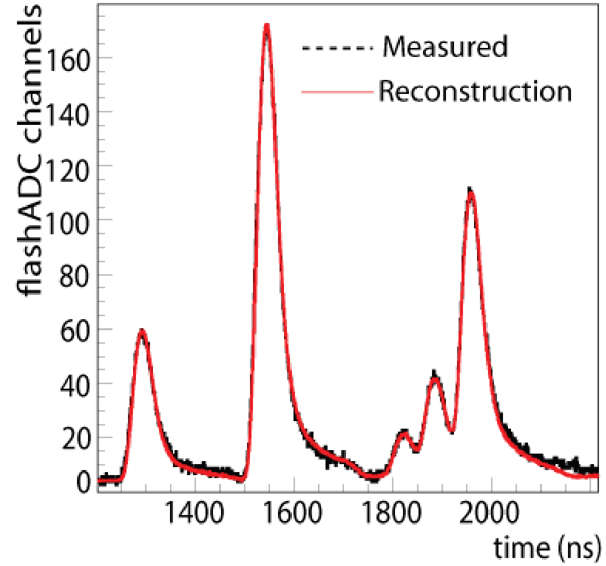
Capture and fission beams:
•Φ=4 cm beam
•Φ=8 cm beam

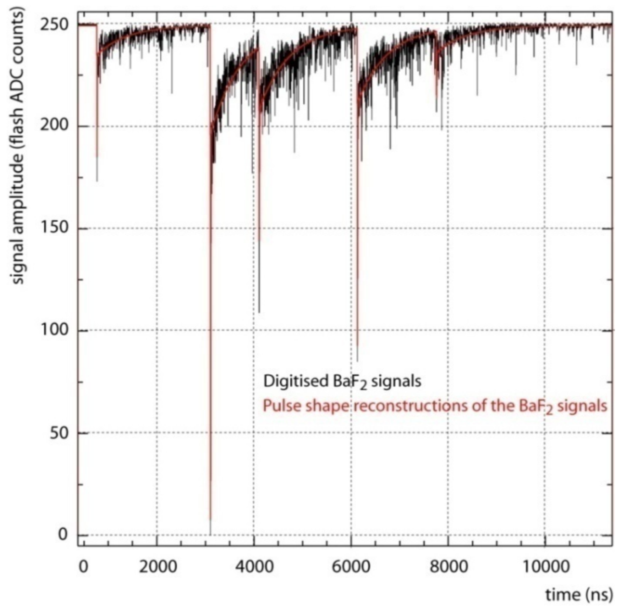
Performance Report, CERN-INTC-2002-037, January 2003,CERN-SL-2002-053 ECT

<u>A fully digital DAQ!</u>









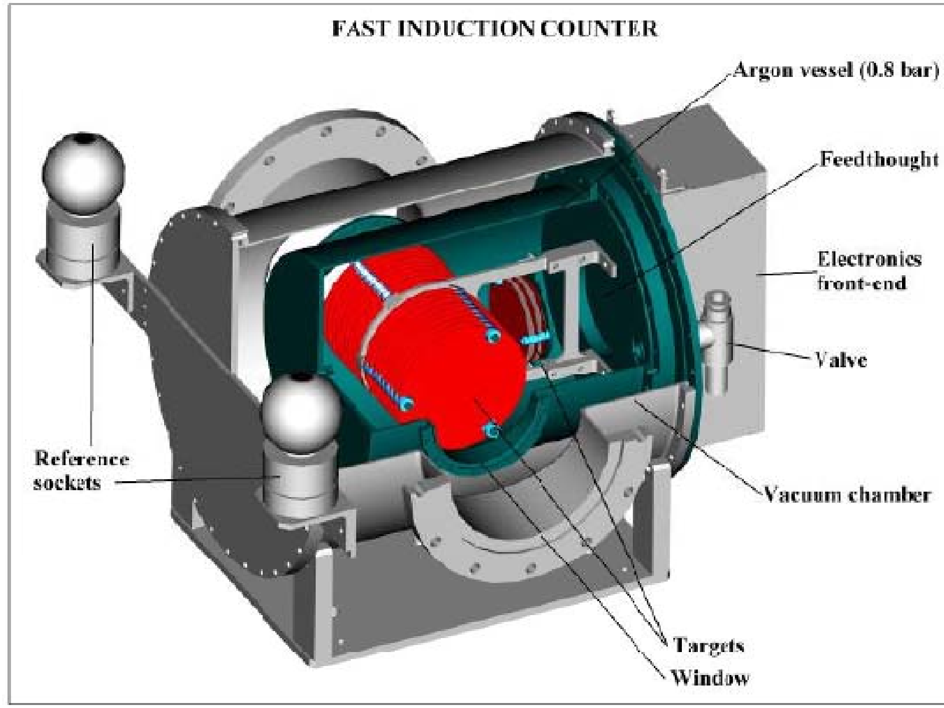
Pulse shape fitting, particle discrimination and pileup reconstruction for C_6D_6 detectors. C. Guerrero et al, submitted to NIM-A.

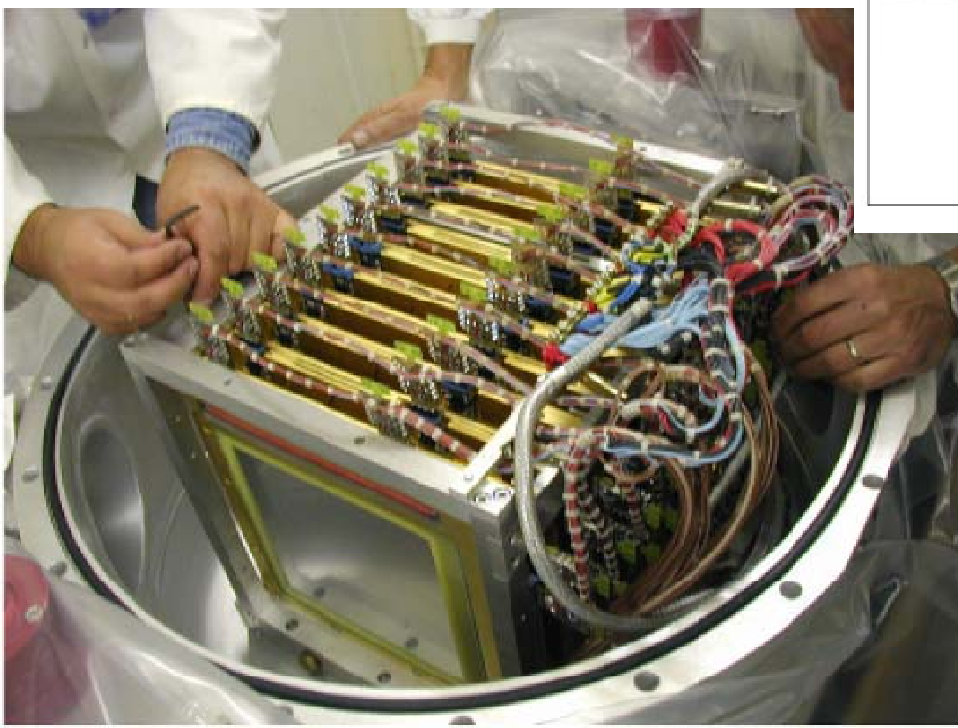
Pulse shape analysis and pileup reconstruction for the BaF_2 detectors. E. Berthomiueux et al. To be submitted to NIM-A.

Advanced detectors

n_TOF fission detectors

Position sensitive Parallel Plate Avalanche Chambers. Allow to reconstruct the trajectories of the fission fragments.
Tassan-Got et al. To be submitted to NIM-A

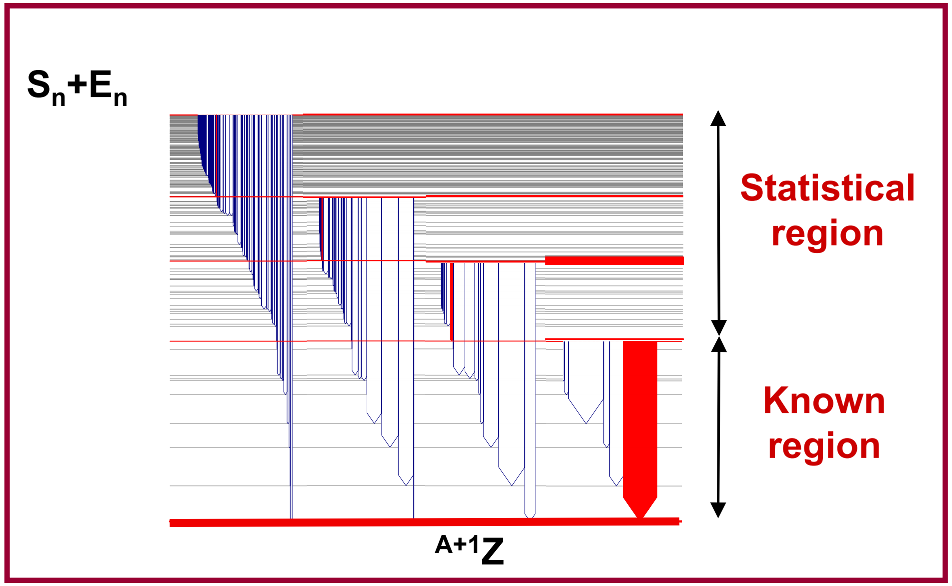


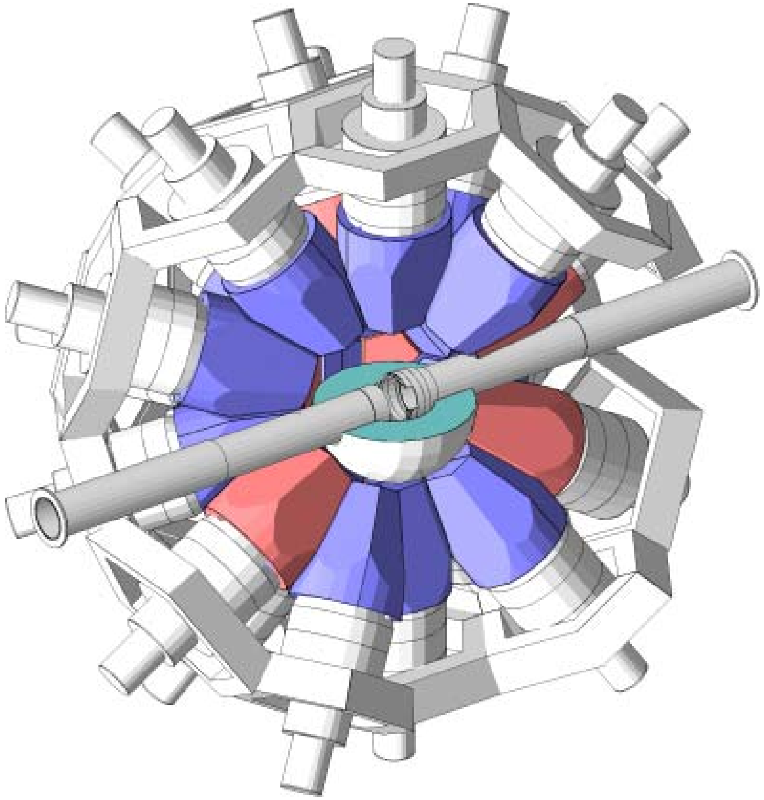


Fast induction chamber (FIC). Large number of samples inside a compact detector.
P. Cennini et al. Submitted to NIM-A

Nuclear structure: TAC as a γ-ray spectrometer

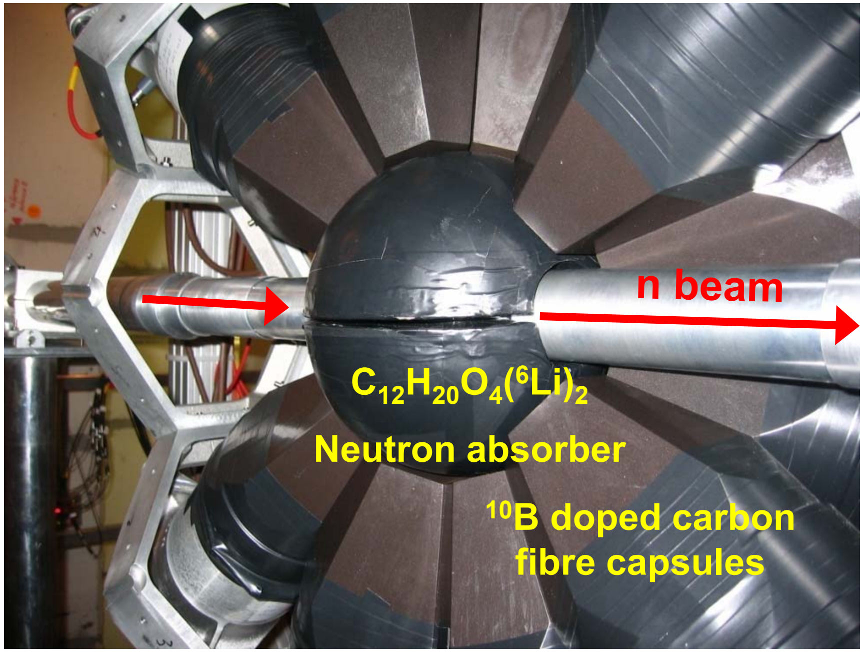
Analysis of the calorimeter data and comparison to realistic Monte Carlo simulations of its response to the EM cascades.

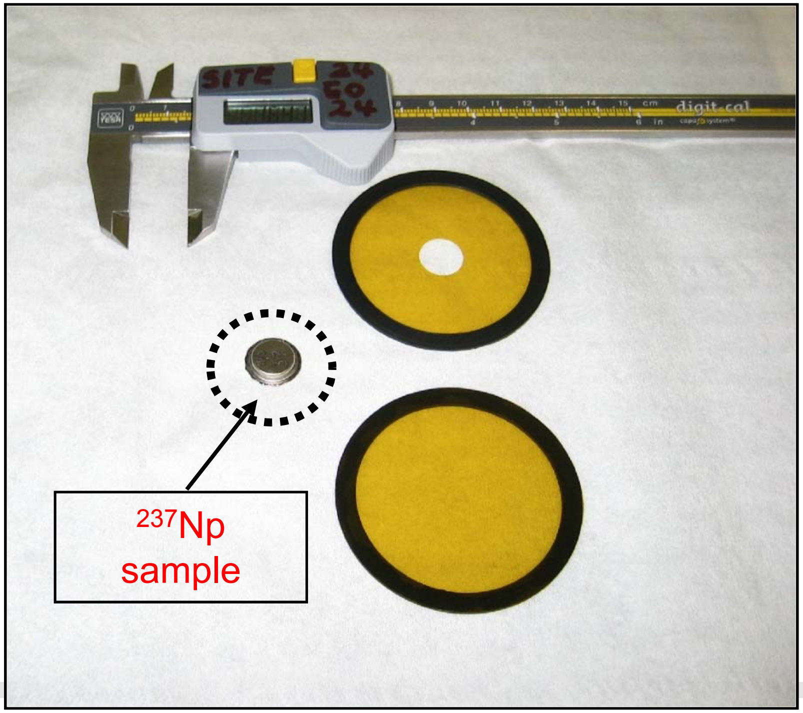




The n_TOF Total Absorption Calorimeter (TAC) for (n,γ) measurements

- 40 BaF_2 crystals covering 95% of 4π.
- 98% detection efficiency for capture γ-ray cascades.





The experimental programme

n_TOF experiments 2002-4

Capture

^{151}Sm

$^{204,206,207,208}Pb$, ^{209}Bi

^{232}Th

$^{24,25,26}Mg$

$^{90,91,92,94,96}Zr$, ^{93}Zr

^{139}La

$^{186,187,188}Os$

$^{233,234}U$

^{237}Np, ^{240}Pu, ^{243}Am

Fission

$^{233,234,235,236,238}U$

^{232}Th

^{209}Bi

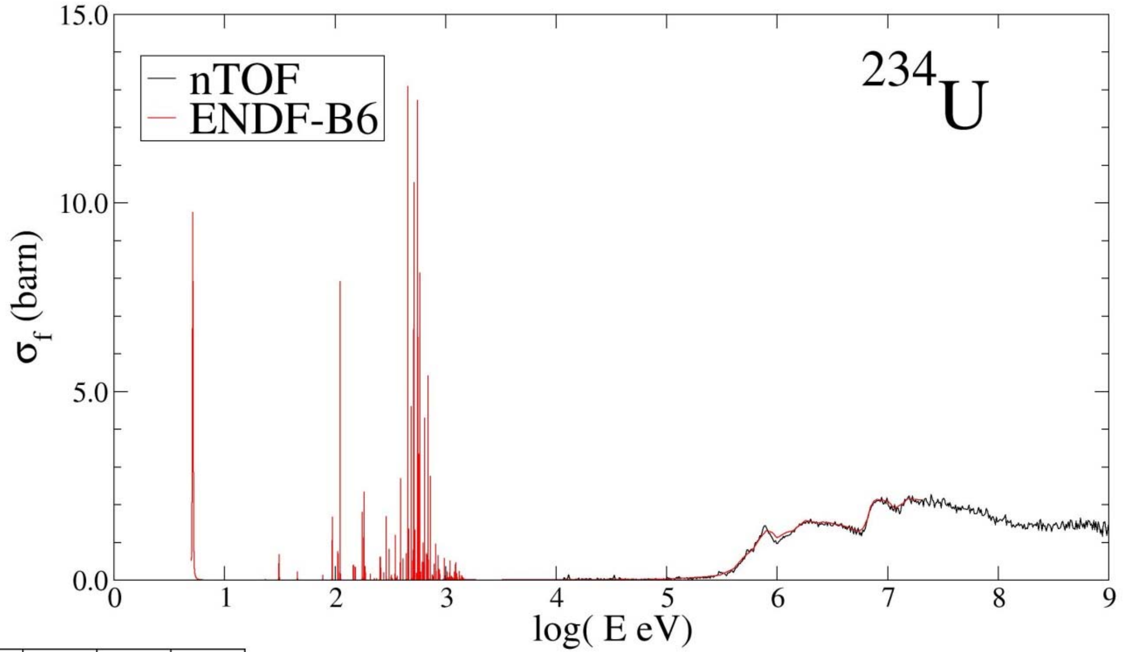
^{237}Np

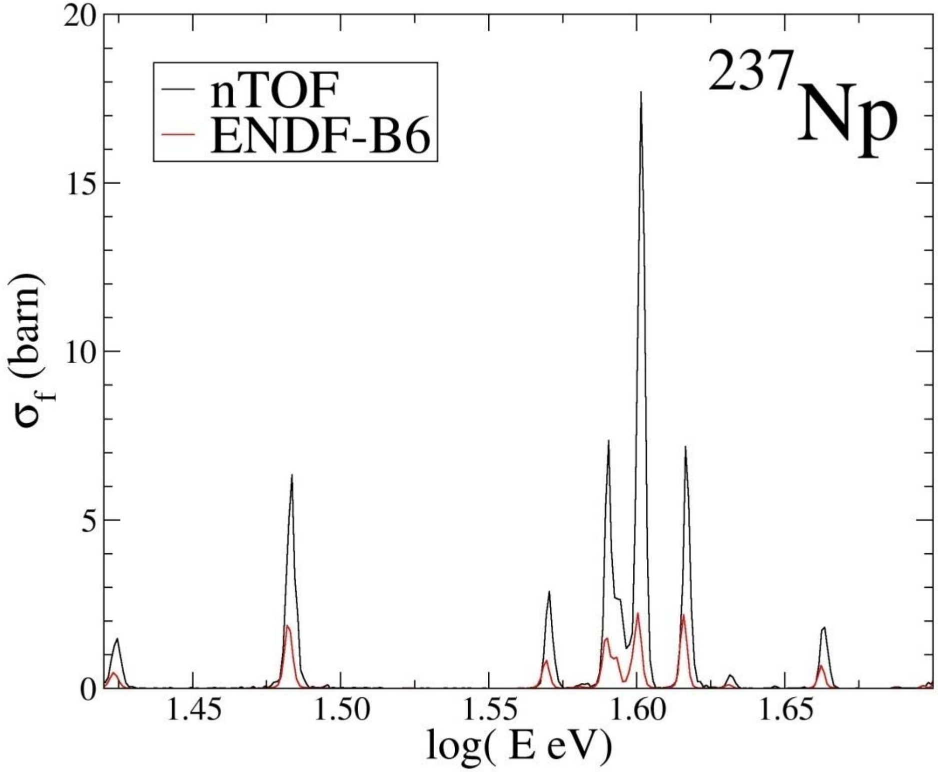
$^{241,243}Am$, ^{245}Cm

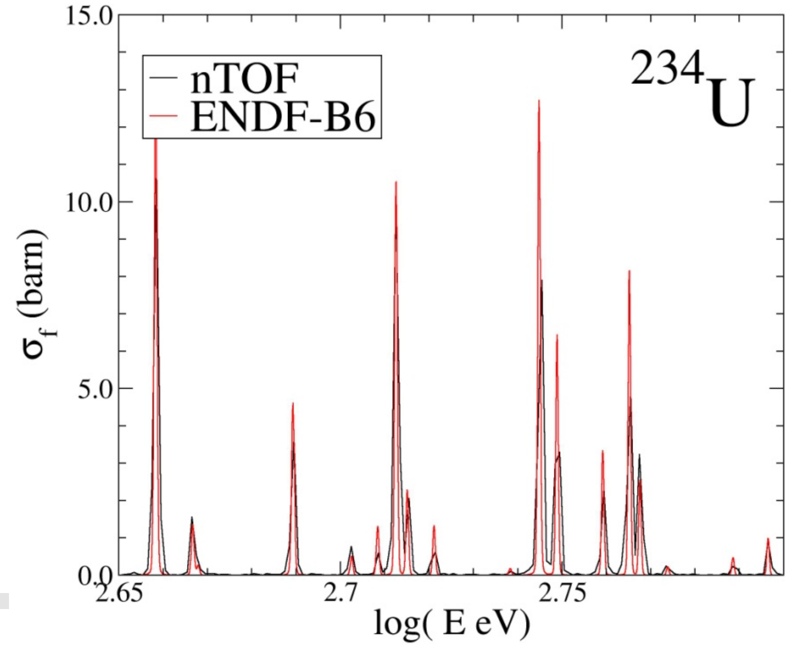
- **Measurements of neutron cross sections relevant for Nuclear Waste Transmutation and related Nuclear Technologies**
  - Th/U fuel cycle (capture & fission)
  - Transmutation of MA (capture & fission)
  - Transmutation of FP (capture)
- **Cross sections relevant for Nuclear Astrophysics**
  - s-process: branchings
  - s-process: presolar grains
- **Neutrons as probes for fundamental Nuclear Physics**
  - Nuclear level density & n-nucleus interaction

(n,f) cross sections

Measurements with PPACs.
L. Tassan-Got et al. In preparation.
C. Paradela et al. In preparation





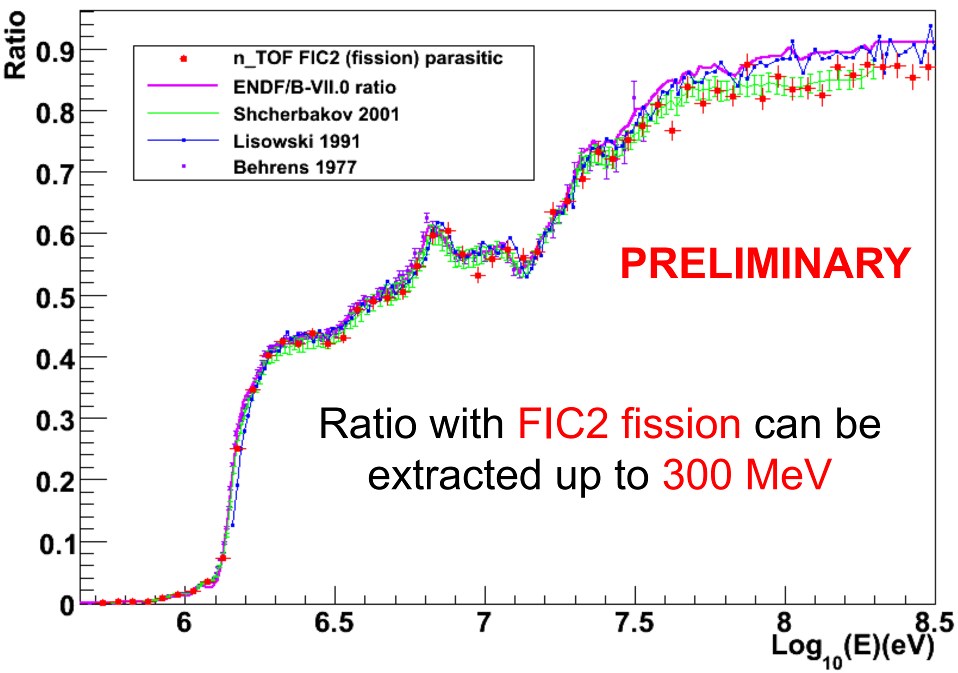


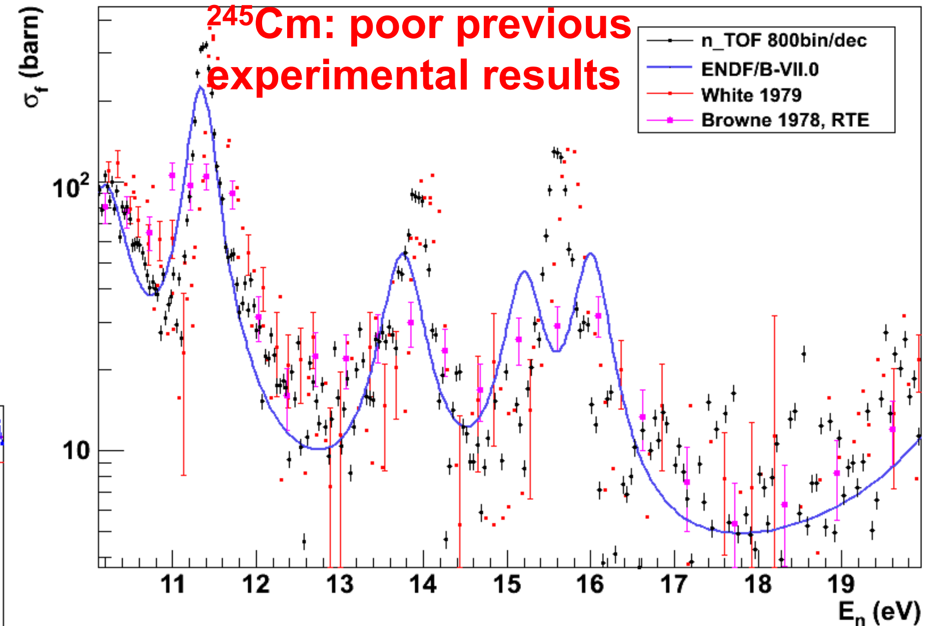
Fission measurements with the Fission Ionization Chamber (FIC) – 2/2

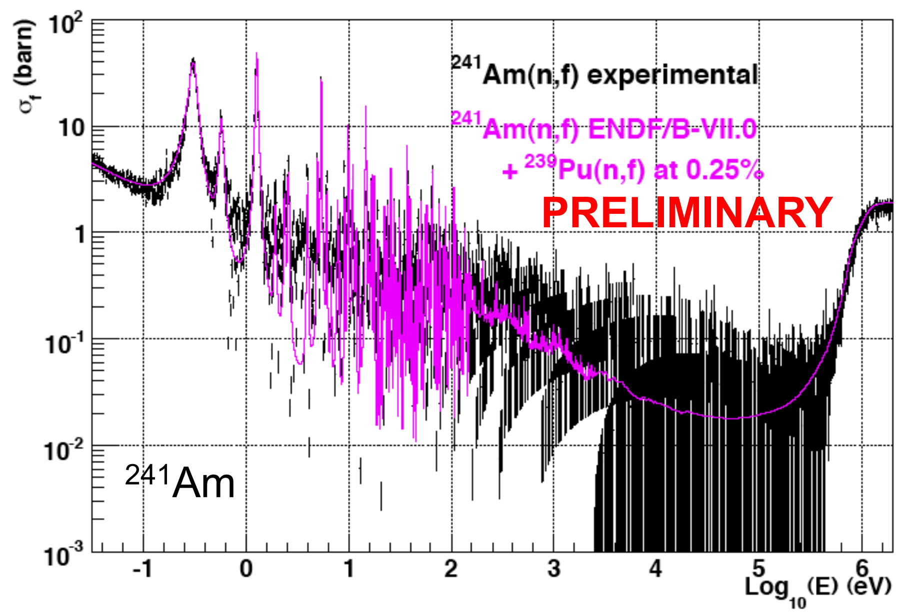
Statistical error bars

Sistematic errors ~8%

$^{238}U/^{235}U$: both isotopes are fission standards up to 200 MeV.



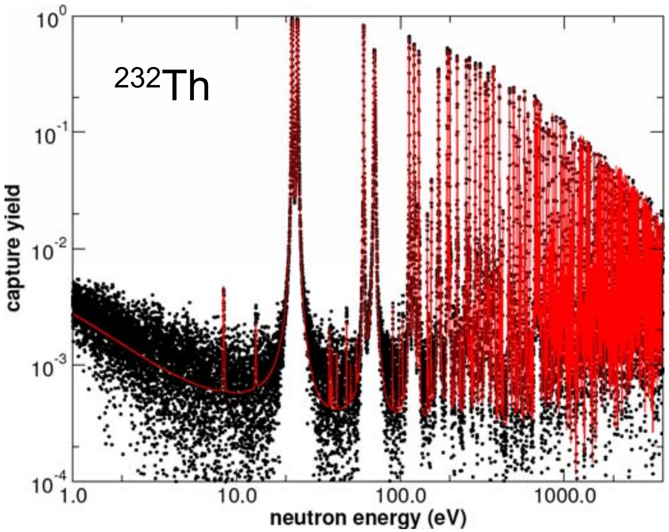




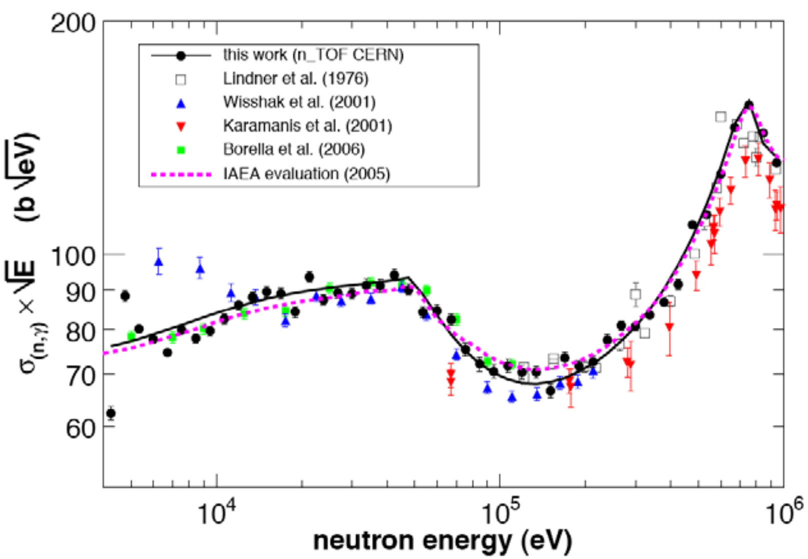
^{241}Am has ^{239}Pu contaminant which significantly contributes to the total σ.

(n,γ) cross section measurements with C_6D_6 detectors

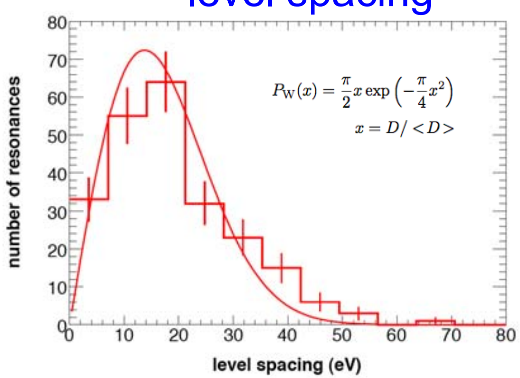
Resolved Resonance Region



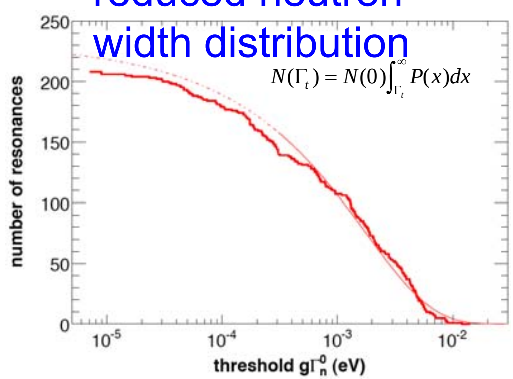
Unresolved Resonance Region



level spacing



reduced neutron width distribution



mass (^{232}Th) = 2.8037 g

diameter = 1.5 cm

purity = 99.5%

F.Gunsing, Nuc. Data. Conf.-2007
G. Aerts et al. (n_TOF Collaboration), Phys. Rev. C 73, 054610 (2006)

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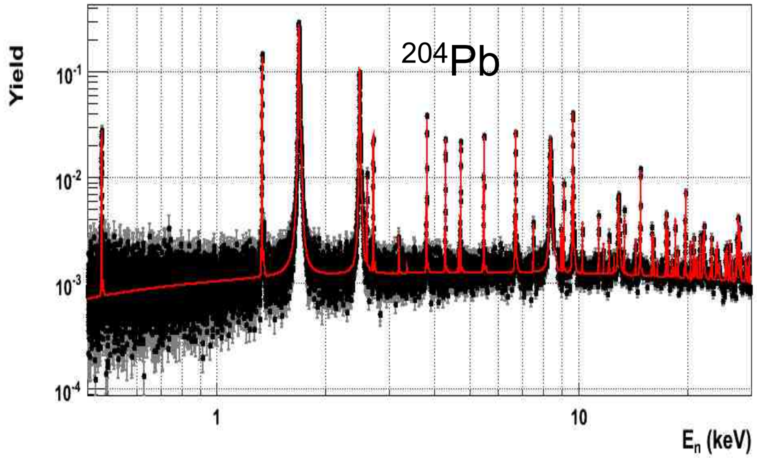
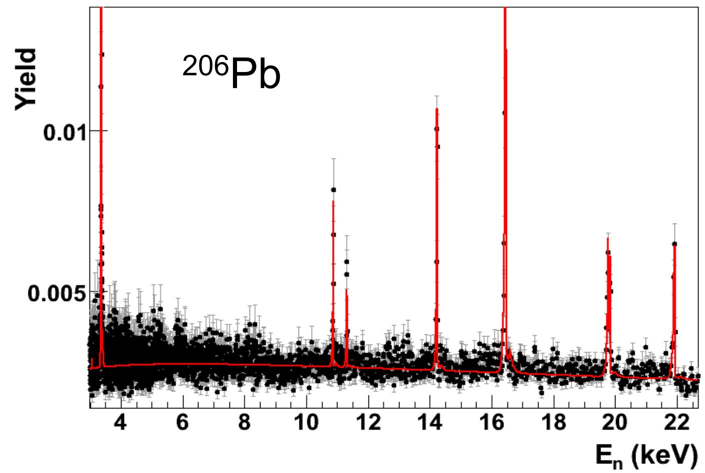
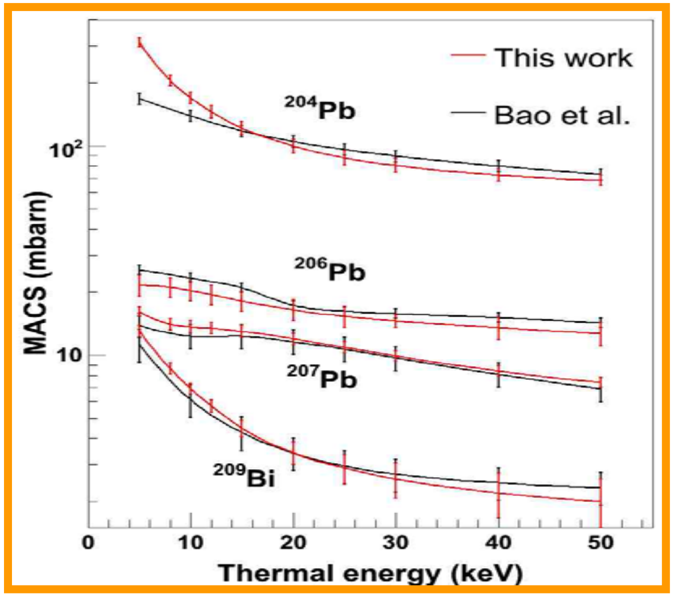
$^{204,206,207}Pb$, ^{209}Bi (n,γ) measurement



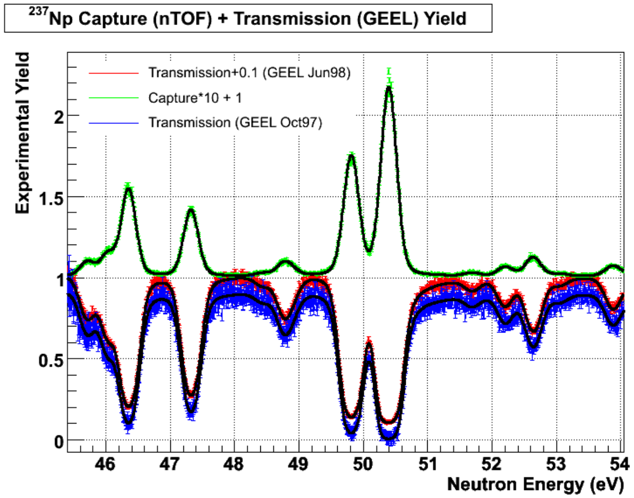


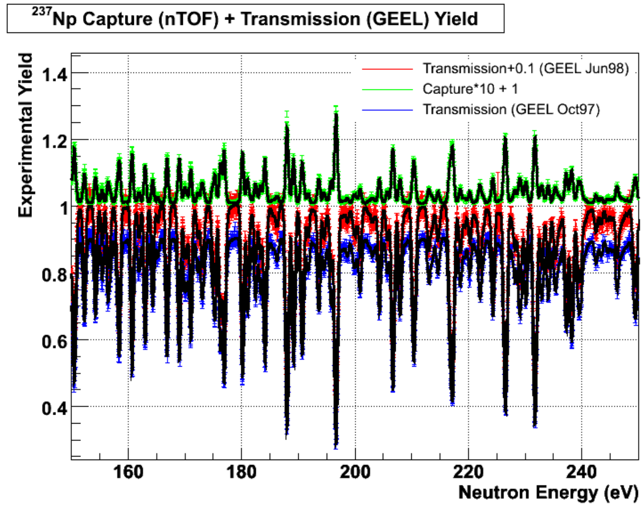


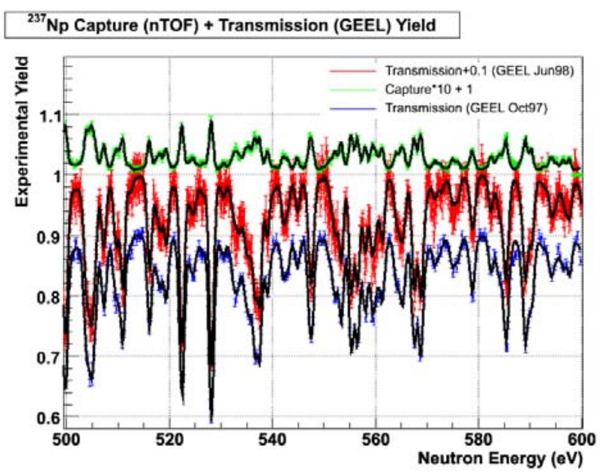
C.Domingo-Pardo et al. (n_TOF Collaboration), Phys. Rev. C 74/75, 2006/7

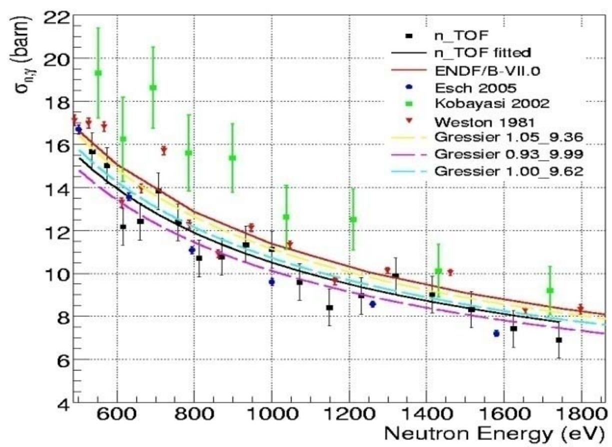
^{237}Np (n,γ) measurement with the TAC

43.3 mg, 1.29 MBq





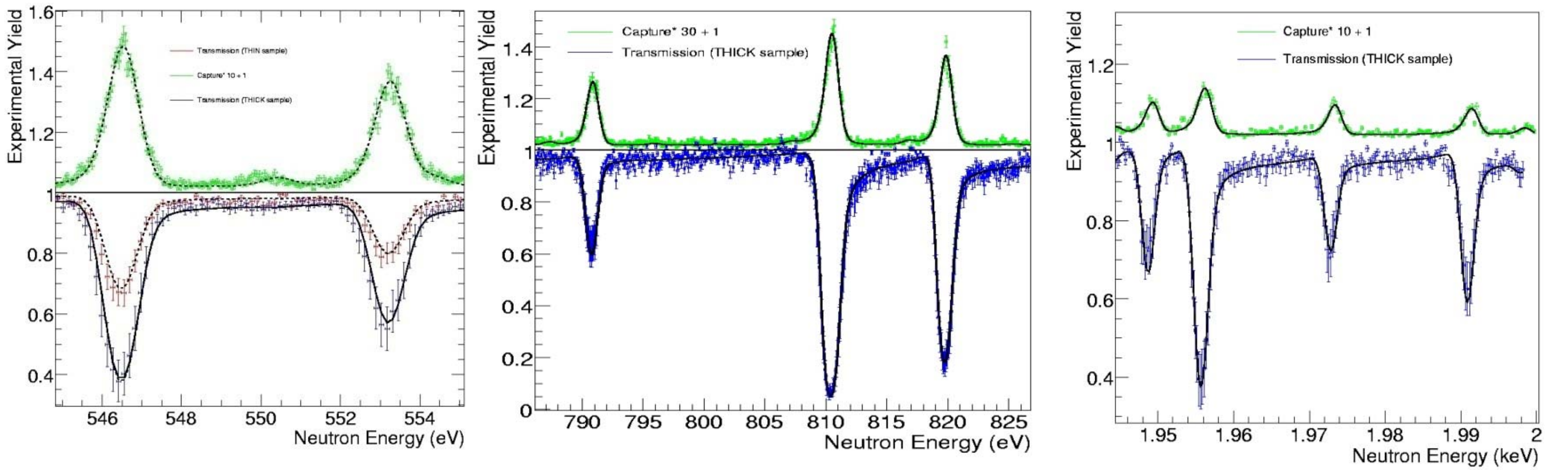




C. Guerrero et al. (n_TOF Collaboration), Proc. Int. Conf. Nuc. Data for Sci. and Tech. 2007, Nice.

^{240}Pu (n,γ) measurement

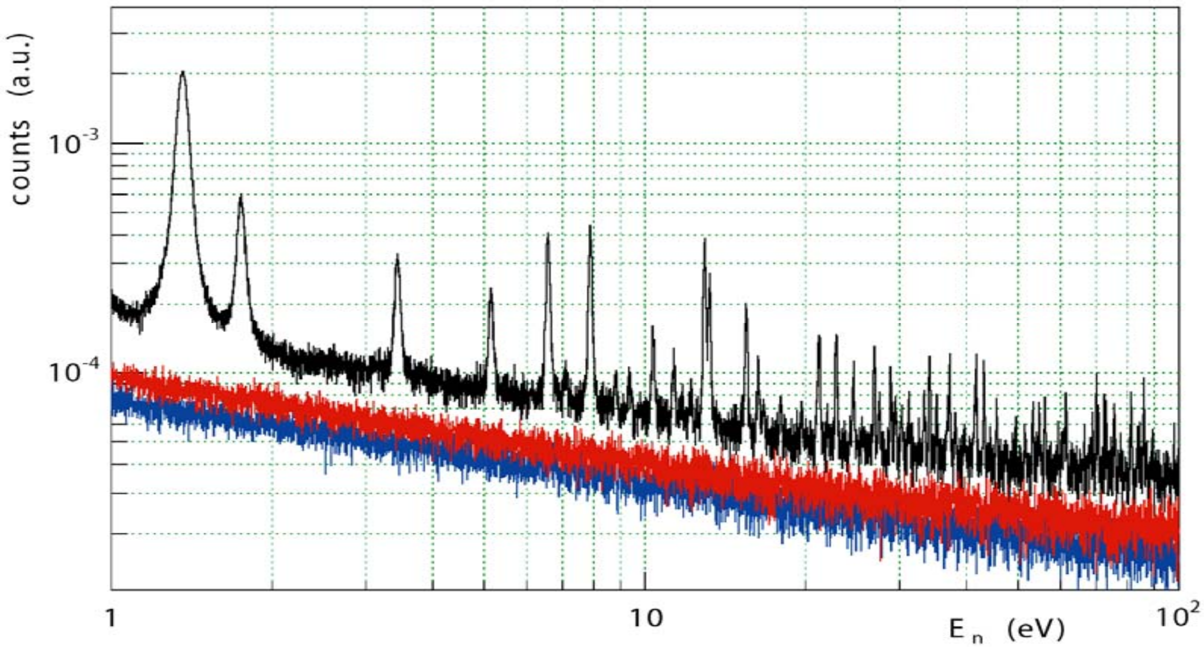
51.2 mg, 458 MBq



	ENDF/B-VII	n_TOF	Bouland et al.
‹D_0› (eV)	12.1	12.1 0.1	12.06
‹Γ_γ›(meV)	30.6	32.4 0.8	31.9
$S_0(10^{-4})$	1.13	1.04 0.08	1.07

C. Guerrero et al. (n_TOF Collaboration), Proc. Int. Conf. Nuc. Data for Sci. and Tech. 2007, Nice.

^{243}Am (n,γ) measurement with the TAC



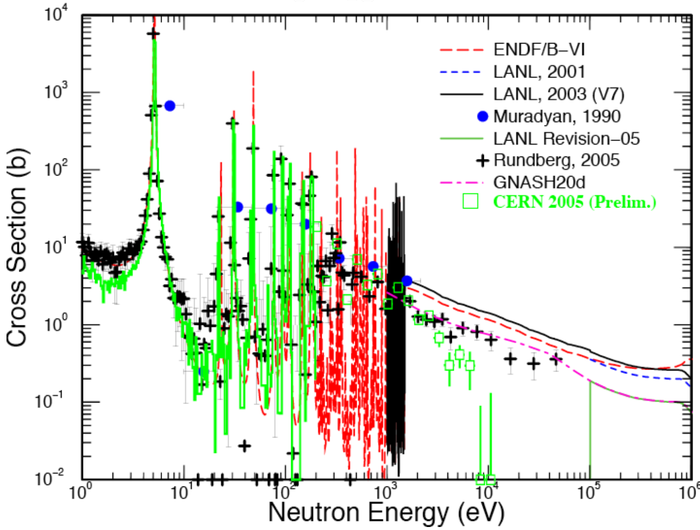
First (n,γ) measurement EVER. 10 mg, 75 MBq

D. Cano-Ott et al. (n_TOF Collaboration), Proc. Int. Conf. Capture Gamma-Ray Spec. 2005, Santa Fe.

^{233,234}U (n,γ),^{233,234,235}U(n,f) measurement

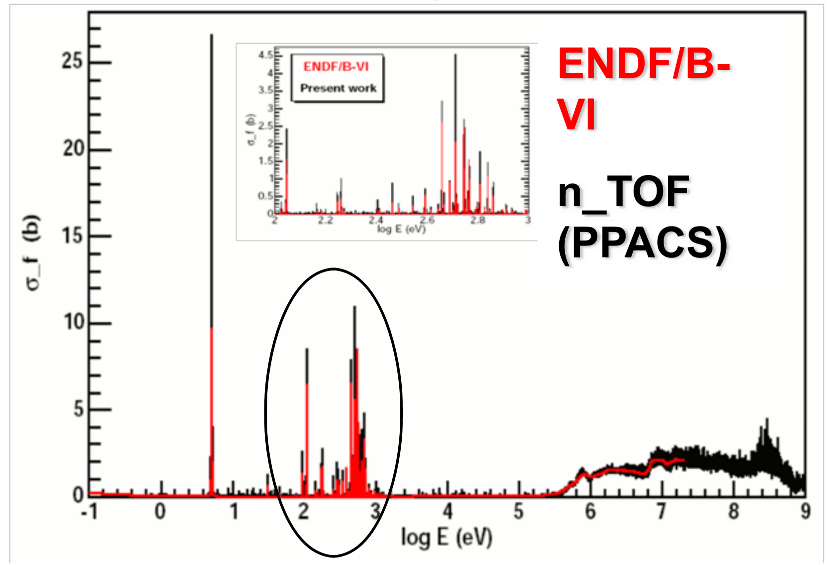
Neutron induced capture

^{234}U(n,γ)



Neutron induced fission

^{234}U(n,f)



W.Dridi, PhD-Thesis, 2006

C.Paradela, PhD-Thesis, 2005

Future plans

n_TOF experiments 2008-…

Capture

Stable Isotopes:

Mo,Bi, Ru: r-process residuals

Fe, Ni, Zn, Se: s-process and structural materials

Radioactive Isotopes:

^{234,236}U, ^{231,233}Pa: Th/U fuel cycle

^{239,240,242}Pu,^{241,243}Am, ^{245}Cm: transmutation of minor actinides (FP-6 project IP-EUROTRANS/NUDATRA)

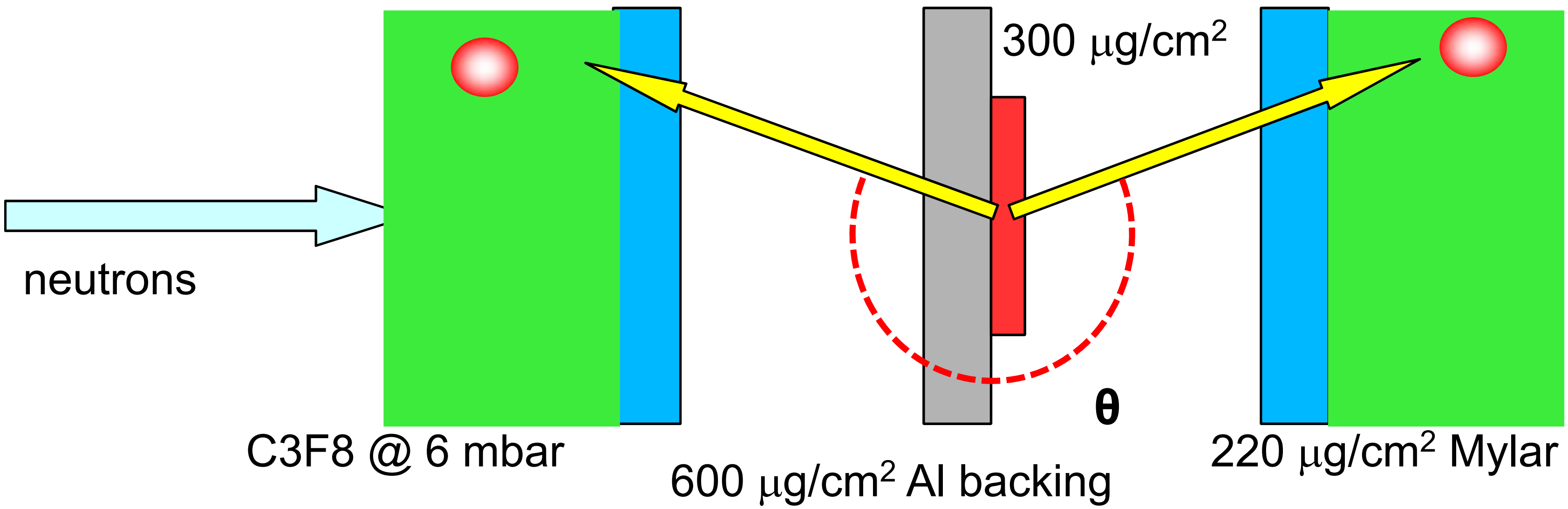
Fission

^{231}Pa,^{234,235,236,238}U

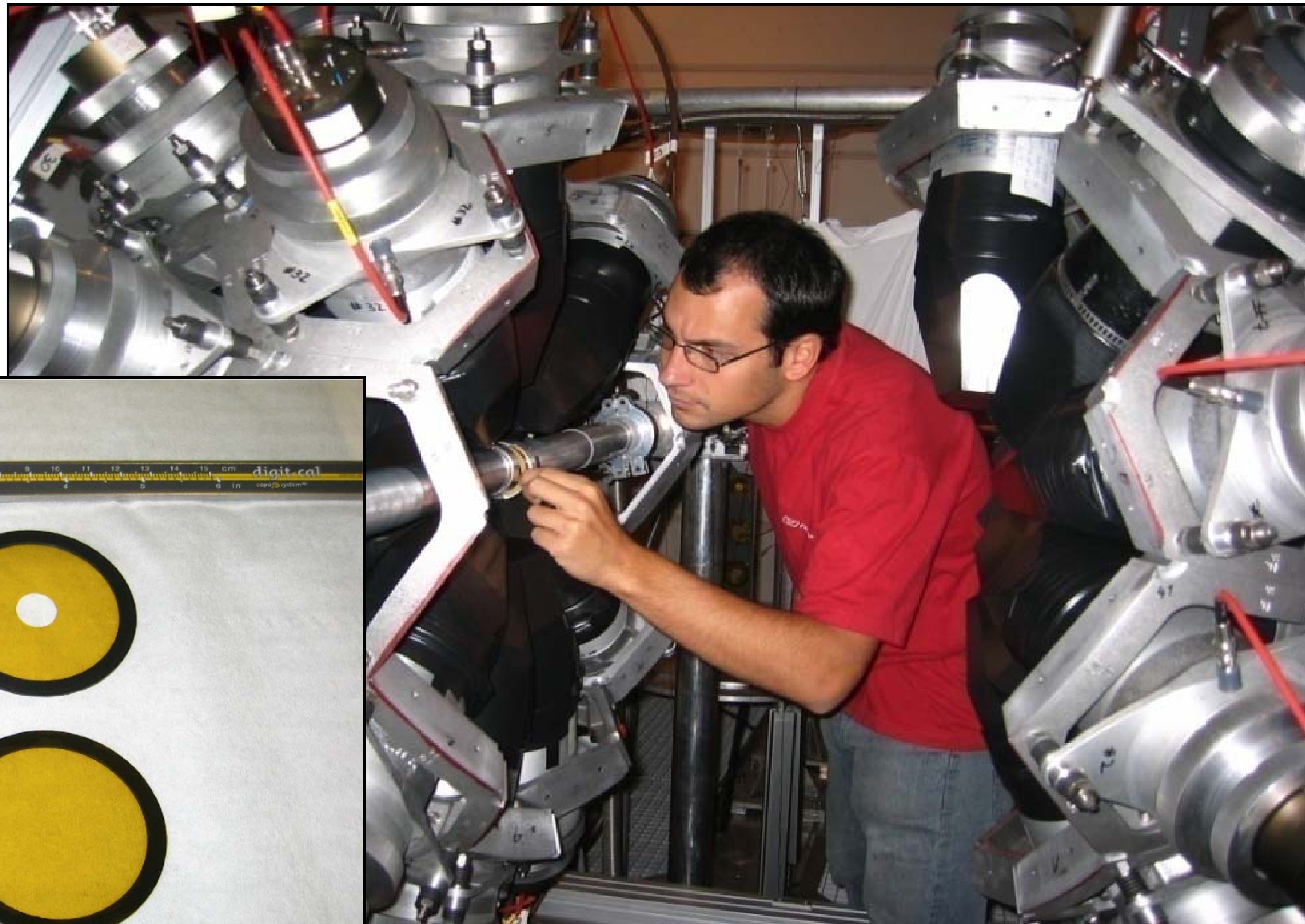
^{241}Pu,^{245}Cm,^{241,243}Am, ^{244,245}Cm

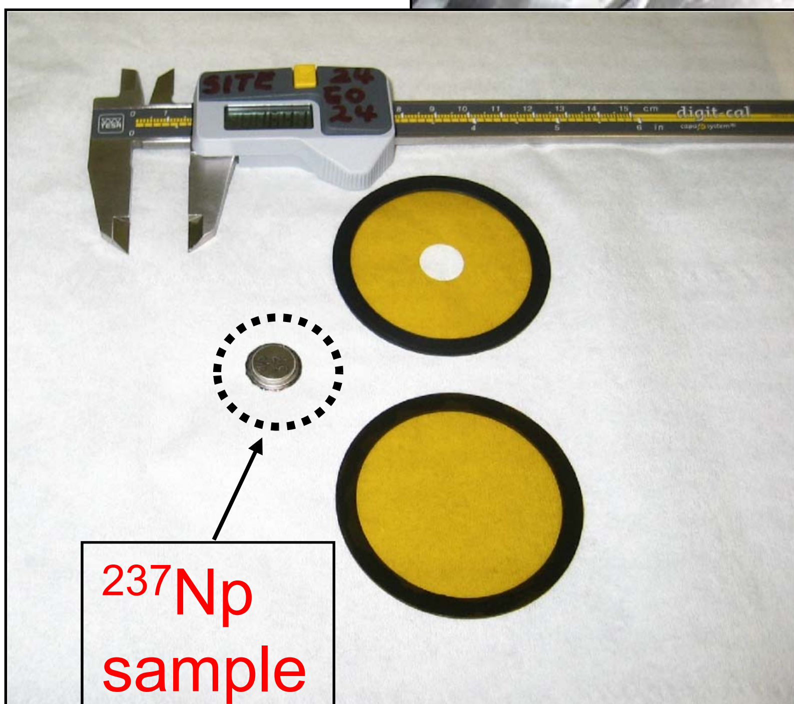
^{234}U: study of vibrational resonances below the bareer

Angular correlations between the fission fragments

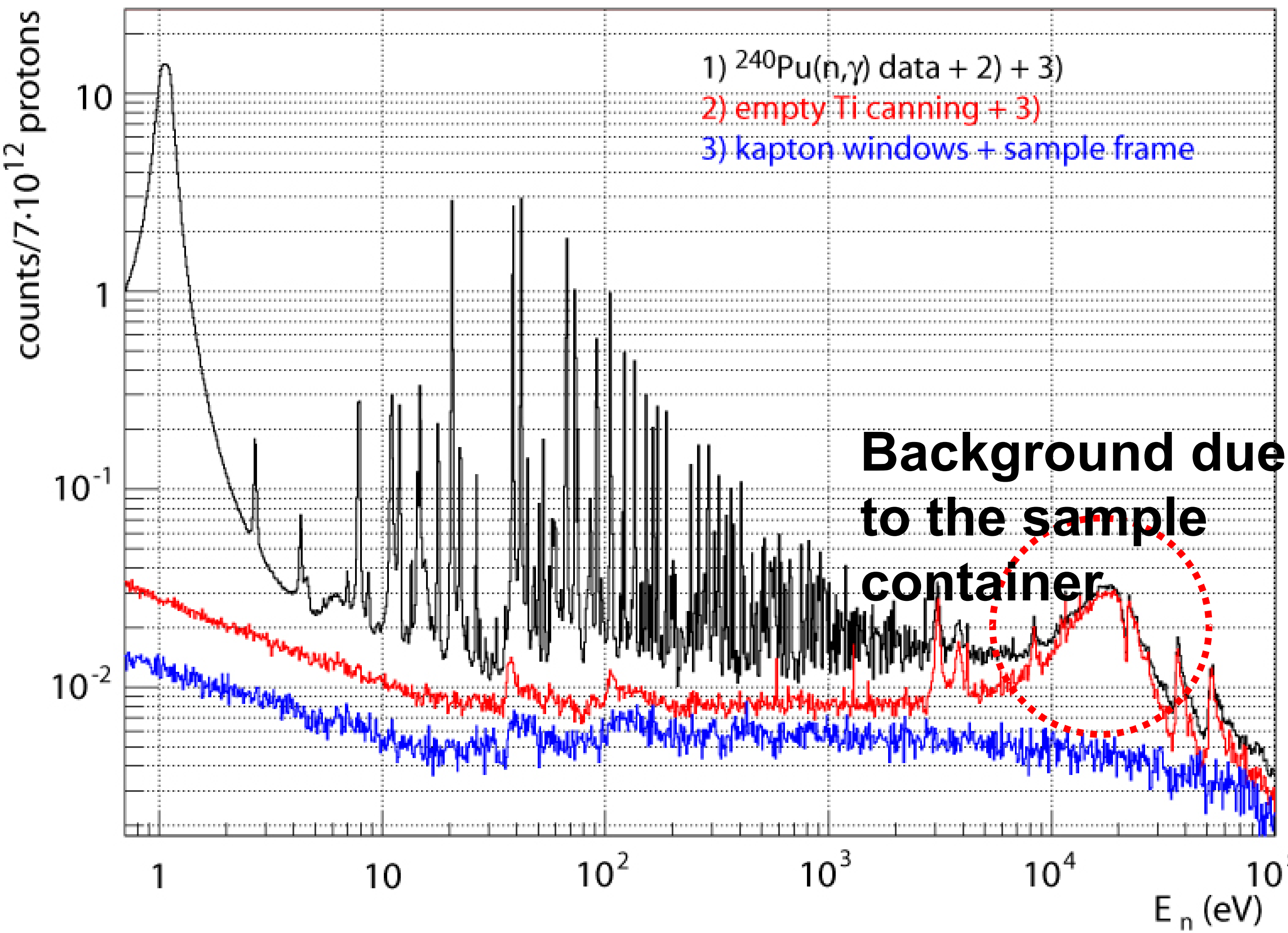


Making better measurements at n_TOF-Ph2

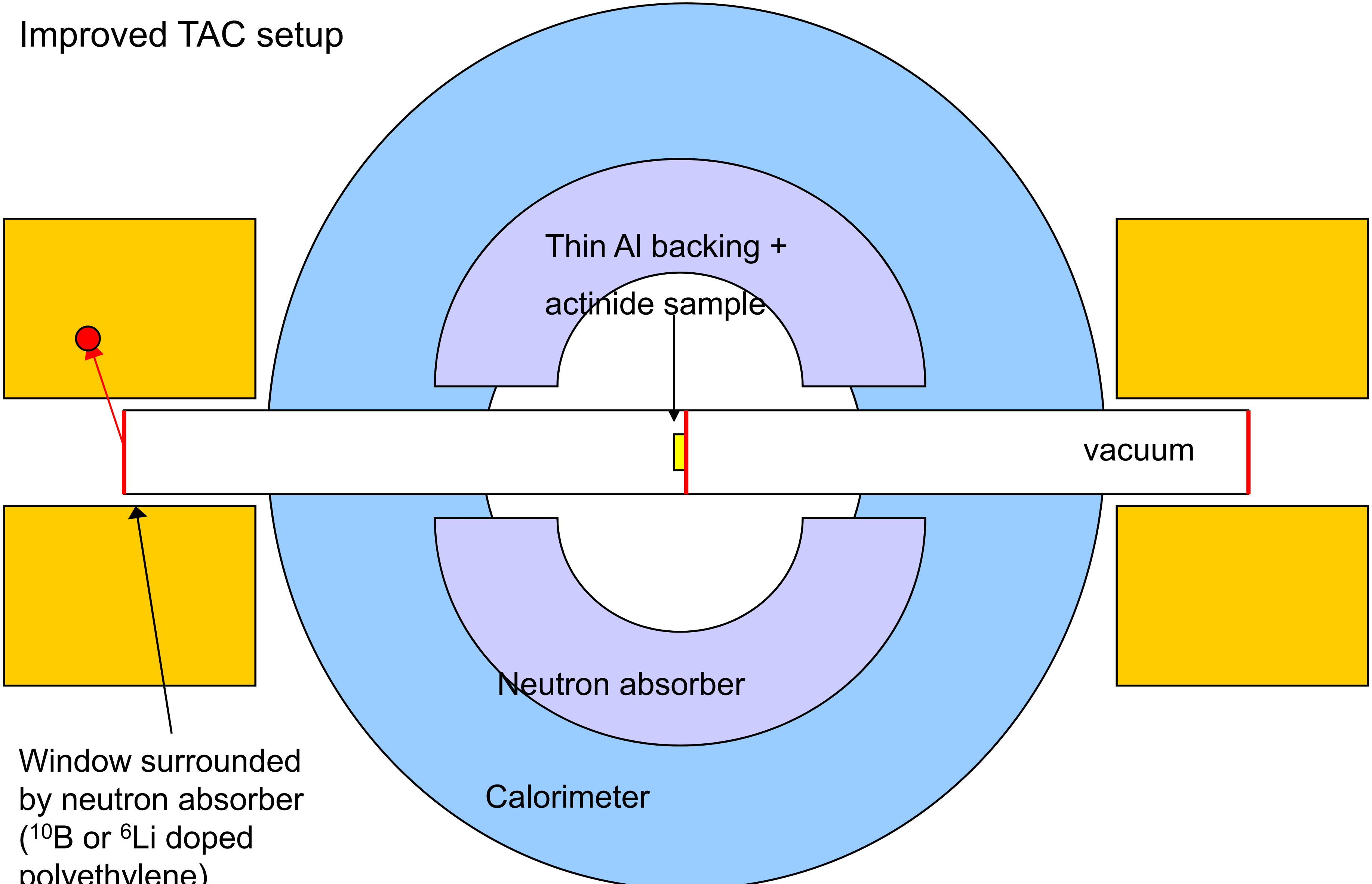




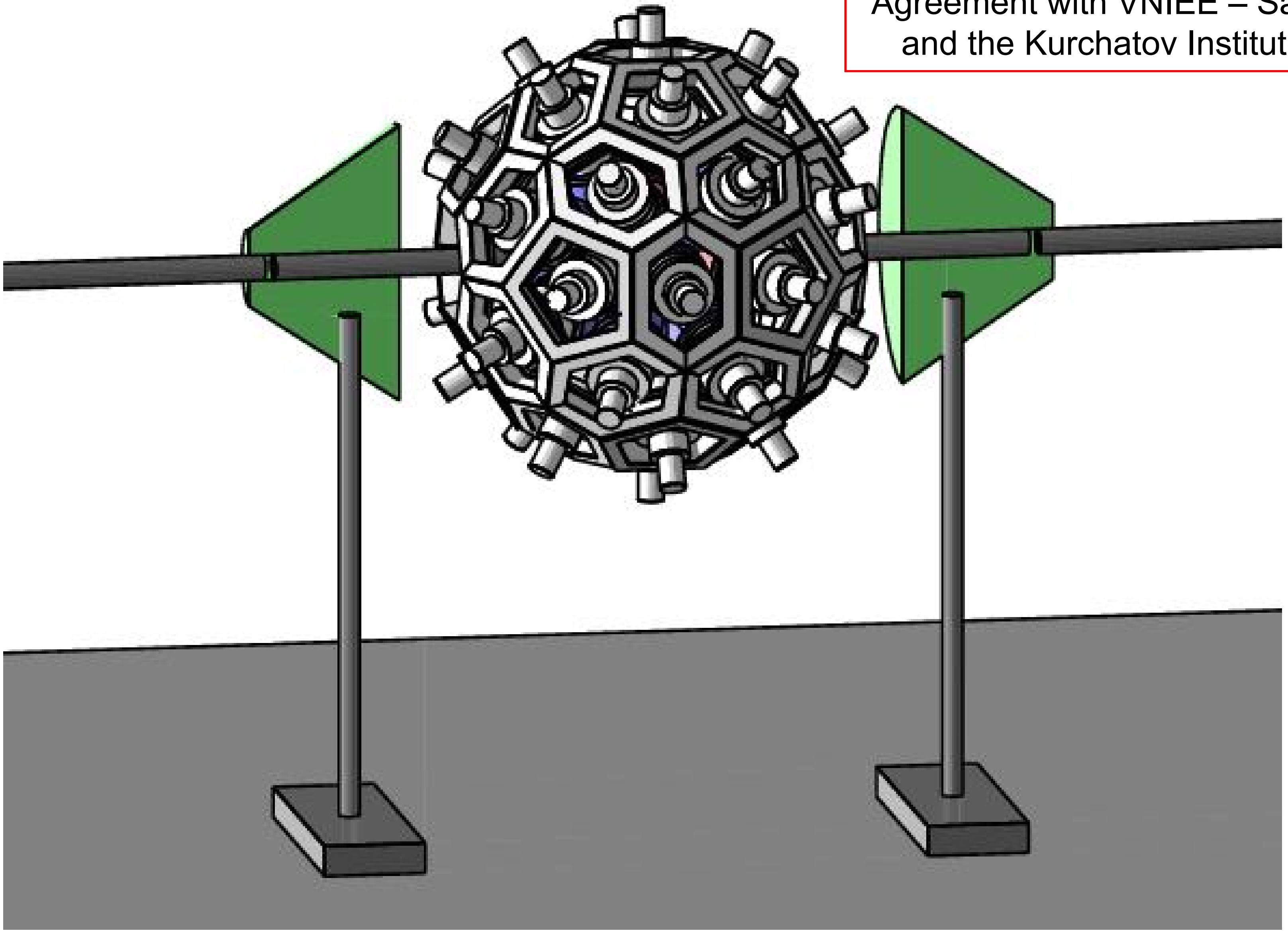
Improvements of the future measurements



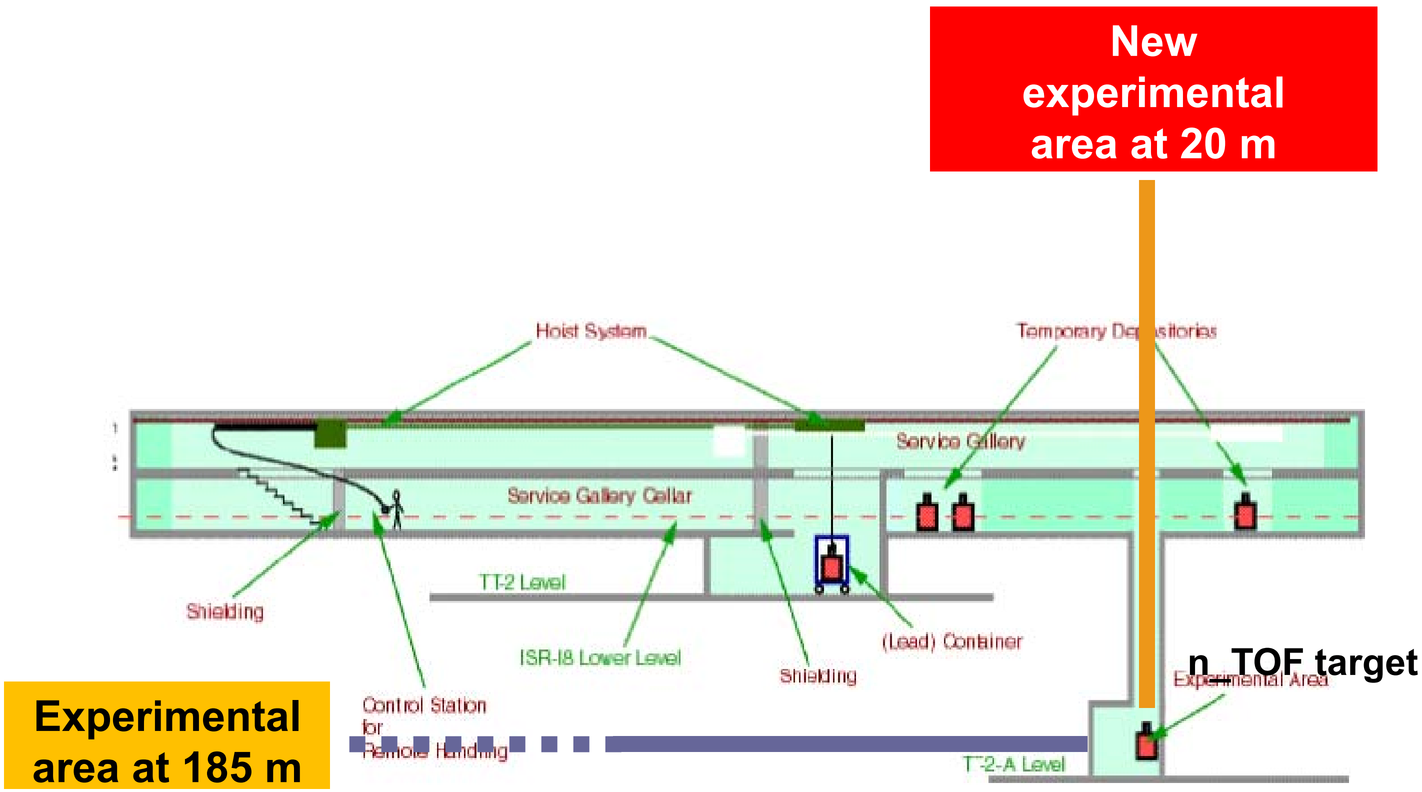
Improved TAC setup



Agreement with VNIEE – Sarov and the Kurchatov Institute.



The (future) second n_TOF experimental area



Flight-path length : ~20 m
at 90 respect to p-beam direction
50 times more intense!

Summary and conclusions

•Differential neutron cross sections for Minor Actinides need to be improved.

•n_TOF @ CERN is a unique facility for it:

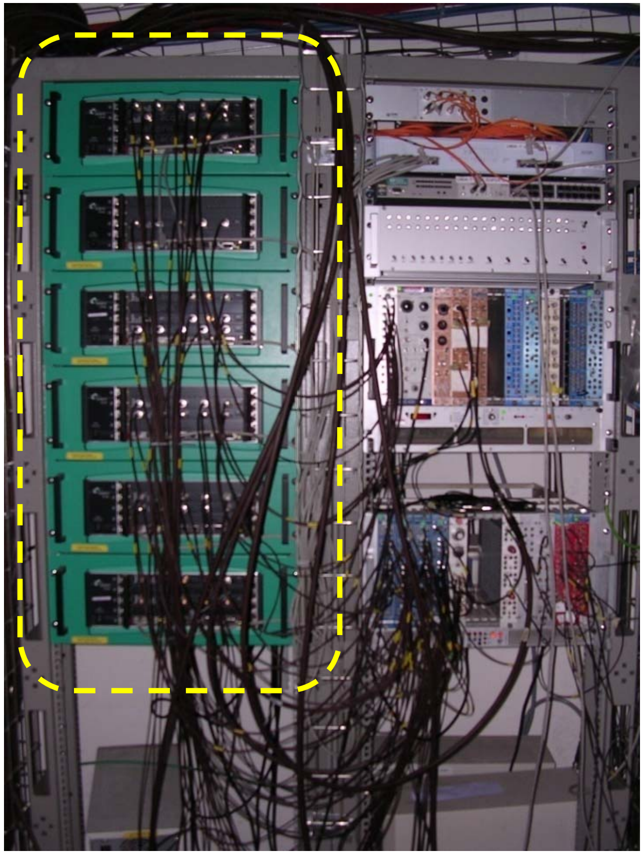
- •High performance detectors for fission (PPAC and FIC) and capture (TAC) cross section measurements
- •Fully digital Data Acquisition System.
- •Already proven for Np & Pu!
- •1 mg mass samples of highly radioactive materials.

We are ready to do so: the n_TOF operation was interrupted in 2004 and will restart in 2008 (3^{rd} of November)!

A brief history of the n_TOF facility

- 1997 - Concept by C.Rubbia validated by TARC exp. [CERN/EET/Int. Note 97-19]
- May ’98 - Further development of the initial idea towards a working facility [CERN/LHC/98-02+Add]
- Aug ’98 – CERN-GELINA joint Letter of Intent [CERN/SPSC/98-15, I220]
- 1999 – Construction started
- Oct 2000 – First proton on the spallation target
- 2001 to 2004 – Experimental program of n_TOF-Ph1 (NTOF-ND-ADS 5th European Union Framework Programme).
- 2005 to 2007 – Shutdown due to the LHC startup + modifications of the spallation target
- June 2007/January 2008 – n_TOF Review Pannels. Green light for a new target!
- December 2007 / January 2008 – Inspection, removal and analysis of the old target.
- March 2008. MoU signed by CERN.
- November 2008 – Start of the n_TOF-Ph2 commissioning.

The n_TOF data acquisition system



n_TOF has been the first neutron beam line world-wide proposing, building and operating a fully digital DAQ. Nowadays, it is becoming a standard at every laboratory.

The n_TOF DAQ consists of ~50 flash ADC channels with 8 bit amplitude resolution and sampling of 500 MSample/s.

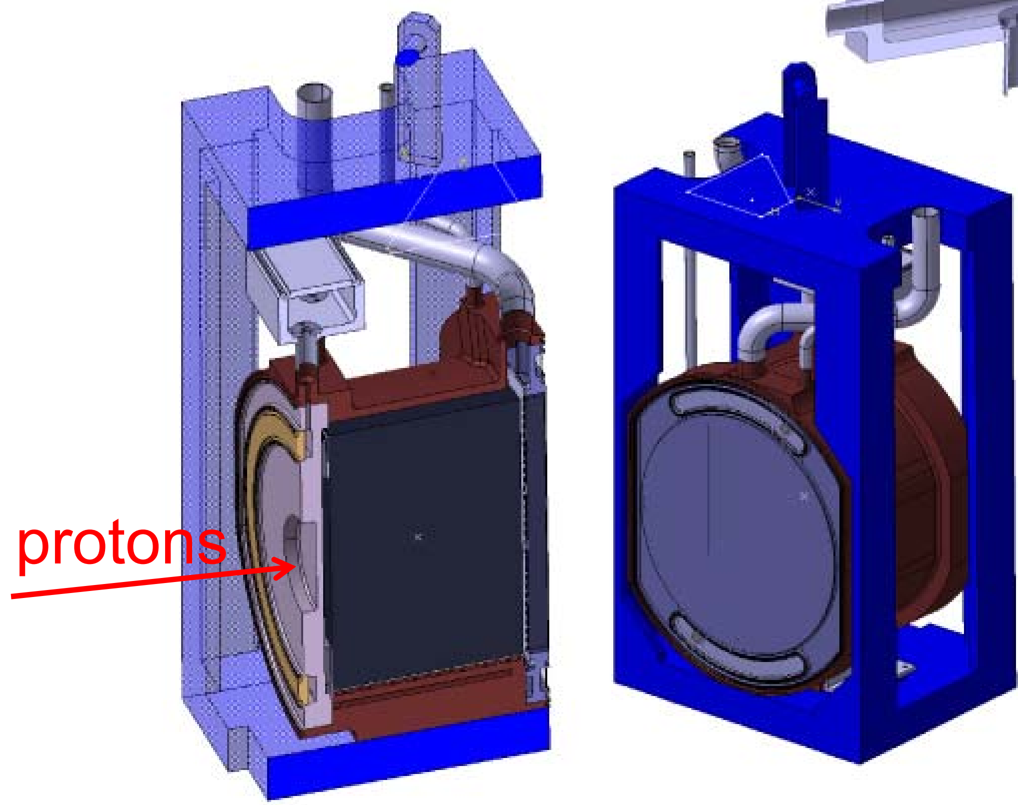
The full history of EVERY detector is digitised during a period of 16 ms (0.7 eV < E_n < 20 GeV) and recorded permanently on tape.

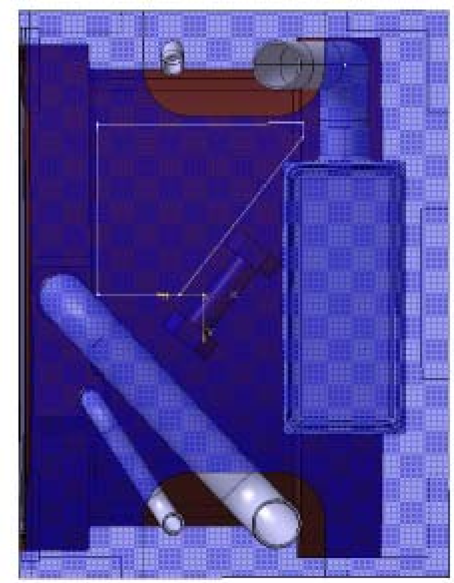
The system has nearly zero dead time.

Simple electronics but everything needs to be done by software.

The new spallation target commissioning

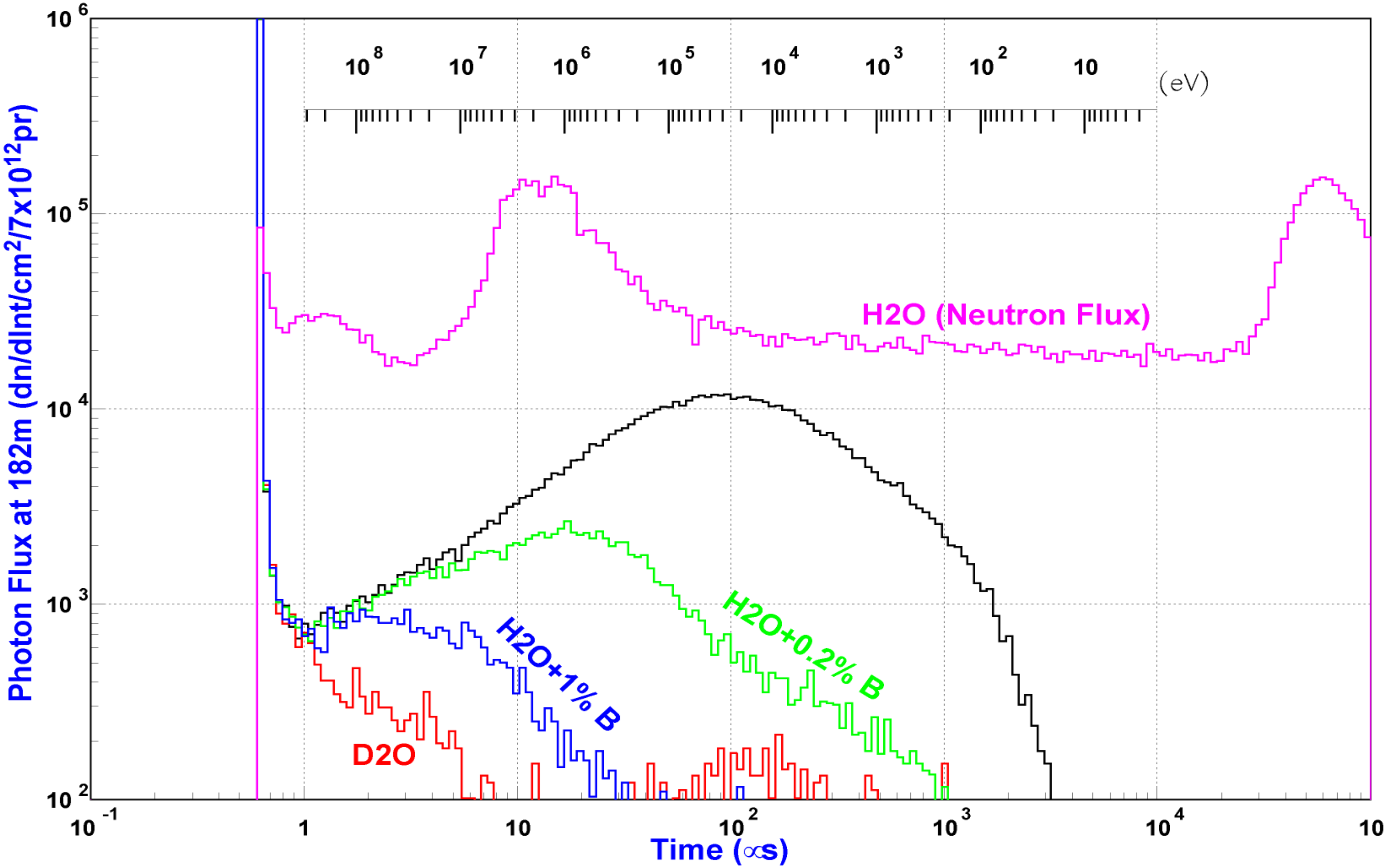
Designed by CERN and CIEMAT





Better experimental conditions

Photon time distribution (E>1MeV)



Isotope	Reaction	Energy range	Original Uncertainty (%)	Required accuracy (%)
^{241}Am	σ_{capt}		40	7.5
			40	5.5
			40	5.1
			20	5.9
			20	6.3
			20	6.9
	σ_{fis}		20	5.6
			20	4.6
			20	3.9
^{243}Am	σ_{capt}			
	σ_{fis}			

T. Yoshida, SND2006-IV.02-1. 240Pu(n,γ) cross section from
WPEC SubGroup 29, http://www.nea.fr/html/science/wpec/index.html
Revisit the resonance capture in ^{235}U, ^{238}U