Nuclear data at n_TOF for fundamental science and technological applications

Enrique M. González Romero
CIEMAT, on behalf of the n_TOF Collaboration

New Opportunities in the Physics Landscape at CERN

CERN, Geneva

12-05-2009

n_TOF: Neutrons at CERN.

Nuclear data at n_TOF for fundamental science and technological applications

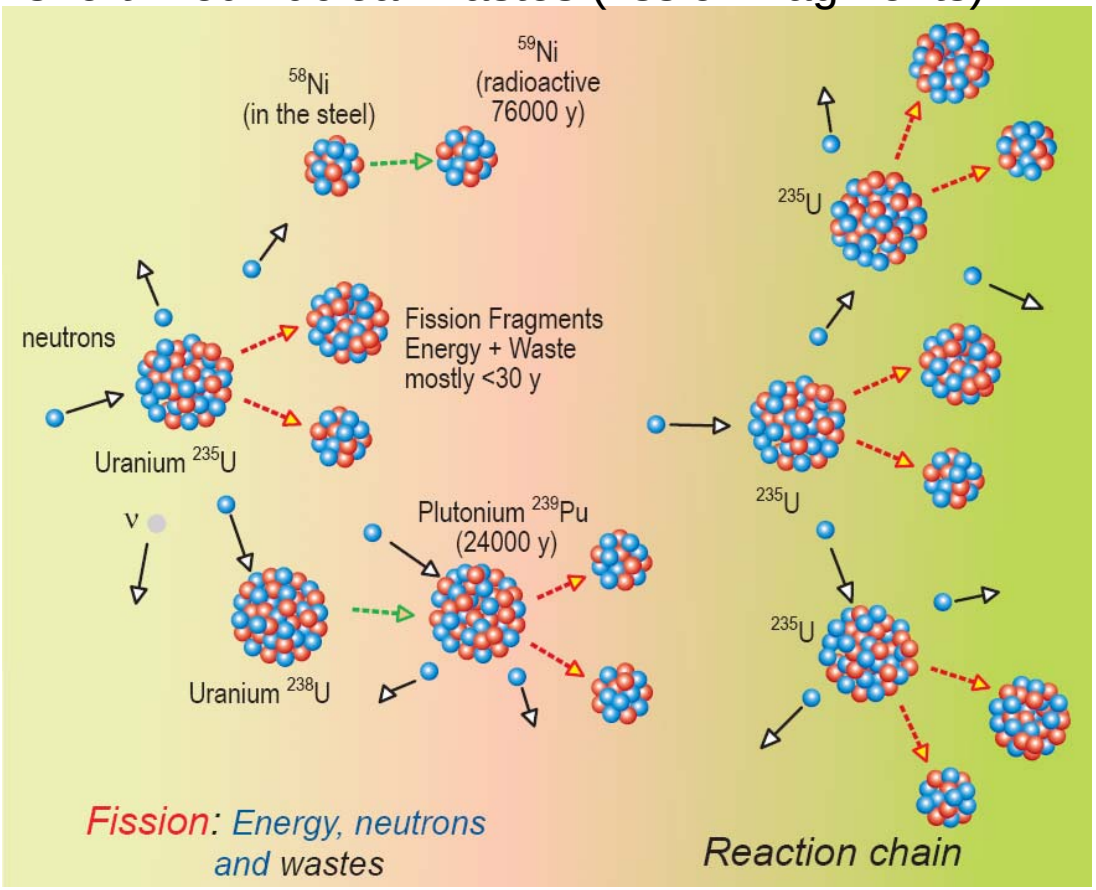
There is a wide range of Nuclear Data applications from:
- Medical treatment and diagnostics to
- search for oil or space exploration,

however the largest social impact is at:
- the electricity production by fission
- the reduction of nuclear wastes.

n_TOF@CERN was designed to provide data for the CERN concept of Energy Amplifier (an ADS) for energy production and Waste Transmutation.

Since then, n_TOF@CERN has become a world-leading facility for this type of measurements.

It is intrinsic to nuclear fission the large energy release per reaction and the production of short lived nuclear wastes (fission fragments)



However, the amount and nature of production of isotopes per activation (Pu and other actinides + structural materials) depends on the technological design.

New reactor concepts under evaluation might allow to reduce (1/100) the existing stock and production of the long lived high level waste (Transmutation).

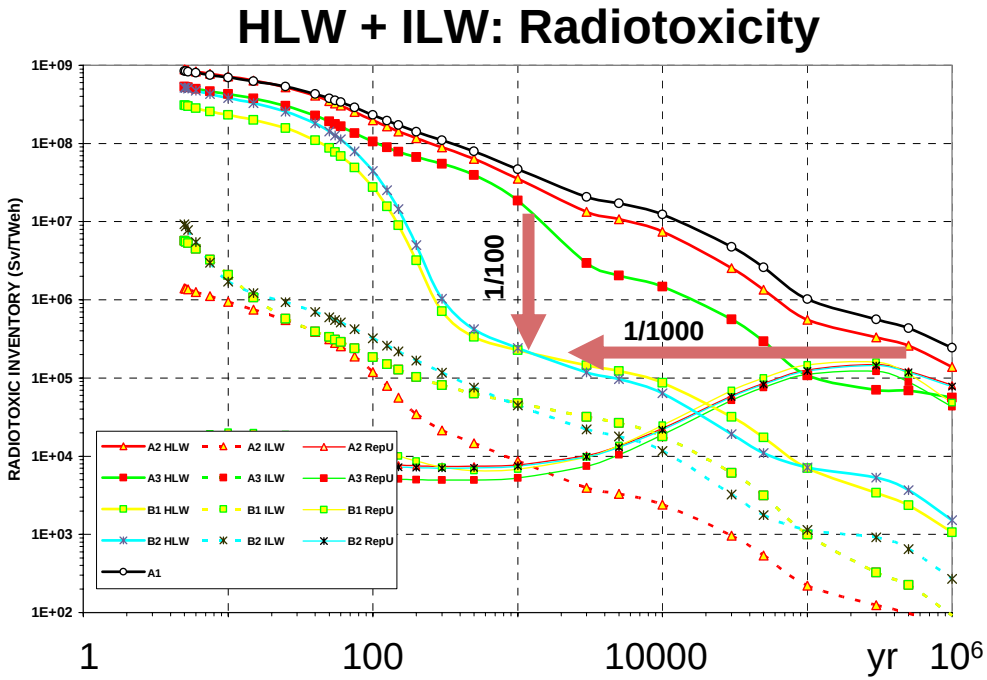
One example: Nuclear waste minimization and P&T

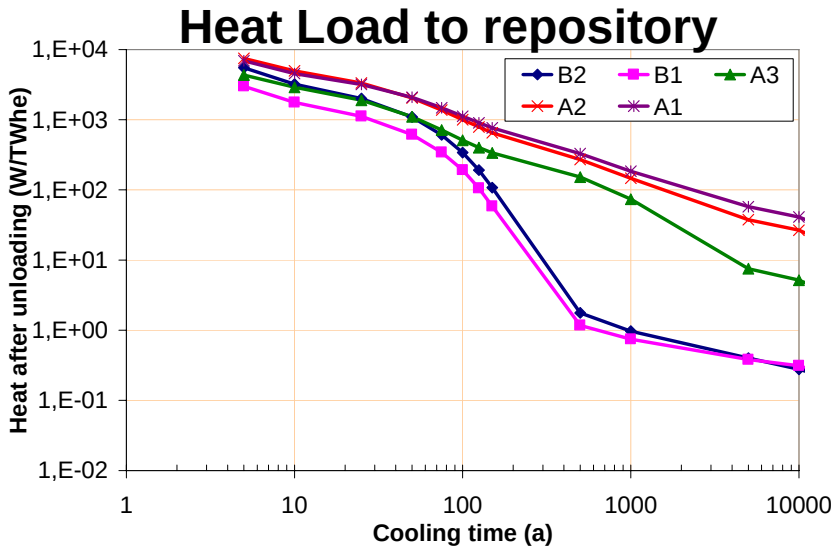
Partitioning & Transmutation
(providing new solutions to social request for nuclear wastes manag.)

Heterogeneity of Spent nuclear fuel Components -> Separate and recycle materials dangerous for environment or valuable to produce energy.

P&T will:

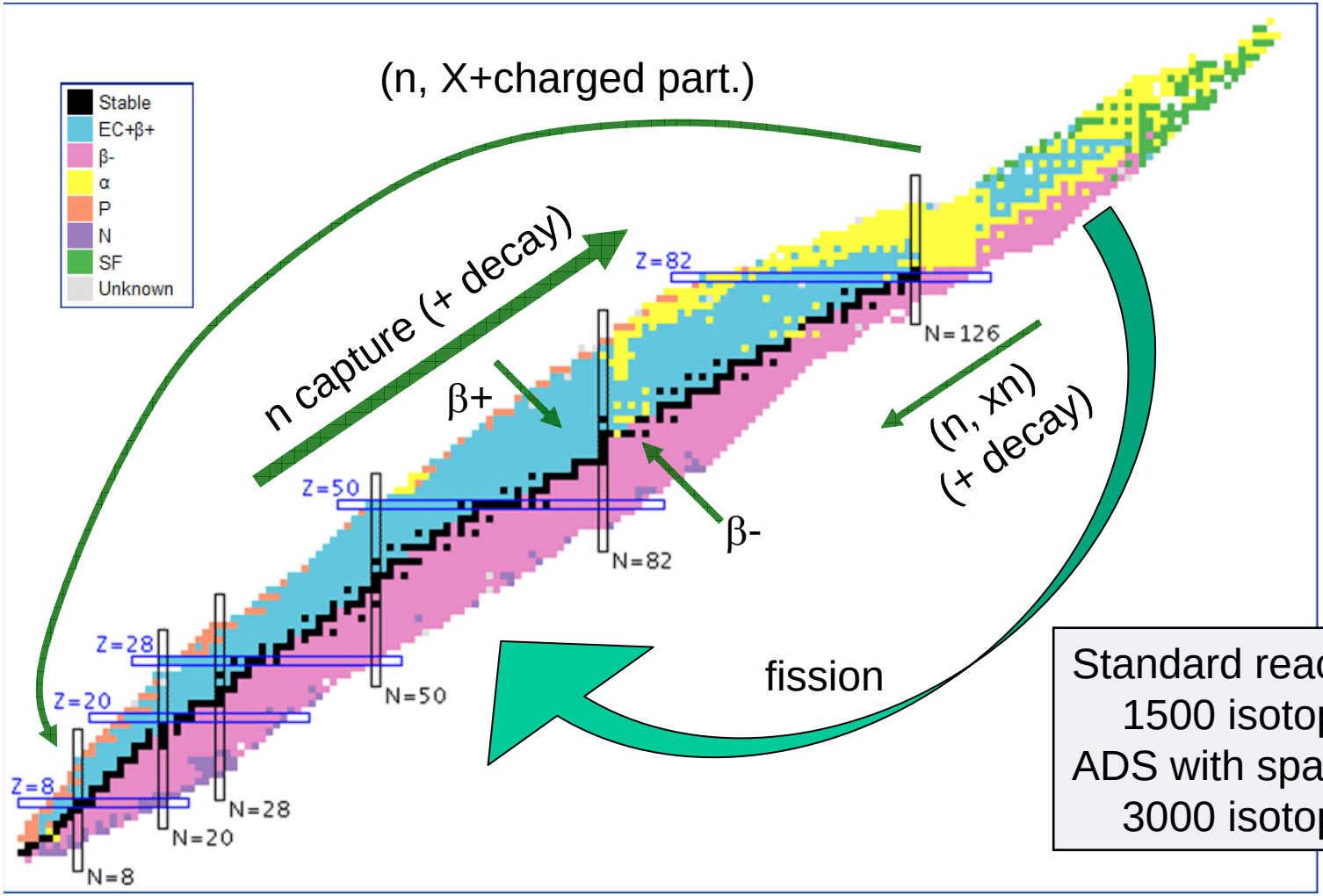
- Reduce by 1/1000 the HLW mass
- Reduce by 1/100 the HLW radiotoxicity
- Reduce by 1/100 the Heat Load to repository
- Reduce by 1/1000 the time
- Reduce by 1/100 the proliferation in repository
- Increase by 5-30 the capacity of hard-rock repositories
- Increase by 30% to 3000% the energy from the fuel





Main reactions in a nuclear reactor or transmutation device

- n- induced fission (energy + wastes)
- neutron capture (activation + breeding)
- elastic and inelastic neutron scattering
- radioactive decay
- (n,xn), (n, charged particle), …



Standard reactor
1500 isotopes
ADS with spallation
3000 isotopes

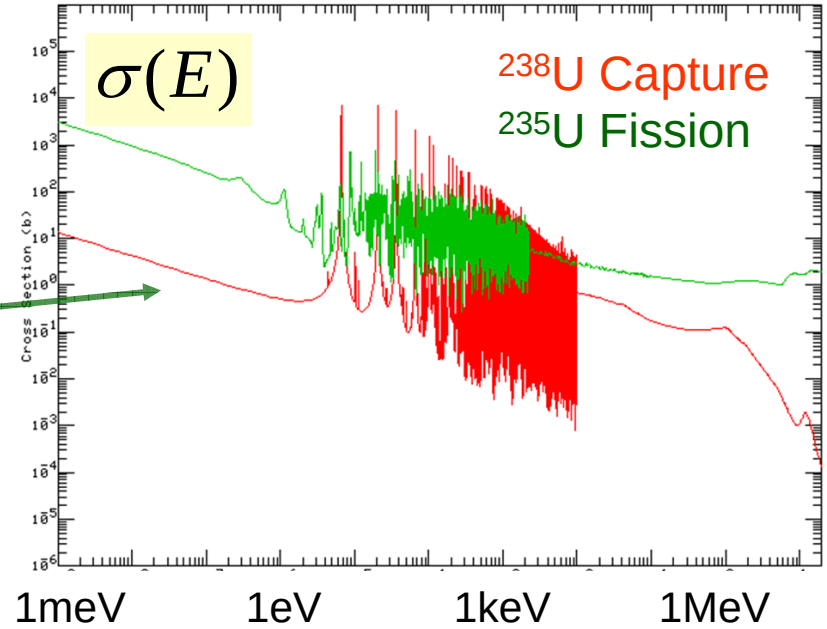
$$Rate = \int n\,\sigma(E)\,\varphi(E)\,dE$$

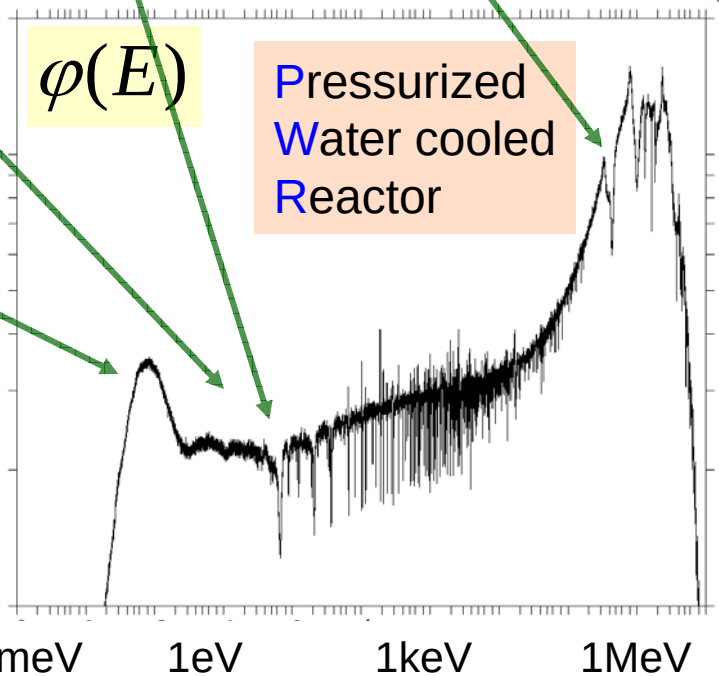
- n- induced fission
- neutron capture

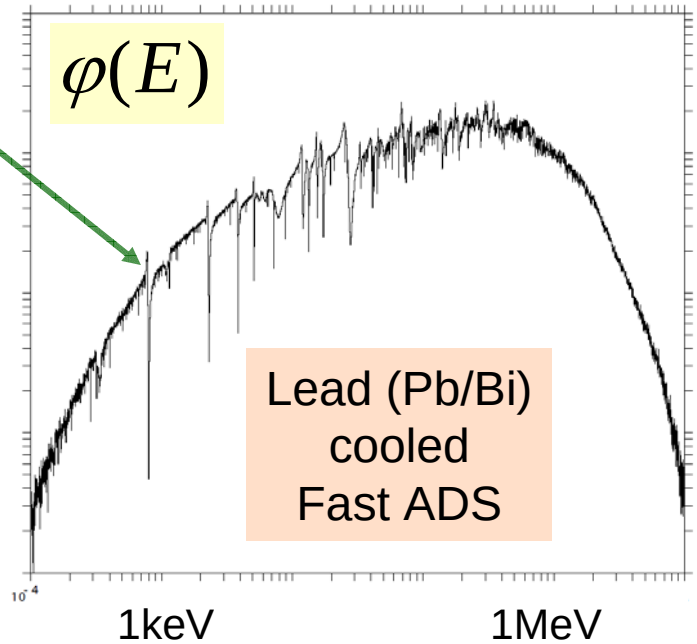
- elastic and inelastic neutron scattering

Thermalization, Moderation

Resonances (absorption, elastic, inelastic,…)





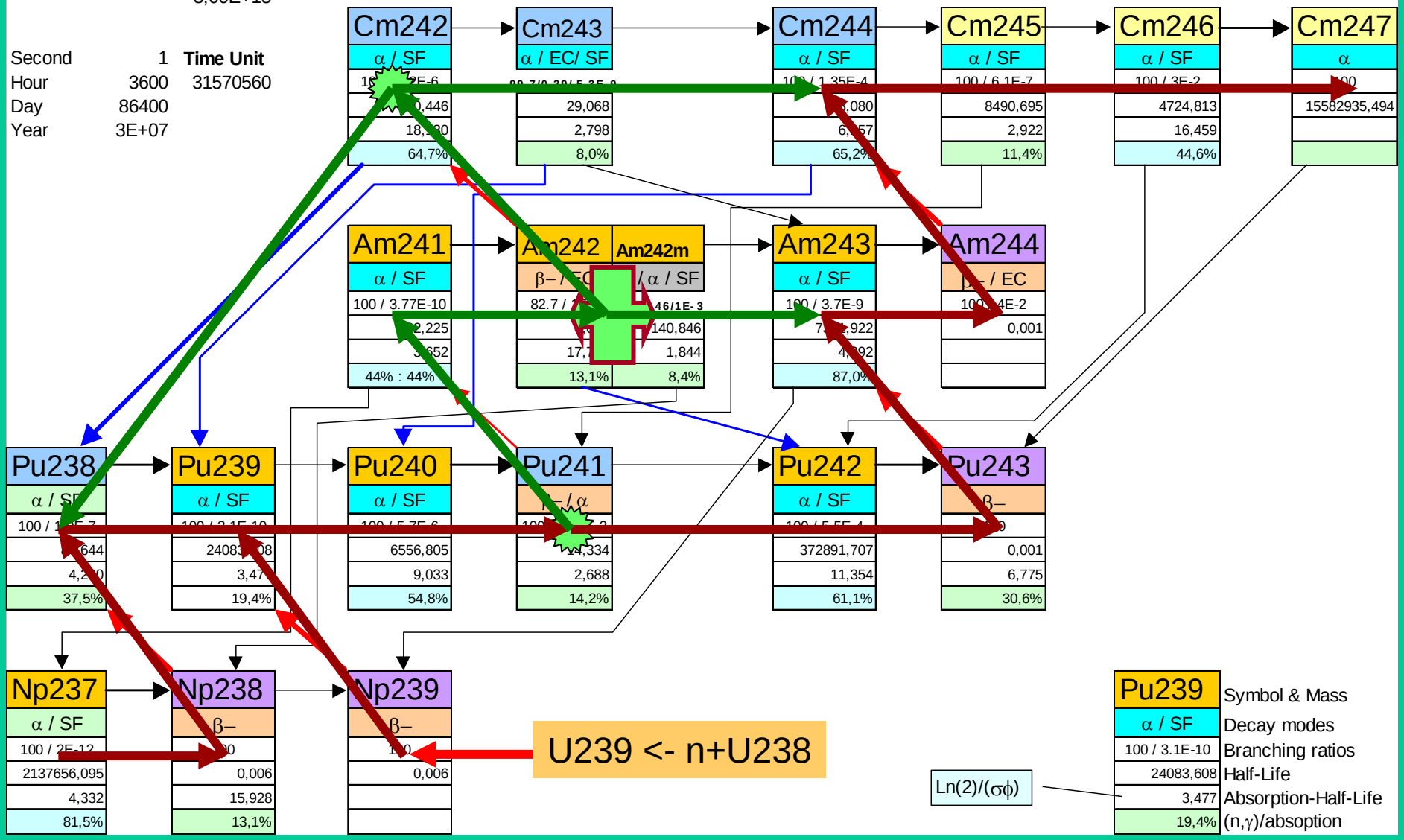


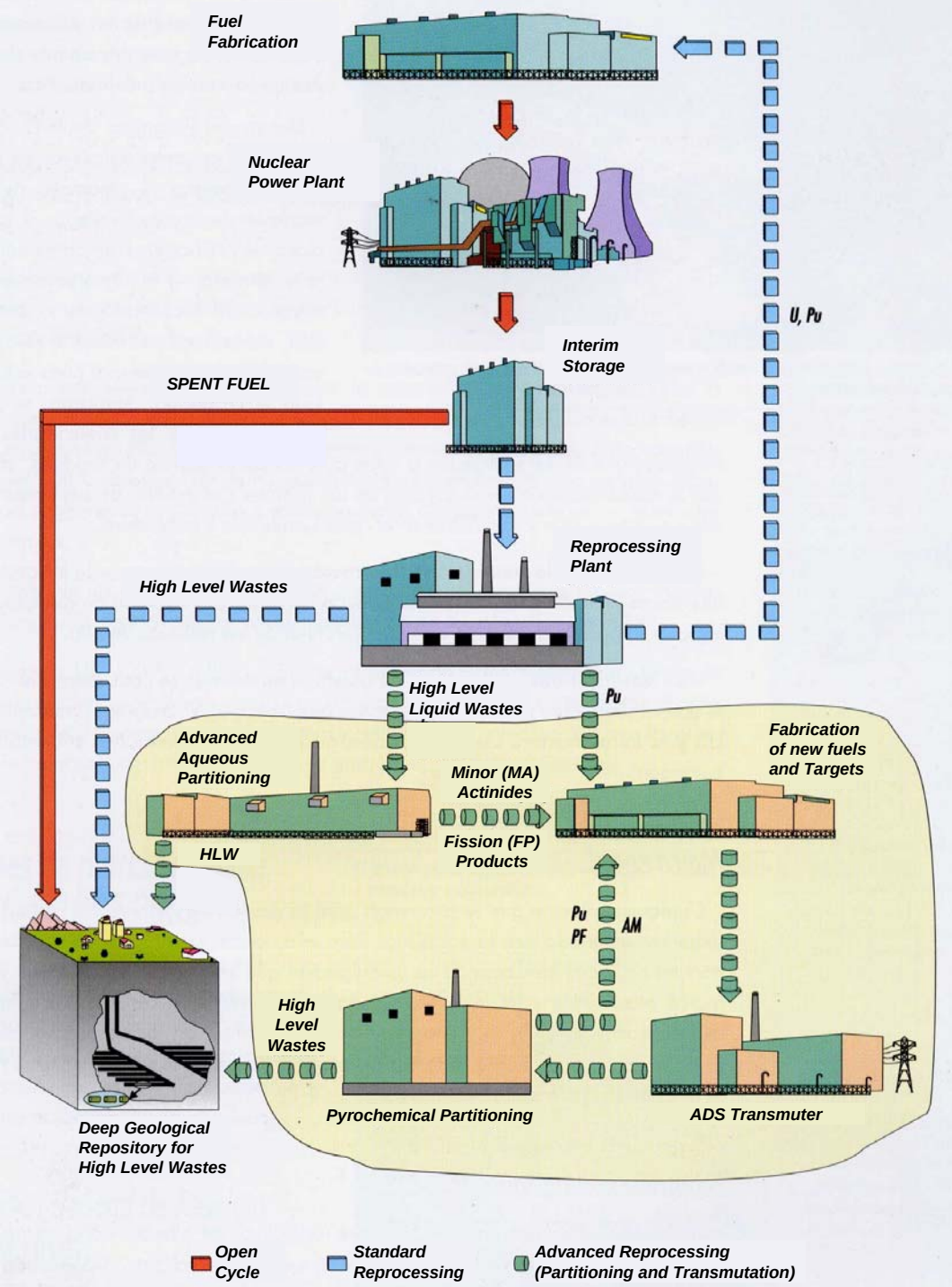
TRU Transmutation Scheme
Fast Spectrum

- Present in nuclear wastes
- Medium Half-Life (<100 años)
- Short Half-Life (< 30 dias)
- High A actinides
- Thermal and Fast Fission
- Fast Fissión
- Low Fission Cross Section

Av. Flux Intensity (n/cm2/s)
3,00E+15

Second	1	**Time Unit**
Hour	3600	31570560
Day	86400	
Year	3E+07	





Reactors: **(Δ<0.5%)**

Performance: Reaction rates, Power distribution, Flux, Energy Spectrum

Safety: Criticality, Feedbacks, Reactivity coeffs, Damage, Shielding

Waste: Isotopic evolution, activation

Storage, Reprocessing and Fabrication plants: **(Δ<5%-10%)**

Isotopic composition !!!

Radioactivity, Neutron emissions, Decay Heat,

Proliferation interest

Storage, Reprocessing and Fabrication plants: **(Δ<5%-10%)**

Isotopic composition !!!

Radioactivity, Neutron emissions, Decay Heat,

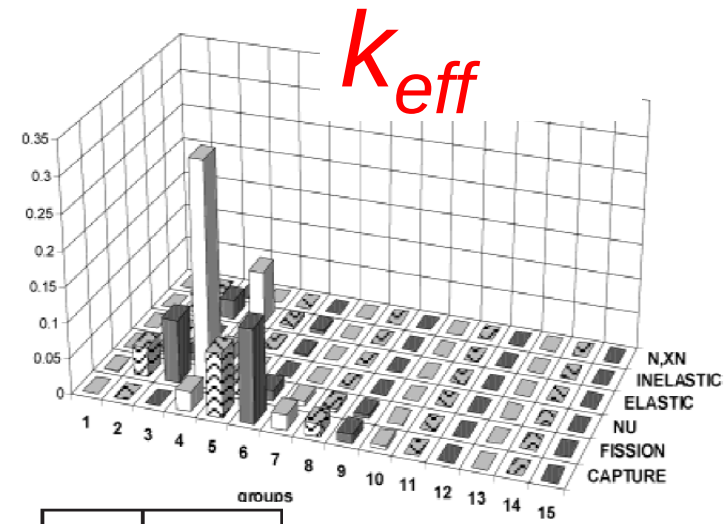
Proliferation interest

Radiotoxicity and Dose to Public and Environment

Effective capacity

Sensitivity analysis – ADS for Transmutation

Isotope	σ_{cap}	σ_{fiss}	ν	σ_{el}	σ_{inel}	$\sigma_{n,2n}$	Total[a]
^{238}Pu	0.01	0.11	0.02	—	—	—	0.11
^{239}Pu	0.04	0.51	0.11	—	0.04	—	0.53
^{240}Pu	0.05	0.18	0.05	—	0.02	—	0.19
^{241}Pu	0.04	0.30	0.03	—	0.01	—	0.31
^{242}Pu	0.01	0.05	0.02	—	0.01	—	0.06
^{237}Np	0.24	0.70	0.21	—	0.14	—	0.78
^{241}Am	1.32	1.12	0.38	—	0.22	—	1.79
^{242m}Am	0.01	0.09	0.03	—	0.01	—	0.10
^{243}Am	0.74	0.59	0.21	—	0.60	—	1.14
^{242}Cm	—	—	—	—	—	—	—
^{243}Cm	—	0.05	0.01	—	—	—	0.05
^{244}Cm	0.13	1.09	0.18	—	0.07	—	1.11
^{245}Cm	0.01	0.41	0.08	—	0.01	—	0.42
^{246}Cm	—	—	—	—		—	—
^{56}Fe	0.03	—	—	0.05	0.49	—	0.50
^{57}Fe	—	—	—	—	0.06	—	0.06
^{52}Cr	0.01	—	—	0.01	0.03	—	0.03
^{58}Ni	—	—	—	—		—	—
Zr	0.03	—	—	0.03	0.07	—	0.09
^{15}N	—	—	—	0.19	0.01	—	0.19
Pb	0.02	—	—	0.10	0.41	0.02	0.43
Bi	0.04	—	—	0.11	0.49	0.03	0.50
Total[a]	1.54	1.97	0.54	0.25	1.05	0.04	2.77



Group	(MeV)[a]
1	19.6
2	6.07
3	2.23
4	1.35
5	4.98e−1[c]
6	1.83e−1
7	6.74e−2
8	2.48e−2
9	9.12e−3
10	2.04e−3
11	4.54e−4
12	2.26e−5
13	4.00e−6
14	5.40e−7
15	1.00e−7

a) Upper limit of the group

NEA/WPEC-26.

One possible optimization for target accuracy for innovative systems using recent covariance data evaluations (BOLNA).
M. Salvatores and R. Jacqmin (Eds), NEA/WPEC-26.
ISBN 978-92-64-99053-1

Similar tables for each present or proposed future reactor

Still serious dependence on the reactor and fuel models and on the transmutation model (homogeneous) can slightly modify the target accuracy and details on the priority order

Table 30. ADMAB: uncertainty reduction requirements needed to meet integral parameter target accuracies

Isotope	Cross-Section	Energy range	Uncertainty (%)		
			Initial	Required	
				$\lambda=1$	$\lambda\neq1$ [a]
Pu238	σ_{fiss}	6.07 - 0.498 MeV	20	3	3
	ν	1.35 - 0. 183 MeV	7	3	3
Pu239	σ_{capt}	498 - 2.03 keV	12	4	3
	σ_{inel}	6.07 - 0.498 MeV	25	5	6
Pu240	σ_{capt}	183 - 67.4 keV	14	6	6
	σ_{fiss}	2.23 - 0.498 MeV	6	2	2
	ν	1.35 - 0.498 MeV	4	2	2
Pu241	σ_{capt}	1.35 - 0. 183 MeV	20	7	7
	σ_{fiss}	6.07 MeV-22.6 eV	15	2	2
Pu242	σ_{capt}	24.8 - 9.12 keV	35	10	10
	σ_{fiss}	6.07 - 0.498 MeV	20	4	4
Np237	σ_{capt}	498 - 0.454 keV	6	3	3
	σ_{fiss}	6.07 - 0.183 MeV	8	2	2
	σ_{inel}	2.23 – 0.183 MeV	25	5	6
Am241	σ_{capt}	1.35 MeV- 0.454 keV	8	2	2
	σ_{fiss}	6.07 – 0.183 MeV	10	1	1
	ν	6.07 - 1.35 MeV	2	1	1
	σ_{inel}	6.07 – 0.183 MeV	25	4	5
Am242m	σ_{fiss}	1.35 MeV- 9.12 keV	17	5	5
Am243	σ_{capt}	1.35 MeV- 0.454 keV	10	2	2
	σ_{fiss}	6.07 - 0.498 MeV	10	2	2
	σ_{inel}	6.07 MeV- 24.8 keV	40	2	3
Cm242	σ_{fiss}	6.07 MeV- 67.4 keV	55	26	26
Cm243	σ_{fiss}	1.35 MeV- 67.4 keV	50	8	8
Cm244	σ_{capt}	498 -9.12 keV	20	6	6
	σ_{fiss}	6.07 MeV- 67.4 keV	45	2	2
	ν	6.07 – 0.183 MeV	10	1	1
Cm245	σ_{fiss}	6.07 MeV- 0.454 keV	45	3	3
Fe56	σ_{capt}	183 - 0.454 keV	12	5	3
	σ_{inel}	6.07 - 0.498 MeV	20	2	2
Zr90	σ_{inel}	6.07 - 2.23 MeV	18	3	4
N15	σ_{el}	2.23 MeV - 67.4 keV	5	1	1
Pb	σ_{capt}	9.12 - 2.03 keV	20	20	14
	σ_{inel}	6.07 - 2.23 MeV	12	3	4
Bi209	σ_{inel}	2.23 - 0.498 MeV	34	3	3

[a] See Table 24 for $\lambda \neq 1$, case A

Important isotopes for Transmutation Fuel Cycles: The multi-recycling point of view

Report of the Numerical results from the Evaluation of the nuclear data sensitivities, Priority list and table of required accuracies for nuclear data. E. Gonzalez-Romero (Ed), NUDATRA Deliverable D5.11 from IP-Eurotrans

Isotopes	Uncertainty in the abundance %			Important for:			
	Burnup (GWd/t) 150	500	800				
^{234}U	4.6	16.1	32.4	T	DH		
^{235}U	13.1	18.4	15.5	T			
^{236}U	1.8	7.6	12.6	T			
^{237}Np	6.3	23.7	28.1	T			
^{238}Pu	4.3	10.8	19.3	T	DH		R
^{239}Pu	4.6	12.9	17.8	T	DH		R
^{240}Pu	2.0	7.0	14.4	T	DH		R
^{241}Pu	8.2	14.7	17.0	T			
^{242}Pu	2.1	7.9	16.2	T	DH		R
^{241}Am	7.2	20.7	26.0	T	DH		R
^{242m}Am	12.8	28.6	34.4	T			
^{243}Am	6.6	15.6	20.2	T	DH		R
^{242}Cm	10.7	7.7	15.6	T	DH		
^{243}Cm	23.3	32.6	35.7				
^{244}Cm	6.0	13.3	19.1	T	DH	N	R
^{245}Cm	13.3	18.8	16.3	T	DH		R
^{246}Cm	7.5	21.7	31.5	T	DH	N	R
^{247}Cm	15.4	27.2	31.6	T			
^{248}Cm	6.4	19.8	31.4			N	
^{250}Cf	31.9	28.9	36.9			N	
^{252}Cf	52.4	46.1	48.9			N	

T= Transmutation efficiency
DH= Decay Heat load
N = Neutron emission
R = Radiotoxicity

Identifying and setting priorities of Nuclear data for applications: An international endeavor

- Applications set the problems to be analyzed and the required accuracies for the simulation.
- Detailed uncertainty and covariance propagation to evaluate the accuracy.
- Sensitivity analysis identify the relevance of each data for each isotope/reaction/energy on the most significant parameters
- Linear Optimization with expert assessment of “cost” (experimental difficulties) to set priorities

Efforts coordinated by dedicated expert groups of NEA/OECD, IAEA, and dedicated EU framework programs

EU support and demand for nuclear data measurements:

- Clear and repeated demand from the Nuclear Waste community, Sustainability of Nuclear Energy (Resources, Safety, Waste) by EU Framework Program calls:
  - FP5: nTOF_ND_ADS (*start of n_TOF facility at CERN*)
  - FP6: NUDATRA (inside EUROTRANS),
  - FP6: EFNUDAT (Transnat. Access) + CANDIDE (Roadmap for ND)
  - FP7: ANDES proposal to WP2009
- Collaboration with other measurements at USA, Japan, Russia.

One EU call on Nuclear Data for each FP (FP5-FP7),
n_TOF measurements in all of them

n_TOF beam characteristics

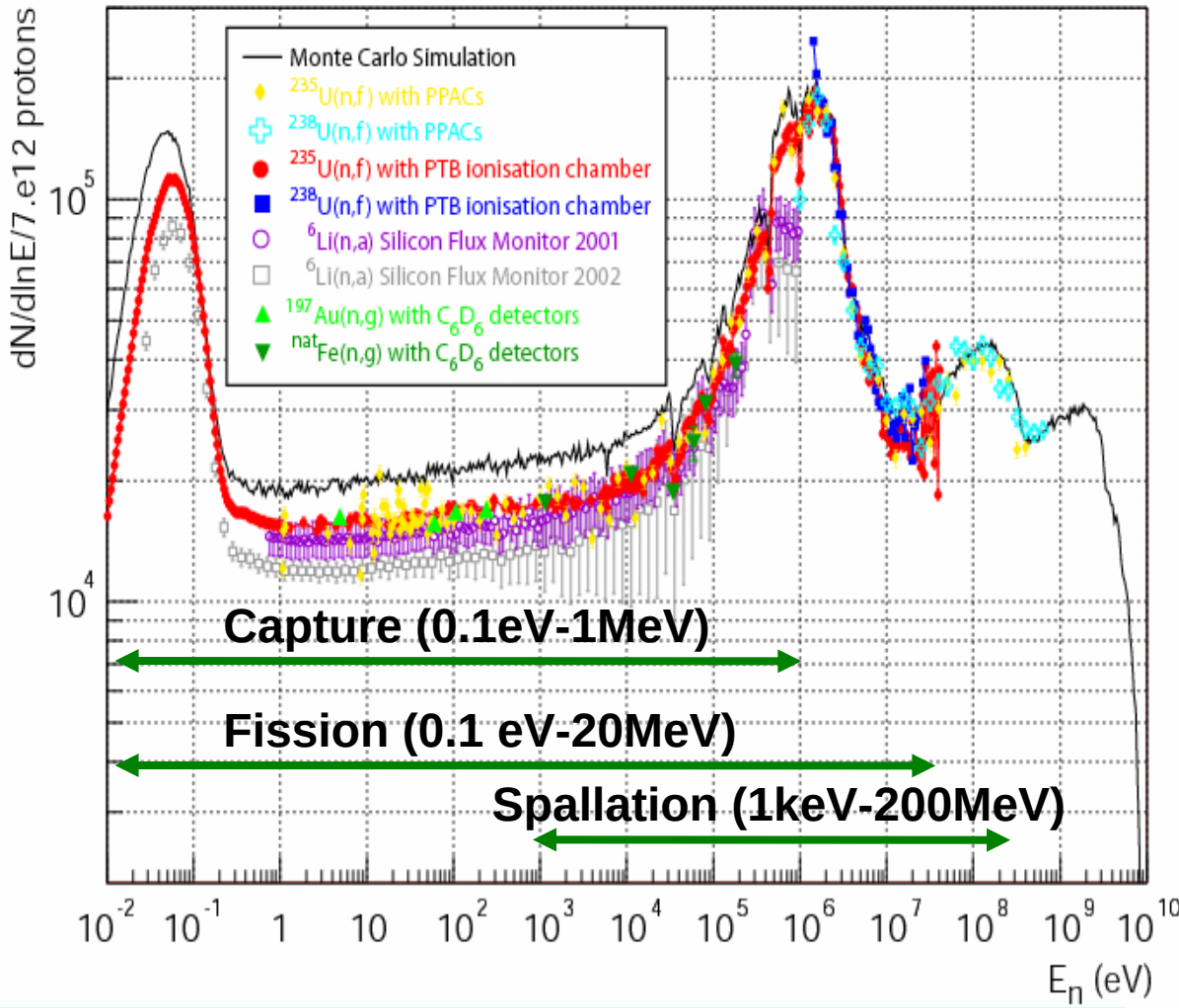
- Wide energy range
- High instantaneous n- flux
- High resolution
- Low ambient background
- Low repetition frequency
- Favorable duty cycle for radioactive samples.

The neutron fluence in EAR-1

Energy range	Uncollimated [n/pulse/cm2]	Capture mode [n/pulse]	Fission mode [n/pulse]
< 1 eV	**2.0E+05**	**3.1E+05**	**2.0E+06**
1 eV - 10 eV	2.7E+04	4.5E+04	2.9E+05
10 eV - 100 eV	2.9E+04	4.7E+04	3.1E+05
100 eV - 1000 eV	3.0E+04	5.1E+04	3.3E+05
1 eV - 1 keV	**8.6E+04**	**1.4E+05**	**9.3E+05**
1 keV - 10 keV	3.2E+04	5.4E+04	3.6E+05
10 keV - 100 keV	3.9E+04	7.1E+04	4.7E+05
100 keV - 1000 keV	1.1E+05	2.3E+05	1.5E+06
1 keV - 1 MeV	**1.8E+05**	**3.5E+05**	**2.3E+06**
1 MeV - 10 MeV	8.3E+04	2.4E+05	1.7E+06
10 MeV - 100 MeV	2.8E+04	7.2E+04	5.1E+05
> 100 MeV	4.4E+04	1.2E+05	5.6E+05
1 MeV - > 100 MeV	**1.6E+05**	**4.4E+05**	**2.7E+06**
Total	**6.2E+05**	**1.2E+06**	**8.0E+06**

Note: 1 pulse is 7E+12 protons. Collimated fluence (fission and capture modes) is integrated over the beam surface.

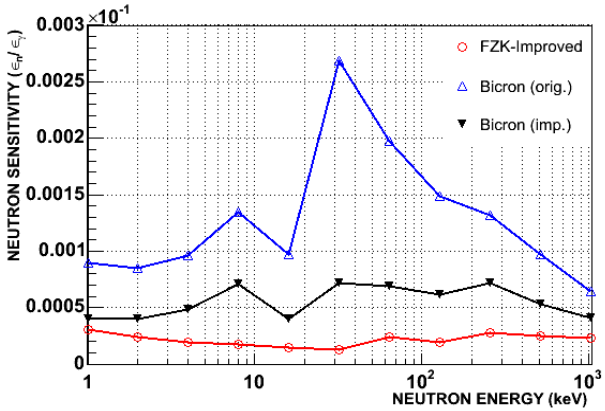
2nd collimator ϕ=1.8 cm (capture mode)



One of the best worldwide facilities for radioactive samples:
Complementary to GELINA (EU JRC-IRMM@Geel, Belgium)
In good position respect to competitors at LANSCE/ORELA(USA) at JPARK (Japan)

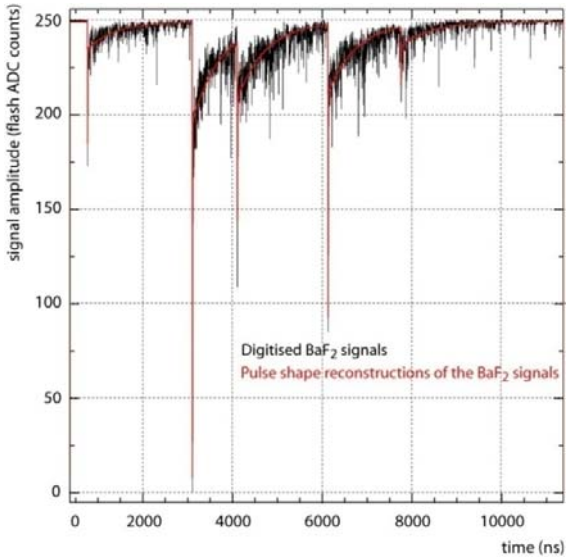
n_TOF: Advanced DAQ and detectors

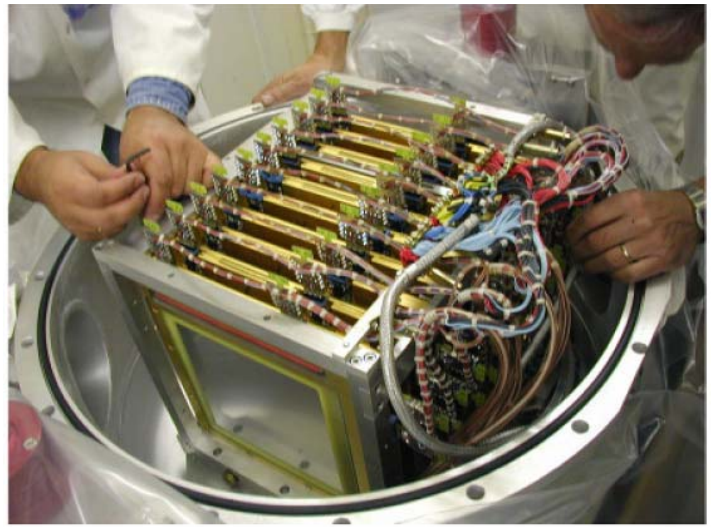
+ Precision simulations (Geant4, MCNPX, Fluka): Utilization and Validation



Low n-sensitivity capture detectors

Total Absorption Segmented Calorimeter





First FADC DAQ for T.o.F facilities

Fission detector reconstructing F.F. trajectories

n_TOF experiments 2000-2004 measurements

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

Sensitivity analysis

NSE 146, 13–50 (2004))

NEA/WPEC-26 (2008)

NUDATRA Deliverable D5.11 of IP-Eurotrans (2009)

Other types of reactor & cycles (Th-U, PWR)

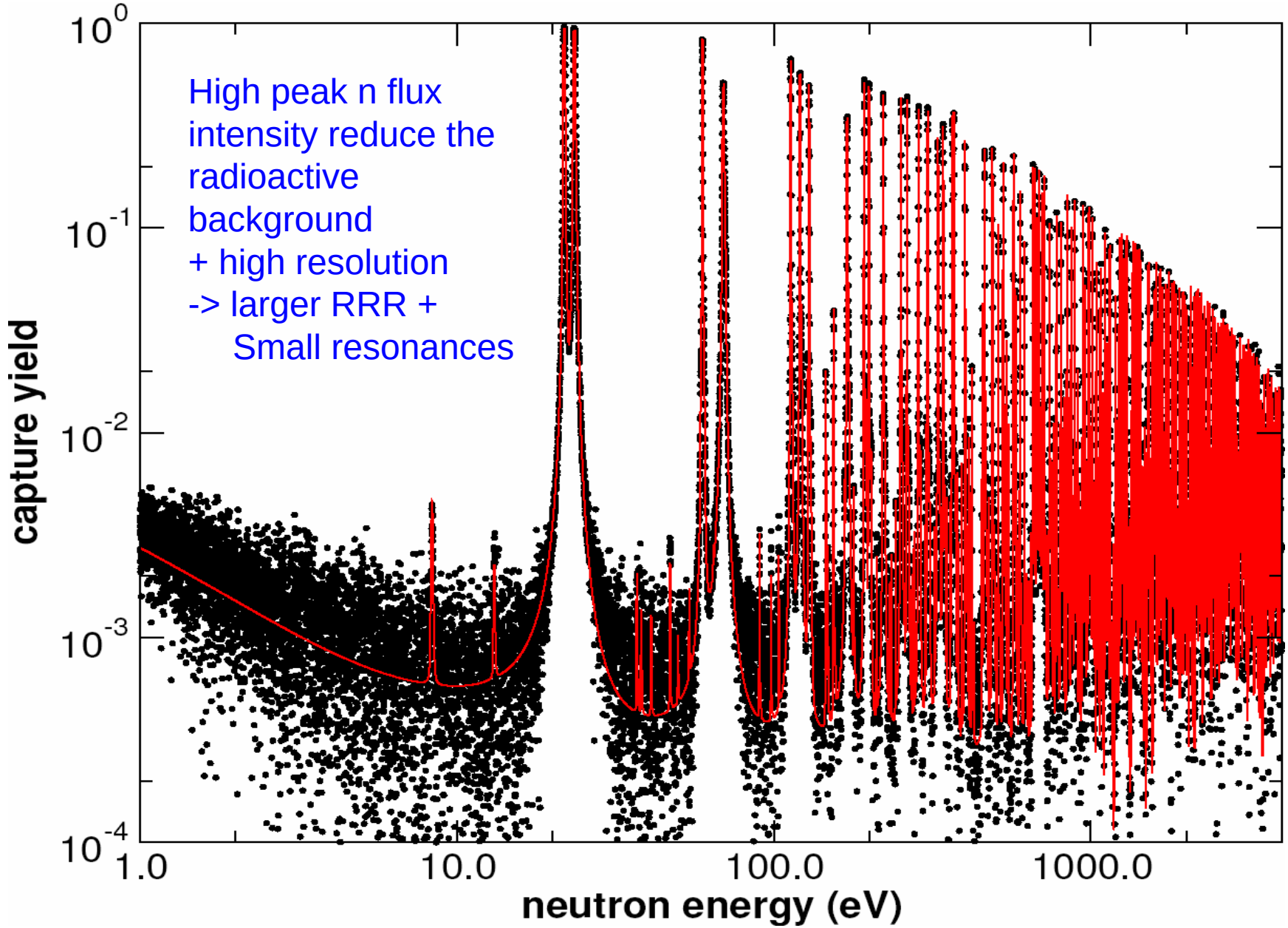
Challenge for the first n_TOF campaign:
- To improve the quality of previous measurements
- Demonstrate feasibility of challenging isotopes

All data first published then stored in the EXFOR database

n_TOF measurements are designed to obtain the maximum information for basic nuclear physics

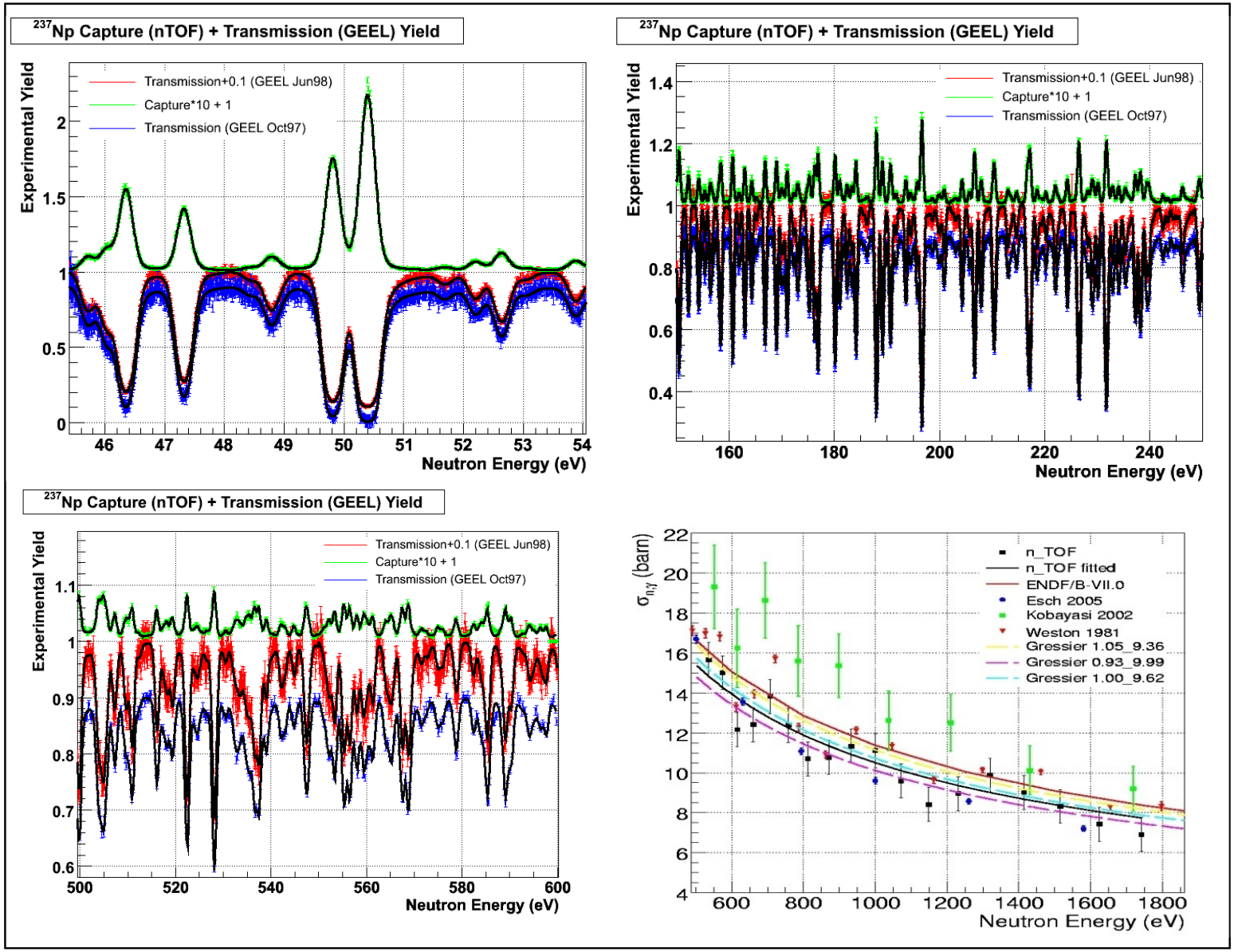
- **Nuclear structure models**
  - Improving the accuracy and statistical information from resolved resonances (RR)
    - Extending the RR region
    - Level densities and criteria for the estimation of missed resonances
  - Photon Strength functions from the TAC
  - Direct vs. compound nuclei mechanism
  - Measurements in closed-shell nuclei and light nuclei (Pb, Mg,…)
- **Fission: towards a better understanding of the process**
  - High resolution measurements over large energy ranges in the same setup
  - FF kinetic energy and angular distributions determination
  - Fissile (^{233}U, ^{235}U, ^{245}Cm) and Fissionable isotopes (^{234}U, ^{232}Th, …)
  - Sub-threshold, direct and multiple chance fission
  - Fine structures in the fission barriers (outer fission barrier and hyper-deformation of the fission potential)
- **Basic reactions**
  - n-n scattering by ^{2}H(n,np)n
  - (n, l.c.p.) reactions (l.c.p. = light charged particles like p, α, ^{3}H, Li,…)

High resolution low backgr. of radioactive samples: ^{232}Th by C_6D_6



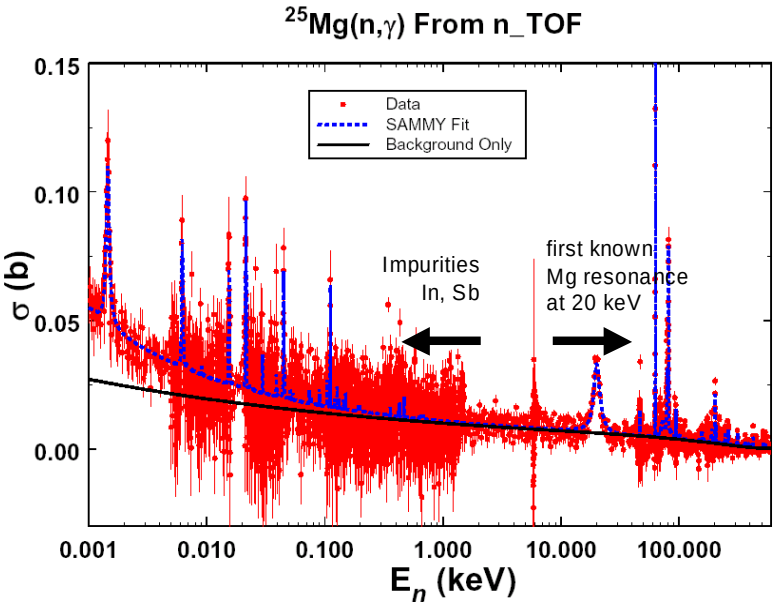
High resolution low backgr. of radioactive samples: ^{237}Np TAC

n_TOF capture + GELINA transmission = One of the best measur. made at Europe.

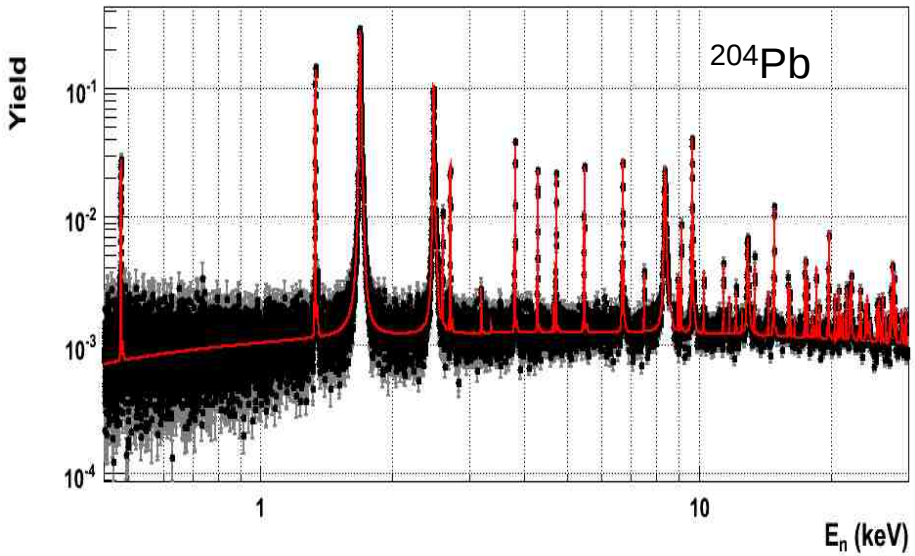


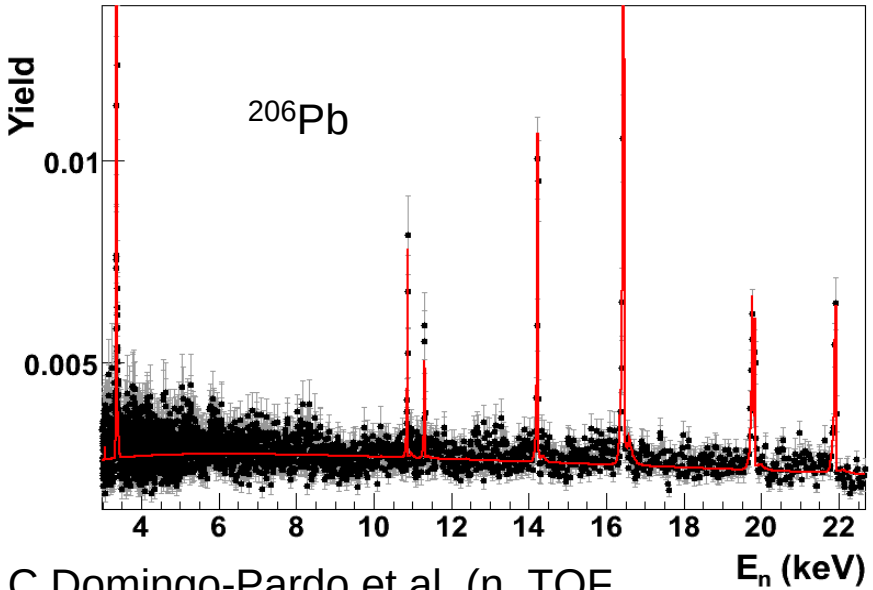
C. Guerrero et al. (n_TOF Collaboration), Proc. Int. Conf. Nuc. Data for Sci. and Tech. 2007, Nice.

High resolution low capture cross section samples: C_6D_6 + TAC



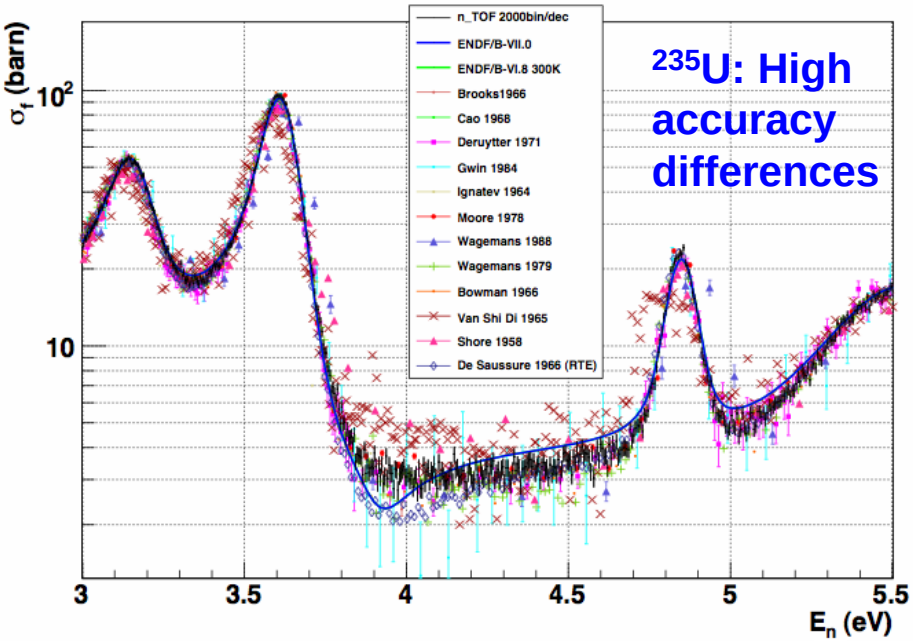
M.Heil (FZK), Nuclei in the Cosmos IX, Geneva 2005

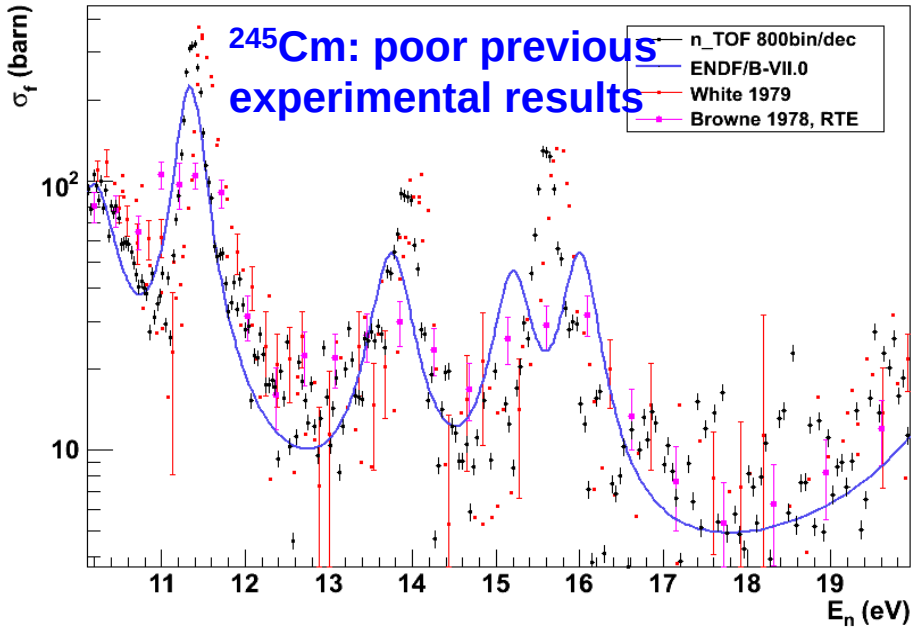


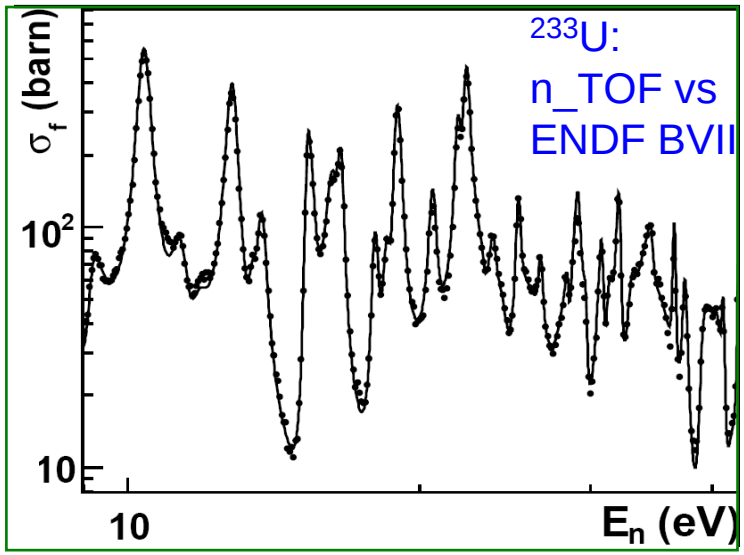


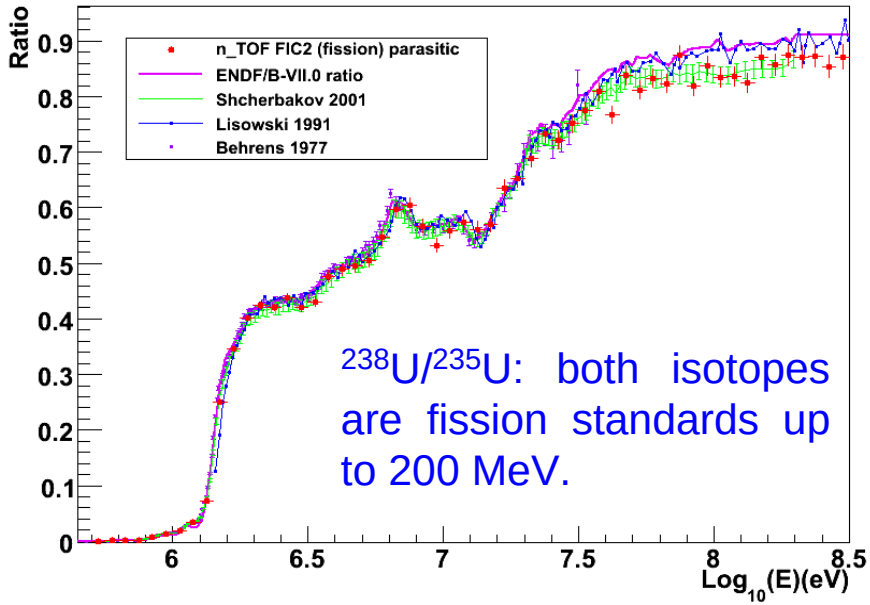
C.Domingo-Pardo et al. (n_TOF Collaboration), Phys. Rev. C 74/75, 2006/7

High resolution and large energy range accurate fission data



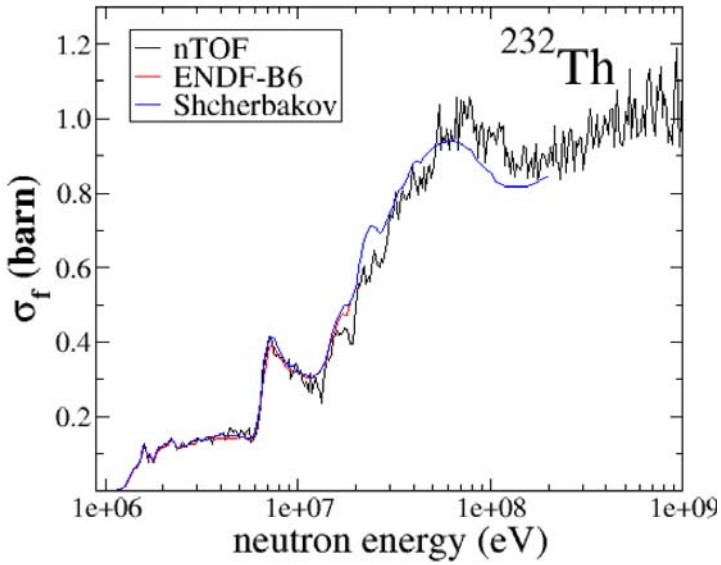


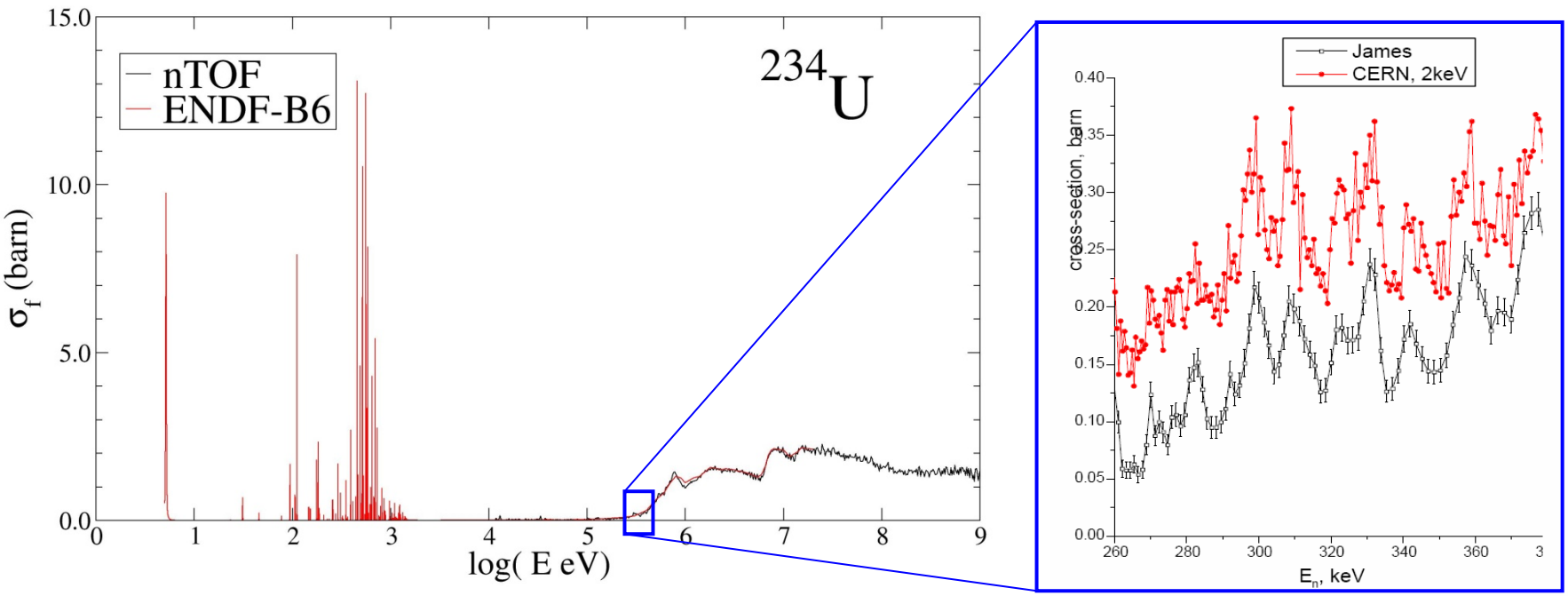




High resolution and large energy range accurate fission data

Fine structures in the fission barriers (outer fission barrier and hyper-deformation of the fission potential)





n_TOF_ph2 experiments
Current program

Reaching required accuracy indicated by the sensitivity analysis : (5-10%) M.A. and (2%-5%) for main isotopes.

Capture

Stable Isotopes:

Mo, Bi: Materials for fuel matrix (Mo) and coolant (Bi)

Fe, Ni, Zn, ^{79}Se: Structural materials

^{234,236,238}U, ^{231}Pa: Th/U fuel cycle

^{239,240,242}Pu, ^{241,243}Am, ^{245}Cm: transmutation of minor actinides

Fission

^{231}Pa, ^{234,235,236,238}U : Safety and sustainability of nuclear energy

^{241}Pu, ^{241,243}Am, ^{244}Cm, ^{245}Cm : transmutation of minor actinides

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

Other

n-n scattering by ^{2}H(n,np)n

(n, lcp) (lcp = light charged particles like p, α, ^{3}H, Li,…)

Sensitivity analysis

NEA/WPEC-26 (2008)

NUDATRA Deliverable D5.11 of IP-Eurotrans (2009)

Other types of reactor & cycles (Th-U, PWR)

n_TOF_ph2 experiments

Main upgrades from n_TOF

- New target, target cooling station and ventilation system (improving safety and reliability)
- New fission detectors to measure more physical magnitudes (angle, kinetic energy)
- New capture samples design for the calorimeter with lower beam scattered background
- The possibility to have independent moderator and cooling circuits:
    - Moderation by borated water to reduce in-beam γ background.

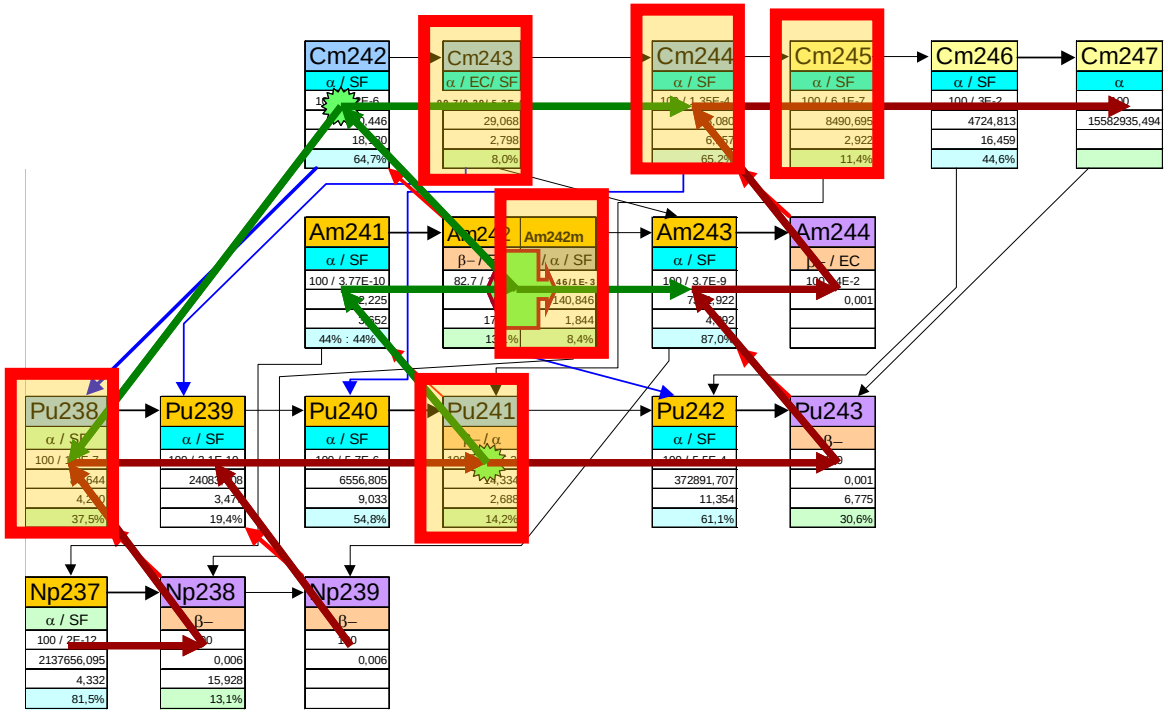
Further upgrades ahead:

- convert EAR1 to Class A/B Rad. Laboratory
- Building a new short flight path and the associated experimental area EAR2 (also expected to be Class A Rad. Laboratory)

Most measurements proposed before can be done with the facility as it is (3 first upgrades), however some fission targets are conditioned by R.P. rules and will require to upgrade the Experimental Area to a Class A/B Rad. Laboratory.

The next frontier: n_TOF @ EAR2

Actinides with very short half life (10-200 yr): **^{238,241}Pu, ^{242m}Am, ^{243,244}Cm**



- These isotopes are key steps for the nuclear waste breeding, but their radioactivity makes their measurement very difficult.
- Very low mass samples (<<1 mg): to reduce the radioactivity induced background and to be compatible with R.P. rules.
- Same conditions allow very rare materials (even deposits from rad beams ISOLDE?), and materials of very low cross section:

^{90}Sr, ^{79}Se, ^{126}Sn, ^{147}Pm, ^{135}Cs: long lived FF

The next frontier: n_TOF @ EAR2 and SPL/PS2

Improving the Signal to noise ratio for very small samples:

(1) Radioactivity background
- High brightness (peak flux intensity) and low duty cycle
- Shorter flight path 1/10 -> 100 times larger flux (EAR2)
- Higher pulse intensity (SPL/PS2)

(2) Scattered beam background
- Very thin sample support no encapsulation
  Class A laboratory (EAR2)
- Distance from samples to walls (EAR2)

(3) Background vs. Detectors
- Low neutron sensitivity of detectors
- Improved background rejection by detectors

(4) In beam background
- Large angle of neutron and proton lines (EAR2)
- Lower proton energies (SPL/PS2)
- Optimized moderator

(5) Ambient background
- Walls distance and detector background rejection

The second n_TOF experimental area (EAR2)

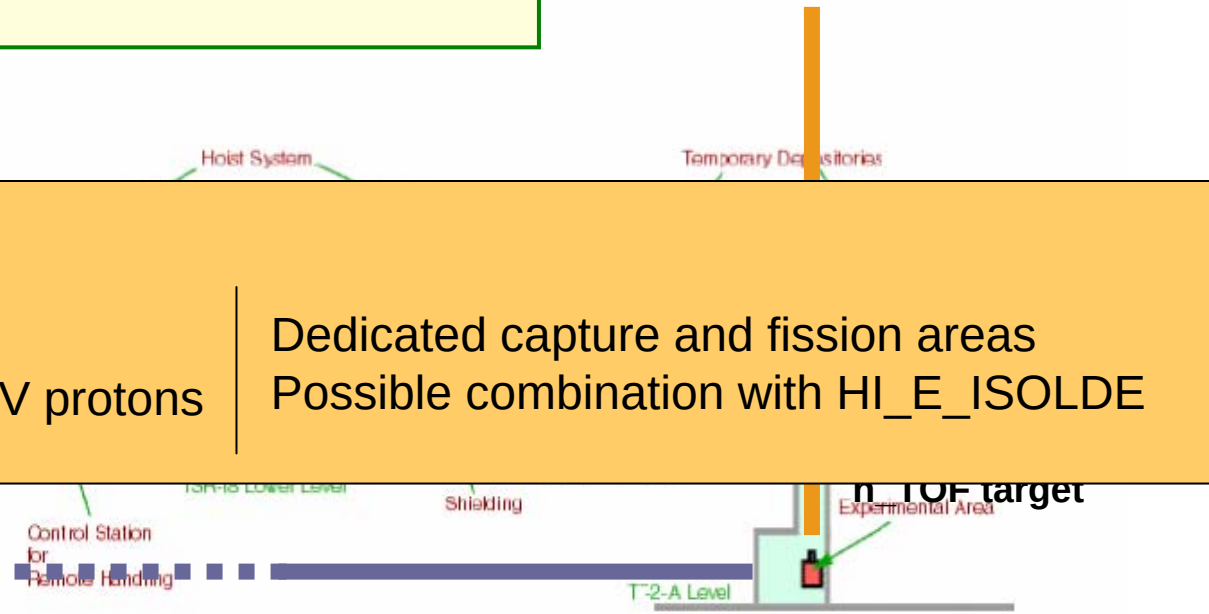
- Flight-path length : ~20 m: 100 time brighter!
- at 90° respect to p-beam direction
- Class A laboratory for minimum encapsulation
- reduced beam induced background
- Wide area for lower ambient background

^{238,241}Pu, ^{242m}Am, ^{243,244}Cm

^{90}Sr, ^{79}Se, ^{126}Sn, ^{147}Pm, ^{135}Cs

- Reducing the mass by 100 from **1mg -> 10µg**
- **Acceptable half-life from 500y -> 10y**
- **Improve accuracy and efficiency for larger samples**

New experimental area at 20 m



n_TOF @ SPL/PS2:
- Further increase of brightness
- Lower background 20 -> 4 GeV protons

Dedicated capture and fission areas
Possible combination with HI_E_ISOLDE

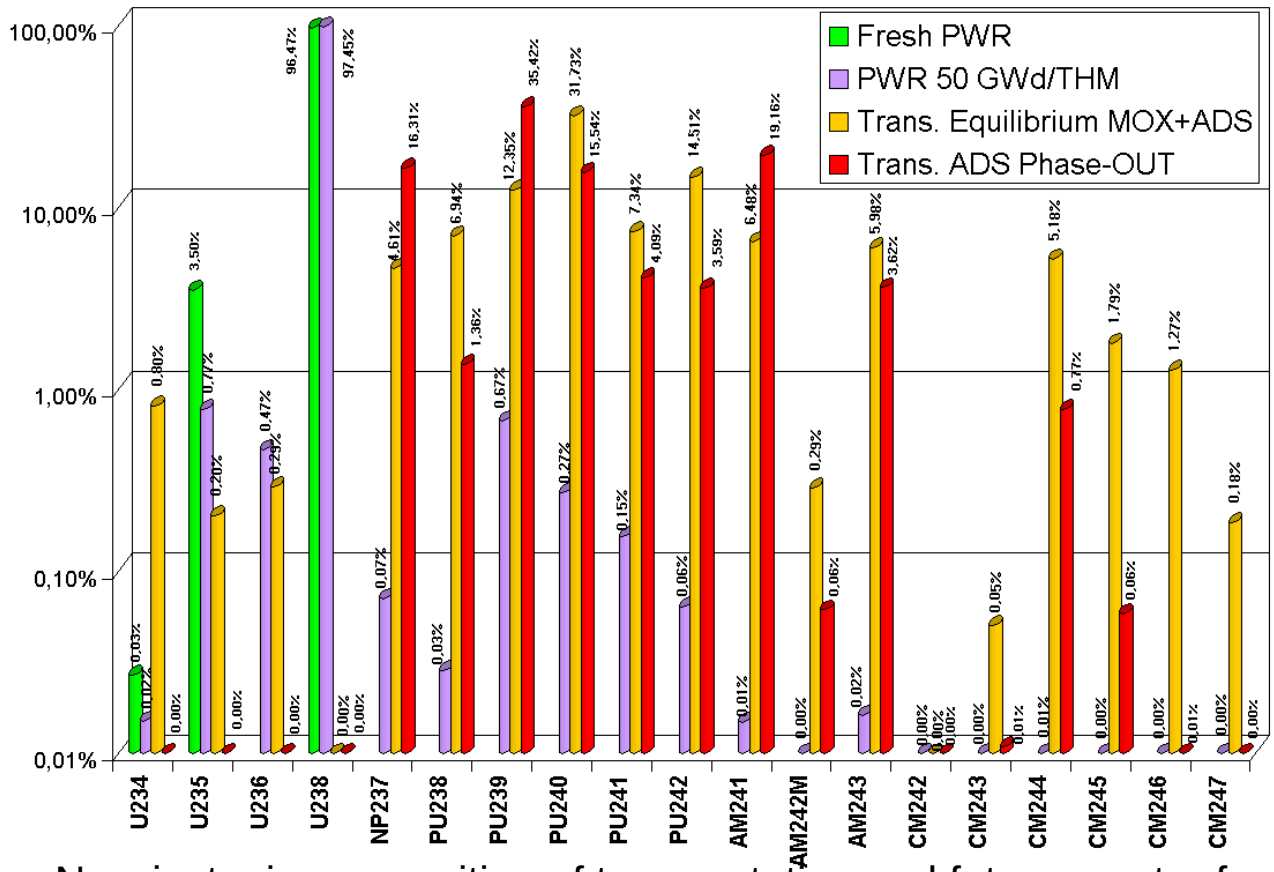
Experimental area at 185 m

Summary and conclusions

- n_TOF @ CERN is a first class neutron Time Of Flight facility
- It is specially well suited for radioactive materials, samples of rare materials or low cross section.
- Excellent facility for measuring neutron capture and fission cross sections and the most needed cross sections identified for nuclear applications (nuclear waste minimization). Sustained support from the EU framework programs.
- The measurements provide very relevant parameters to improve the understanding and physics models of nuclides and reactions.
- Combined with high performance detectors and DAQ allows to perform high accuracy cross section measurements.
- The n_TOF potentiality was proved by successful operation from 2000 to 2004
- The current campaign, with improved setup, will allow to fully exploit its possibilities to fulfill the request of the highest priority nuclear data needs
- The collaboration proposes to enhance the performance with an additional short flight path and EAR2 that will allow to open a new frontier of sample masses, short lived isotopes and accurate measurements
- We also propose the exploration of the possibilities of the future SPL/PS2 for further improvements by reaching higher neutron source brightness.

	Production	Flight path [m]	E range [eV]	Repetition rate [Hz]	Pulse width [ns]	Φ_n [cm^{-2} dec^{-1} s^{-1}]	Φ_n [cm^{-2} dec^{-1} $pulse^{-1}$]
n_TOF	Spallation	185	10^{-1}-10^{8}	<0.4>	6	10^{5}	$2.5\cdot10^{5}$
LANSCE	Spallation	20	1-10^{8}	20	250	$5\cdot10^{5}$	$2.5\cdot10^{5}$
ORELA	(γ,f)	40	th-$5\cdot10^{6}$	525	8	$2\cdot10^{4}$	20
GELINA	(γ,f)	30	th-$4\cdot10^{6}$	800	1	$5\cdot10^{4}$	50
n_TOF (EAR2)	Spallation	20	10^{-1}-10^{8}	<0.4>	6	$3\cdot10^{6}$	$8\cdot10^{6}$

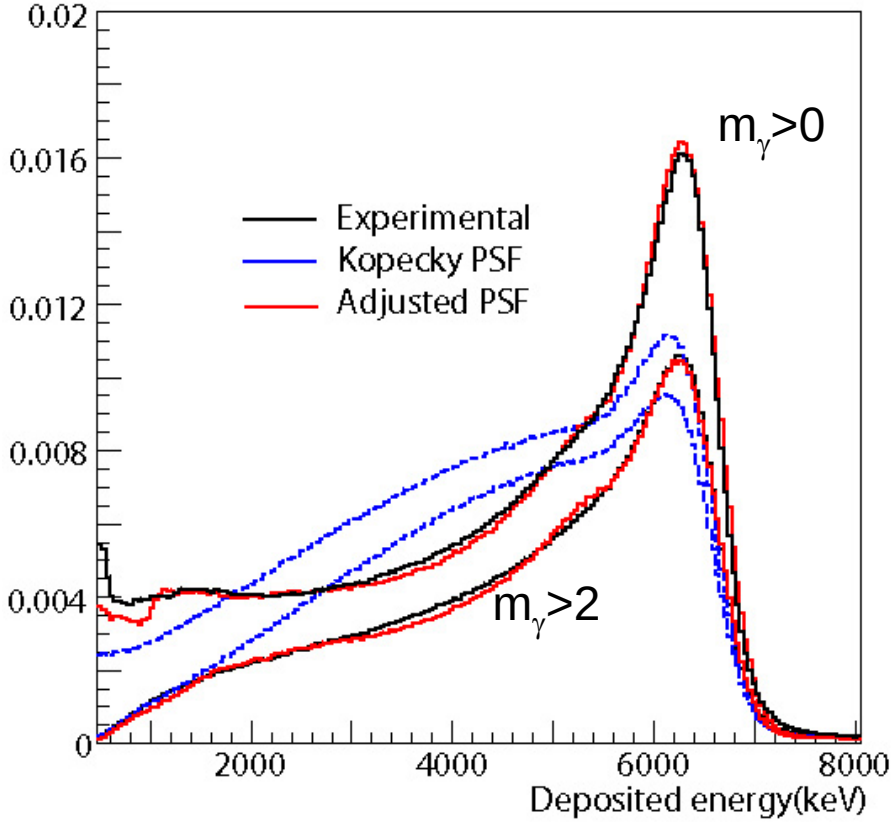
New isotopic composition of transmutation and future reactor fuels
New coolants and fuel matrices
New requirements for structural materials
New neutron spectra
High energy neutrons from the Spallation Source

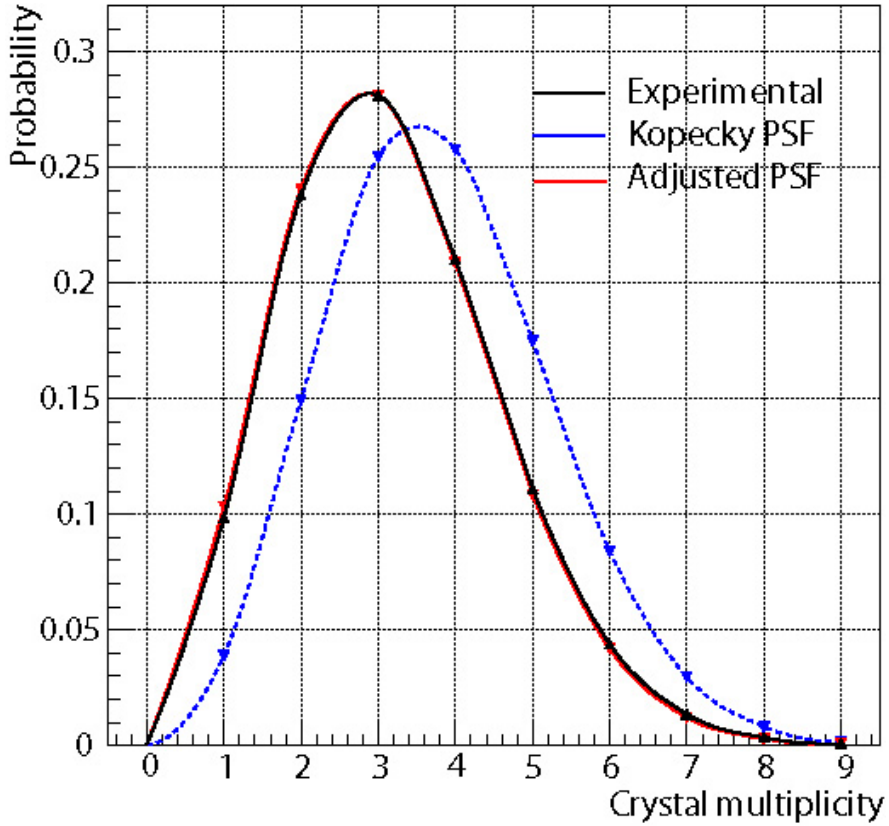


New isotopic composition of transmutation and future reactor fuels

Nuclear structure: TAC response $^{197}Au(n,\gamma)$: E_{sum} and m_γ

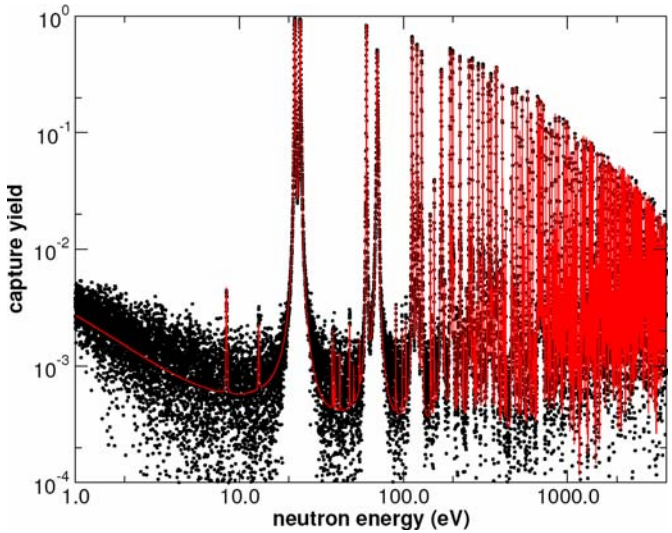
Validation of Adjusted Photon Strength Functions models with $^{197}Au(n,\gamma)$ TAC data for deposited energy (with and without conditions in m_γ) but also for m_γ (E_{sum}>1.5 MeV).
Similar studies for several isotopes.



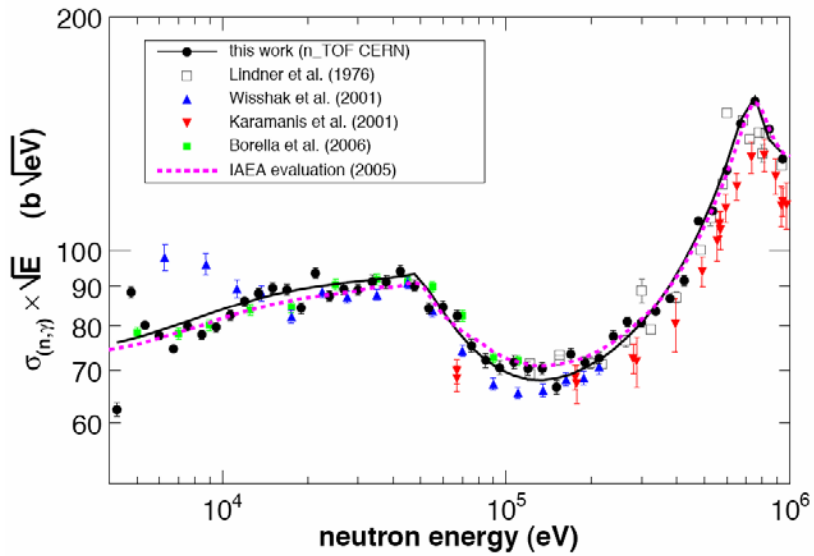


High resolution low backgr. of radioactive samples: ^{232}Th by C_6D_6

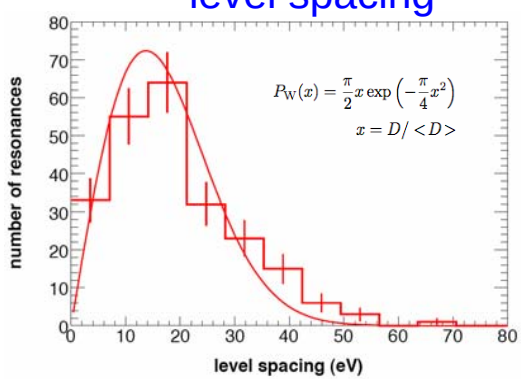
Resolved Resonance Region



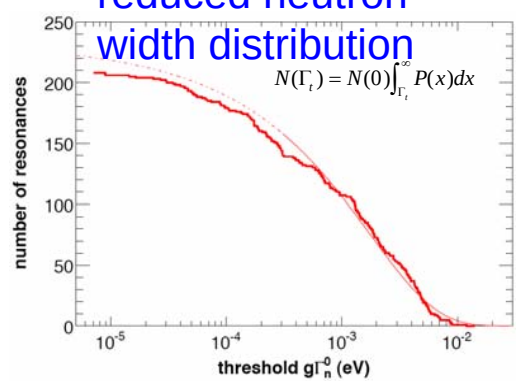
Unresolved Resonance Region



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)