Cross section measurements for thermal neutron-induced reactions on actinides at the ILL reactor

A. Letourneau

Institute of research into the fundamental laws of the Universe / Nuclear Physics Section

CEA - Saclay

The Mini-INCA project

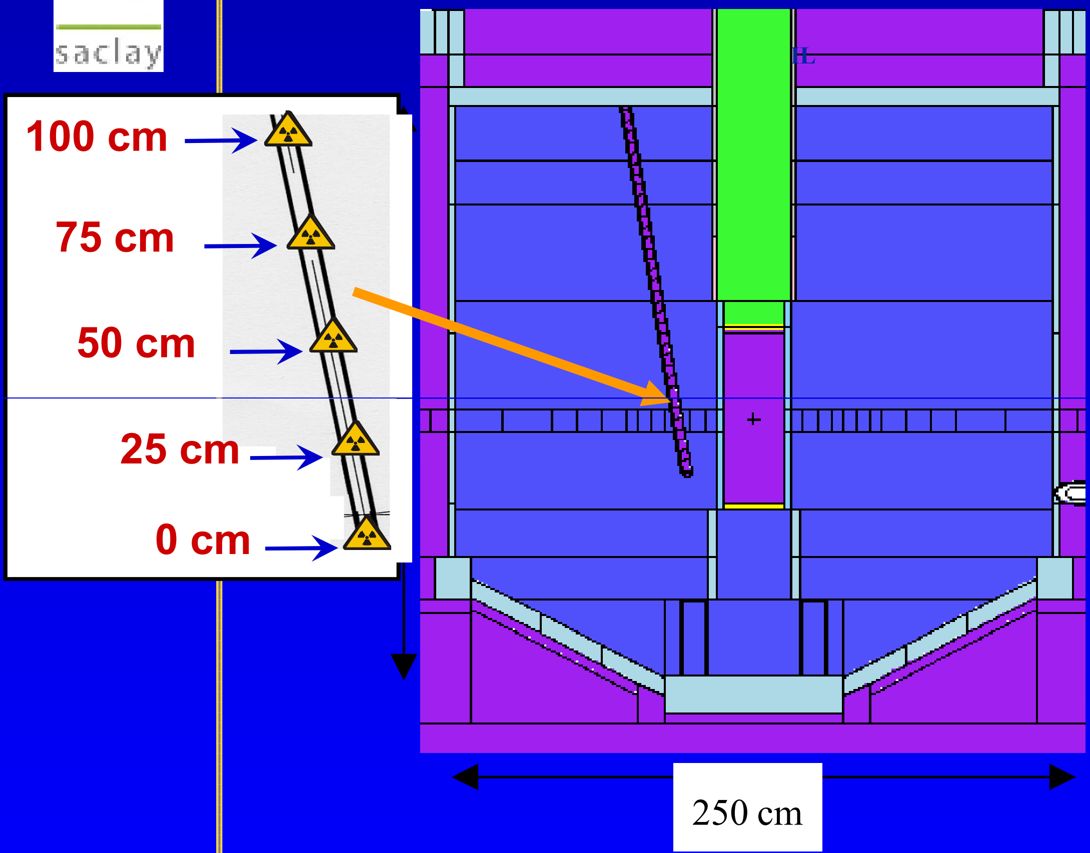
irfu cea saclay

- To provide accurate nuclear data and reduce the uncertainties on minor-actinides for:

  - thermal neutron-induced reaction cross sections
  - decay parameters for short live isotopes or isomers

They do not play a major role on reactor performances (criticality, reactivity, …), but can have potential great impacts for high burn-up or multirecycling fuel, MA transmutation in dedicated systems

Irradiation facility: HFR at ILL



CHARACTERISTICS:

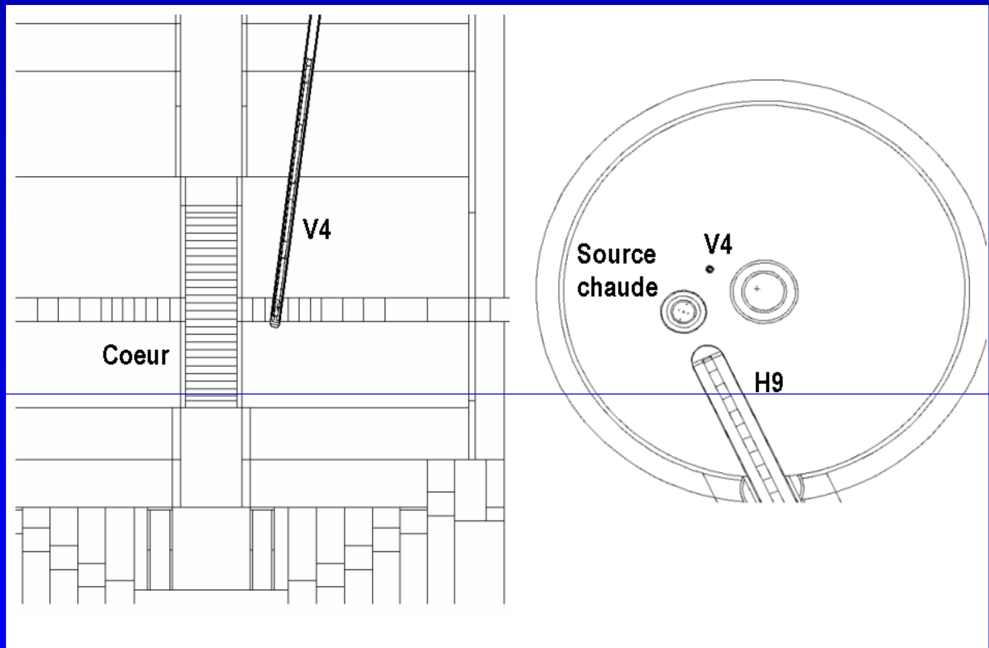
- Heavy water, highly enriched ^{235}U
- Thermal power: 58 MW
- Thermal neutrons: 6 10^{13} to 1.5 10^{15} n/cm^2/s
- Cycle: 50 days

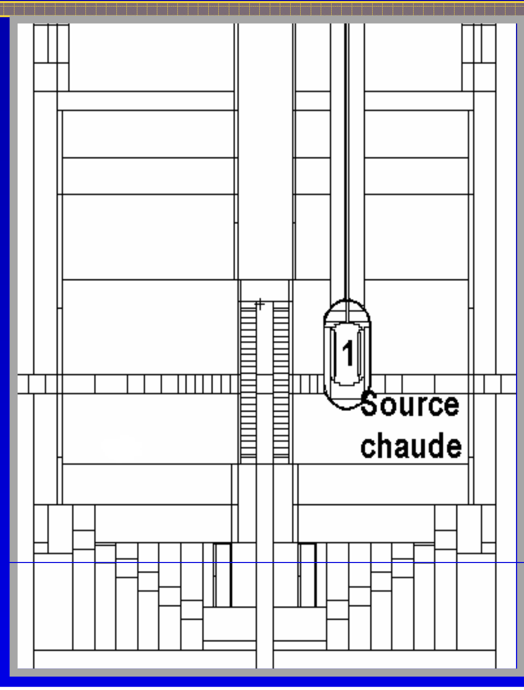
CONSTRAINTS:

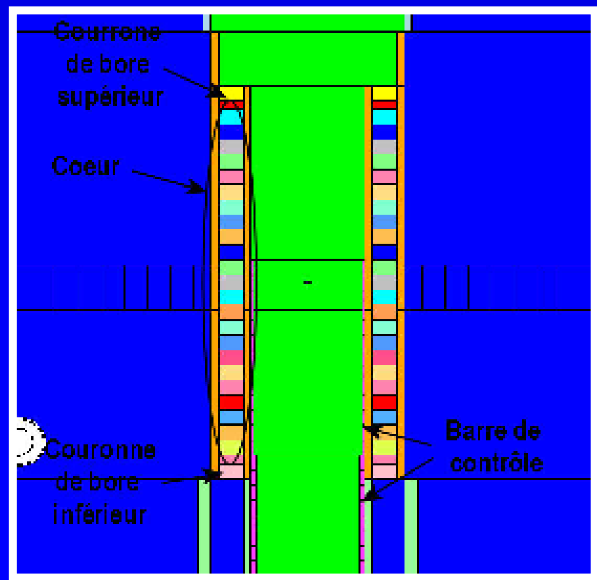
- High burn-up: 1 cycle ~ 1 year in PWR
- γ-radiations: ~ 10^{15} γ/cm^2/s

- Small mass samples (~10-20 μg)
- Low flux perturbation (negligible auto-absorption)
- Deep exploration of transmutation chains ("short-life" isotopes)

Monte Carlo simulation of the HFR

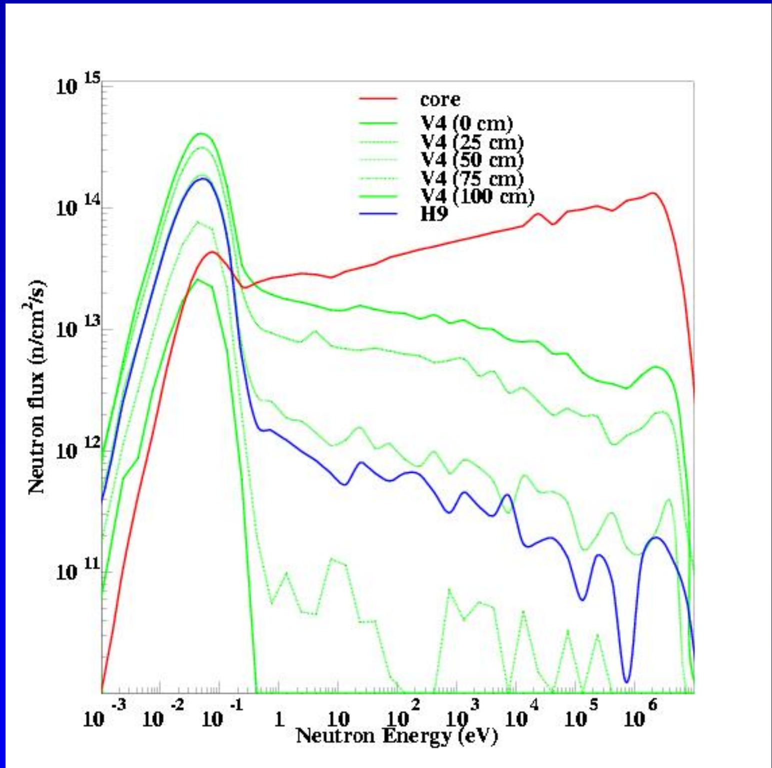


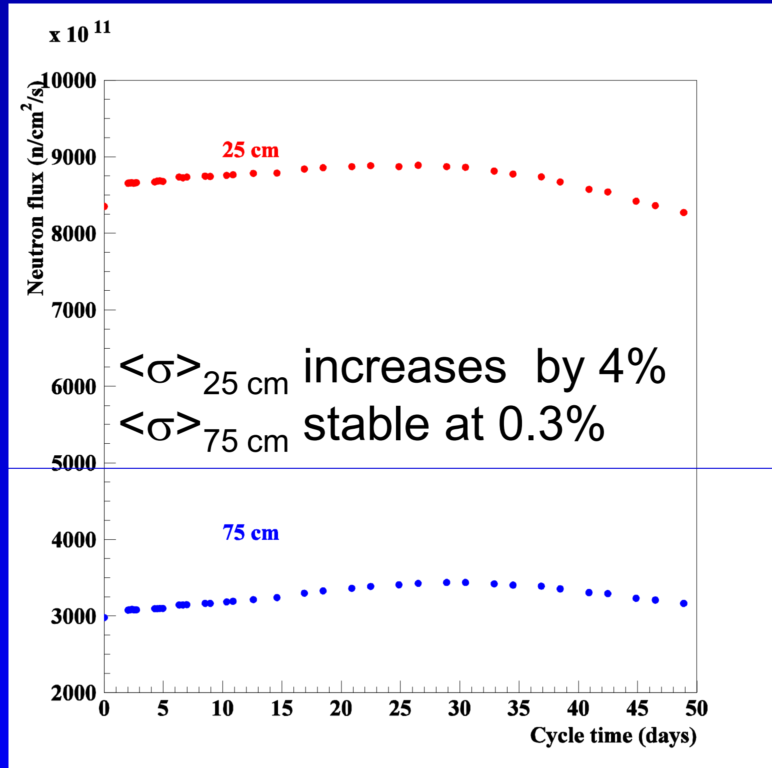




- MCNP2.5 / MURE simulation
- TRIPOLI with evolution

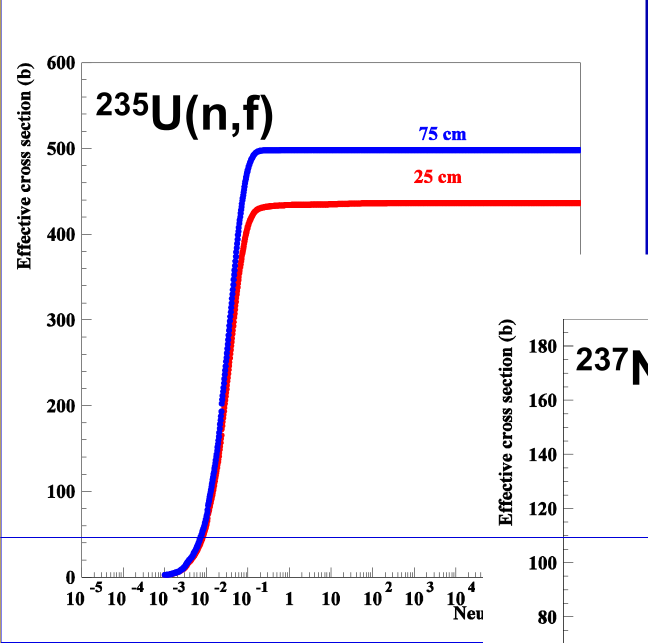
Neutronic features

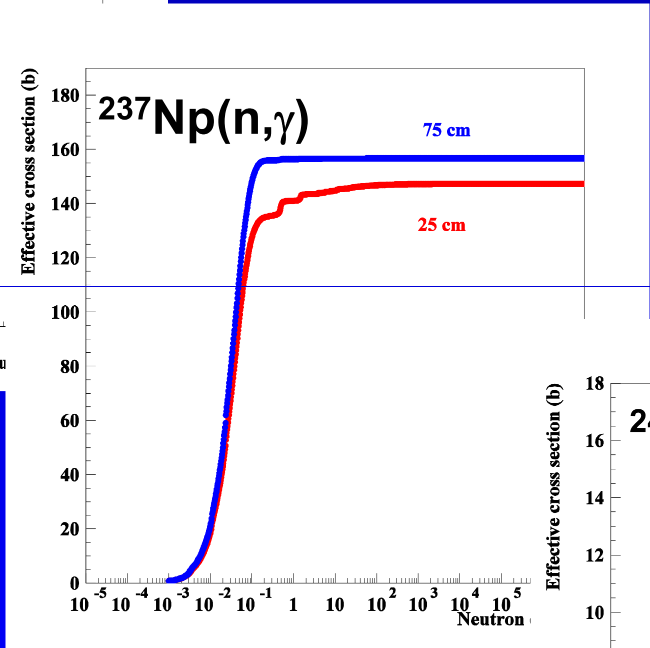


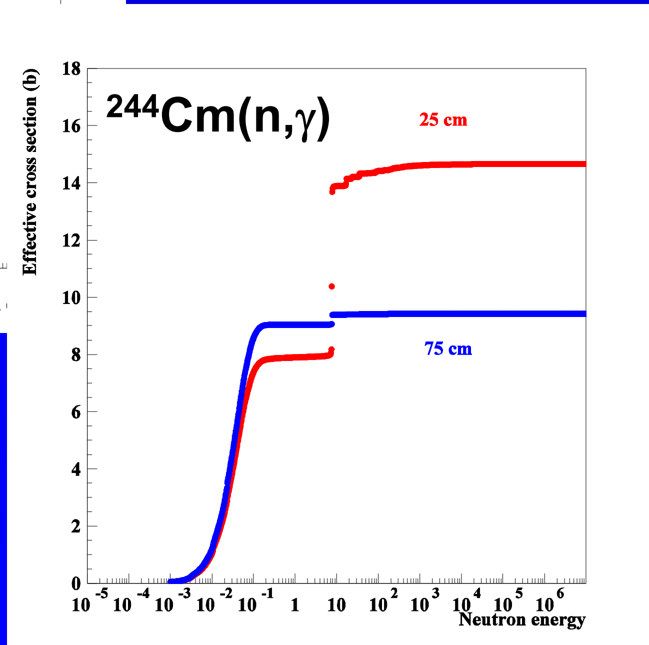


Pos	ϕ (n/cm²/s)	ϕ_{th}/ϕ	$r\sqrt{326/T_0}$
V4 (25 cm)	$8\ 10^{14}$	0.90	0.0144
V4 (75) cm	$4\ 10^{14}$	1.00	0.0006
H9	$6\ 10^{14}$	0.98	0.0021

Effect of resonances on <σ>







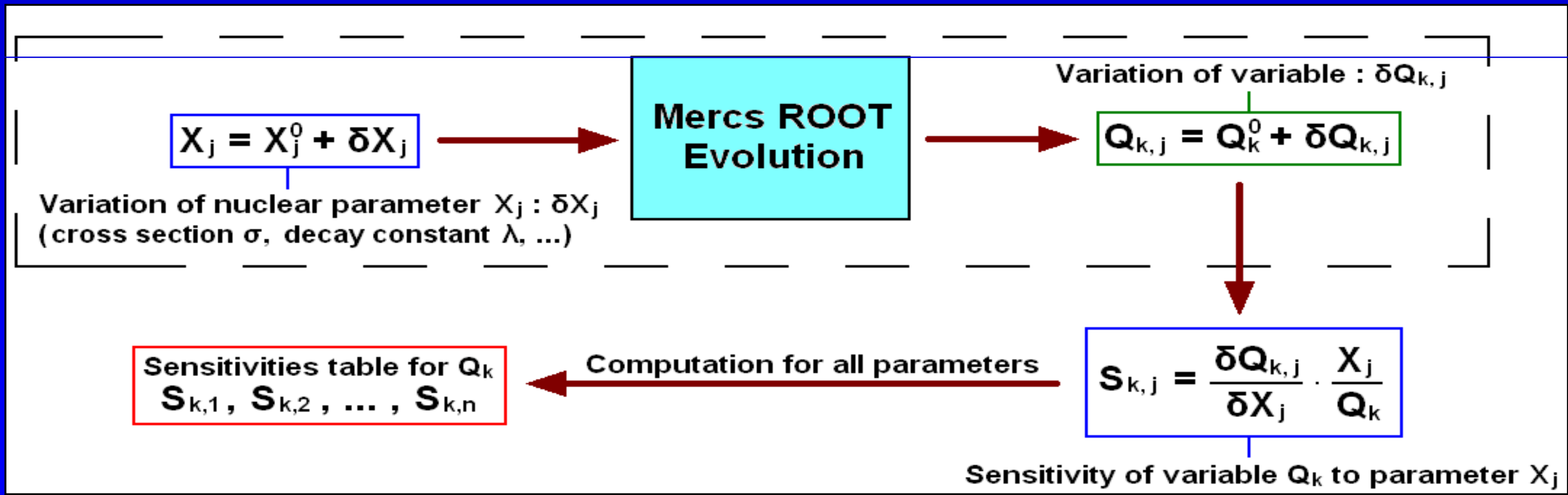
$$< \sigma > = \frac{\int_0^E \sigma(E).\phi(E)dE}{\int_0^\infty \phi(E)dE}$$

Basic principle of the experiments

- Sample characterisation prior to irradiation:
  - Mass spectrometry
  - Spectroscopy α
- Sample analysis:
  - Off-line activation techniques
    - Spectroscopy α and γ
    - Mass spectrometry (Thermal Ionisation, Induced Coupled Plasma MS)
  - On-line monitoring with fission chambers
- Normalisation:
  - $^{59}Co(n,\gamma)^{60}Co$
  - $^{235}U(n,f)$
- Data analysis with MERCS code
- Effective and 25.3 meV cross section values

ROOT MERCS module

- ROOT MERCS evolution code :
  - One dimension, one energy group evolution code
  - Compute the sensitivities of observables to nuclear parameters
  - ROOT minimization package



O. Bringer, PhD thesis, INP Grenoble (2007)

Instrumentation for (n,γ) and β-decay

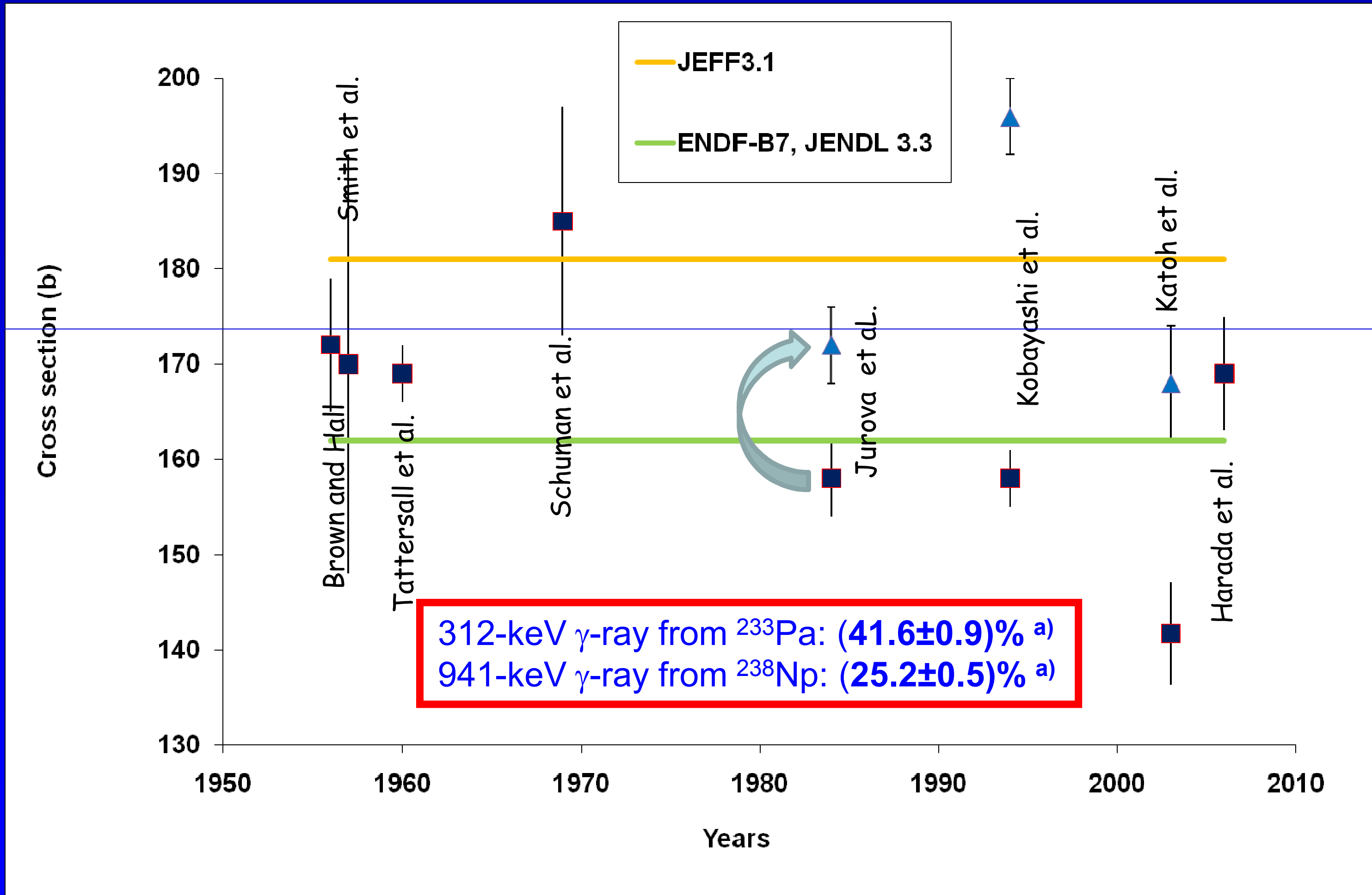
PIPS detector (100 μm)
FWHM=13 keV

from 1 to 30 cm
Count rate < 20KHz with standard fast spectroscopy electronic

HPGe coaxial detector
FWHM=1.7 keV at 1 MeV)

from 40 to 80 cm
Count rate < 80 kHz with DSP2060 Canberra
< 1 MHz with ADONIS

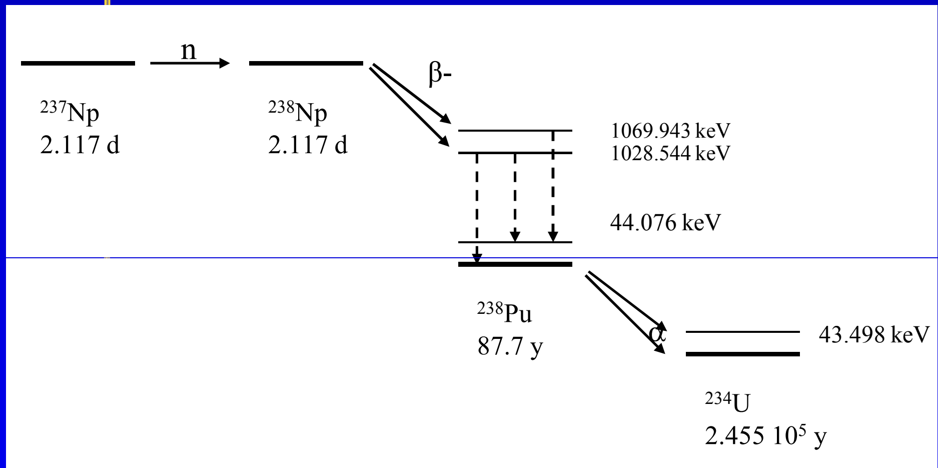
25.3 meV $^{237}Np(n,\gamma)$ cross section

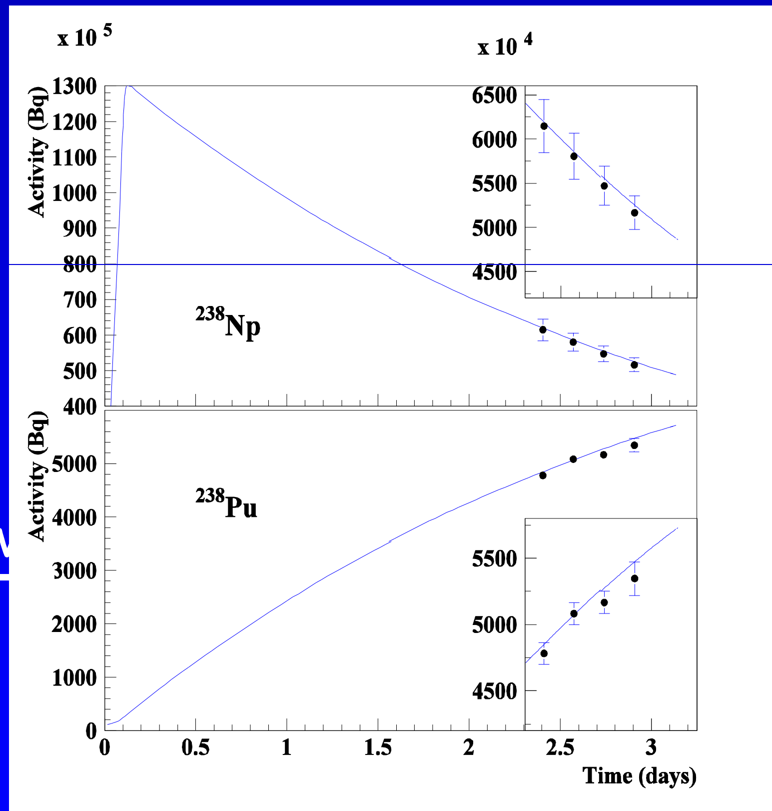


[a] Harada, H., Nakamura S., Ohta M., Fujii T., Yamana H., 2006, J. Nucl. Sci. Technology, 43, 1289.

$^{237}Np(n,\gamma)$ cross section

- $(13.52 \pm 0.14\ \mu g)$ of ^{237}Np target (99.76% purity)
- Simultaneous α (PIPS) and γ (HPGe) spectroscopy

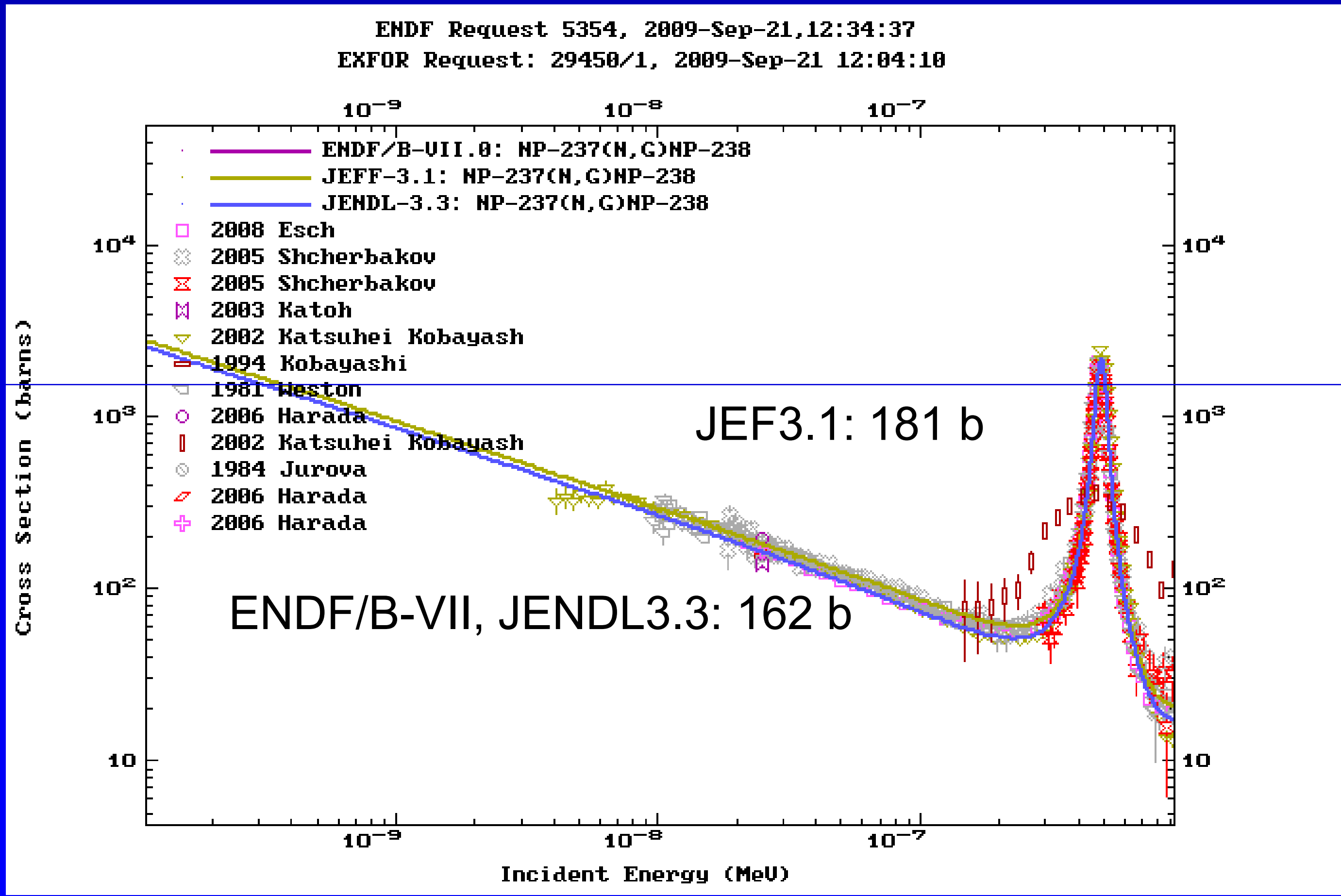




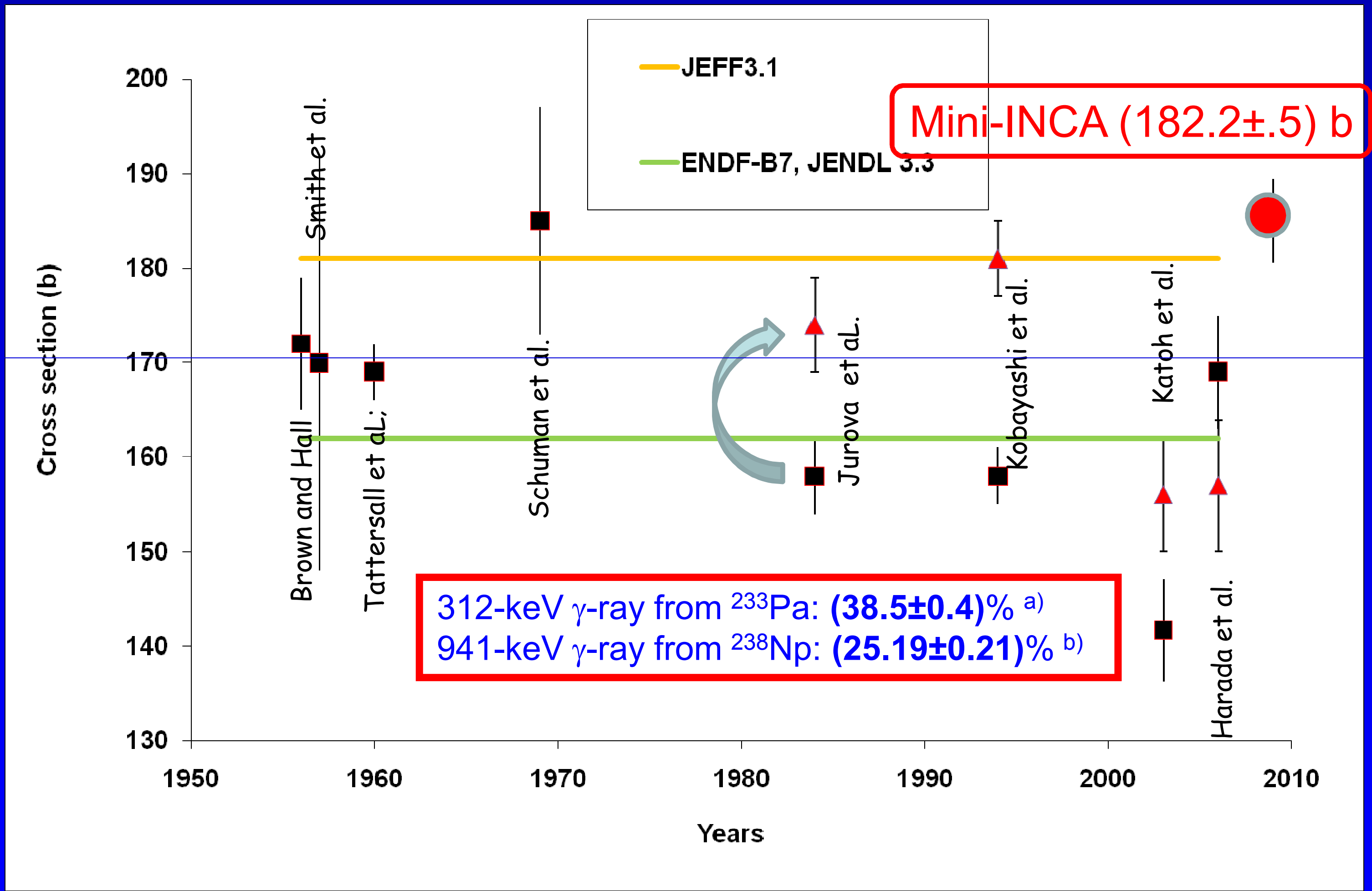
	984.45 keV	1025.87 keV	1028.54 keV
Present work	25.6±0.4	8.90±0.2	18.8±0.3
Chukreev (2002)	25.19 (21)	8.72 (15)	18.29 (23)
Harada (2006)	25.2 (5)		
Rengan (2006)	25.17 (13)	8.766 (45)	18.23 (93)
Lederer (1981)	27.8	9.7 (6)	20.3 (8)

A. Letourneau et al., submitted to App. Rad. and Isot (2009)

$^{237}Np(n,\gamma)$ capture cross section



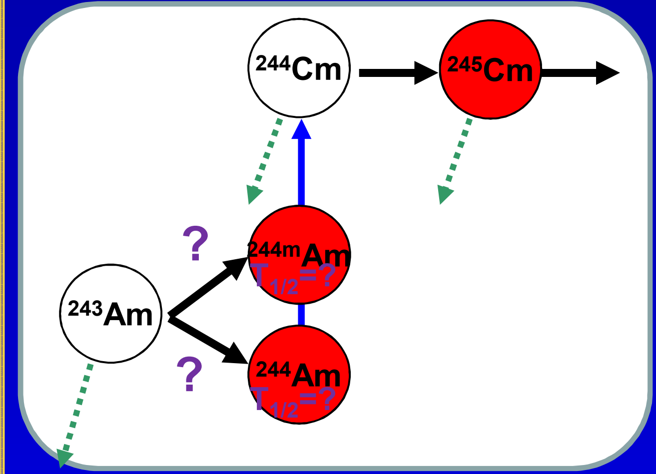
25.3 meV $^{237}Np(n,\gamma)$ cross section

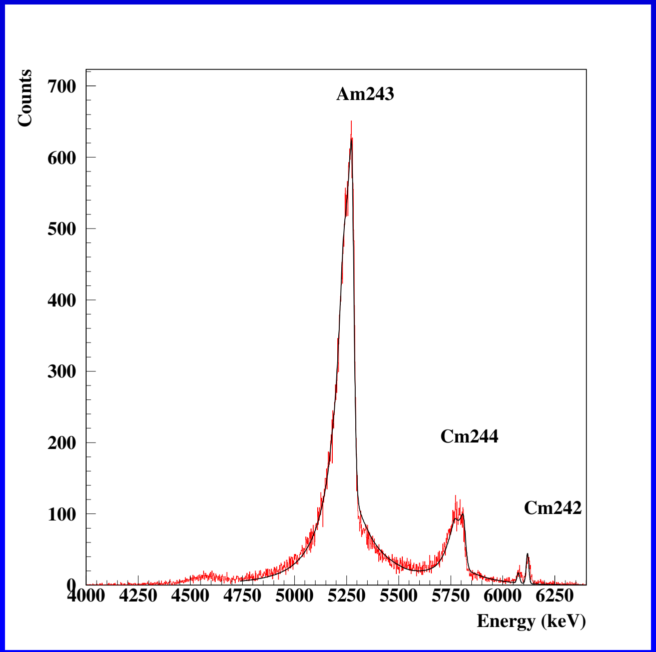


a) Singh, B., Tuli, J.K., 2005, Nuclear Data Sheets 105, 109.

b) Chukreev, F. E., Makarenko V. E., Martin M. J., 2002, Nuclear Data Sheets 97, 129.

$^{243}Am(n,\gamma)^{244gs-m}Am$ cross sections





- (10.926±0.11) μg ^{243}Am
- Two irradiations in H9:
  - (3.277±0.002) hours
  - (24.00±0.02) min
- spectroscopy γ (ADONIS) and α
  - $^{244gs}Am$ and ^{244}Cm activities
- $^{243}Am(n,\gamma)^{244tot}Am$:
  - (73.8±2.3) b
- BR :
  - (0.0474±0.0001)

F. Marie, A. Letourneau et al., NIM A 556 (2006) 547.

A. Letourneau et al., to be submitted to App Rad and Isotop

Half-lives of $^{244gs-m}Am$

- ^{244m}Am:

  $T_{1/2}$: 25-26 min

  (28.25±1.3) min

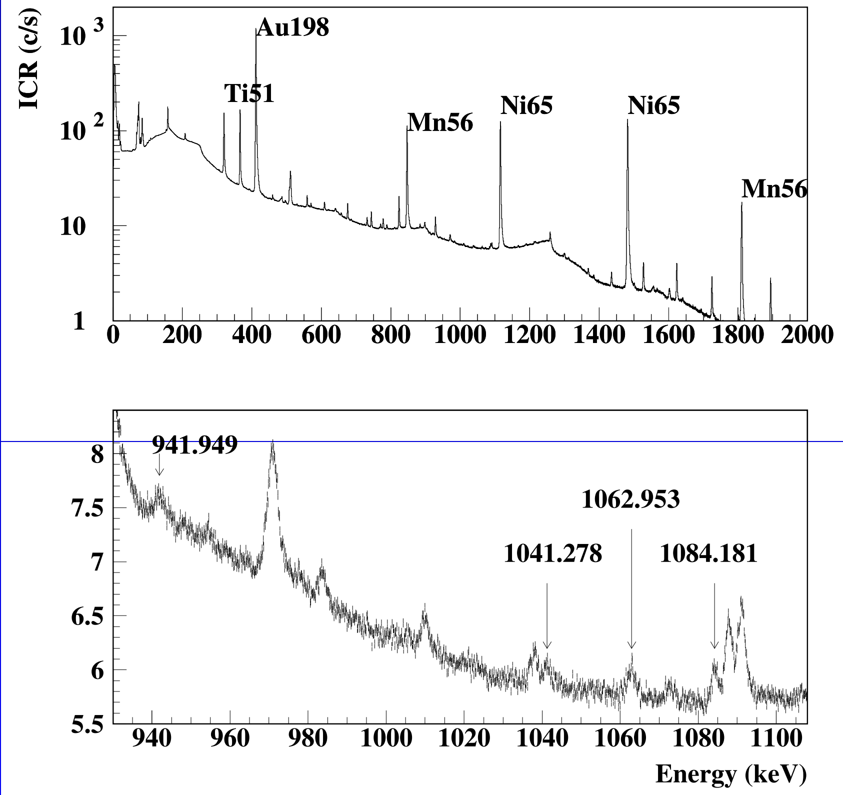
Eγ (keV)	Iγ (new)	Iγ (old)
941.949	0.16±0.011%	0.35
1041.278	0.127±0.001	0.19
1062.953	0.28±0.013	0.27
1084.181	0.30±0.016	0.35

- $^{244gs}Am$:

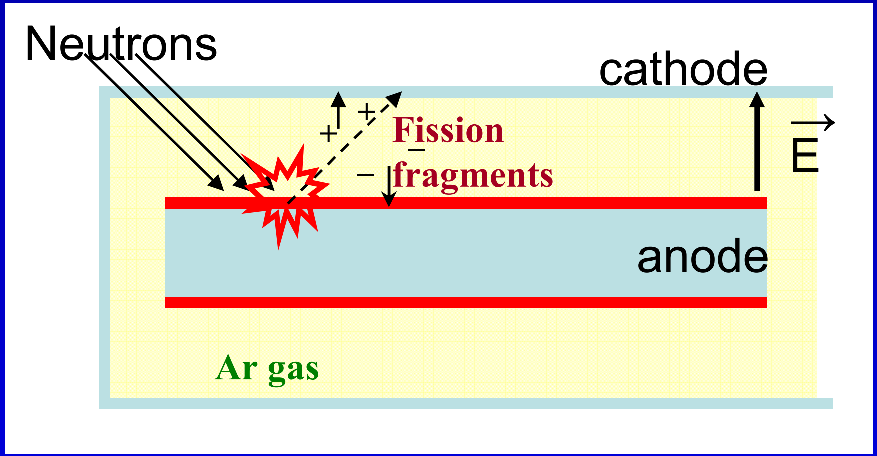
  $T_{1/2}$: 10.1±0.1 h

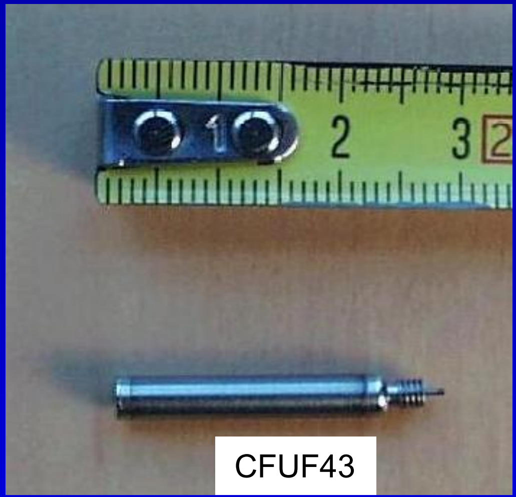
  (10.65±0.12) h

Eγ (keV)	Iγ (new)	Iγ (old)
538.400	<1.11±0.6	0.66
743.971	68.6±0.9	66
897.848	24.2±0.3	28

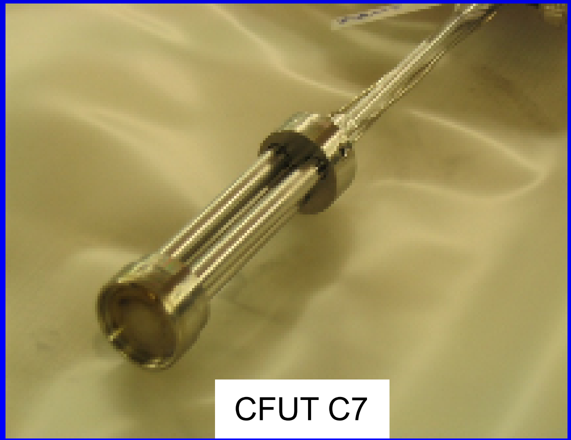


Instrumentation for (n,f) and (n,γ)



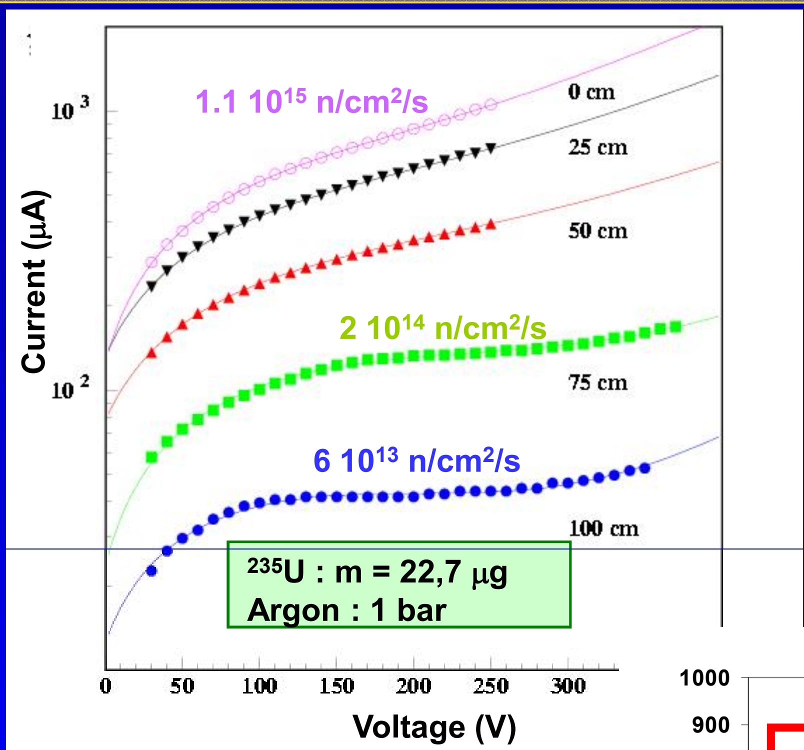


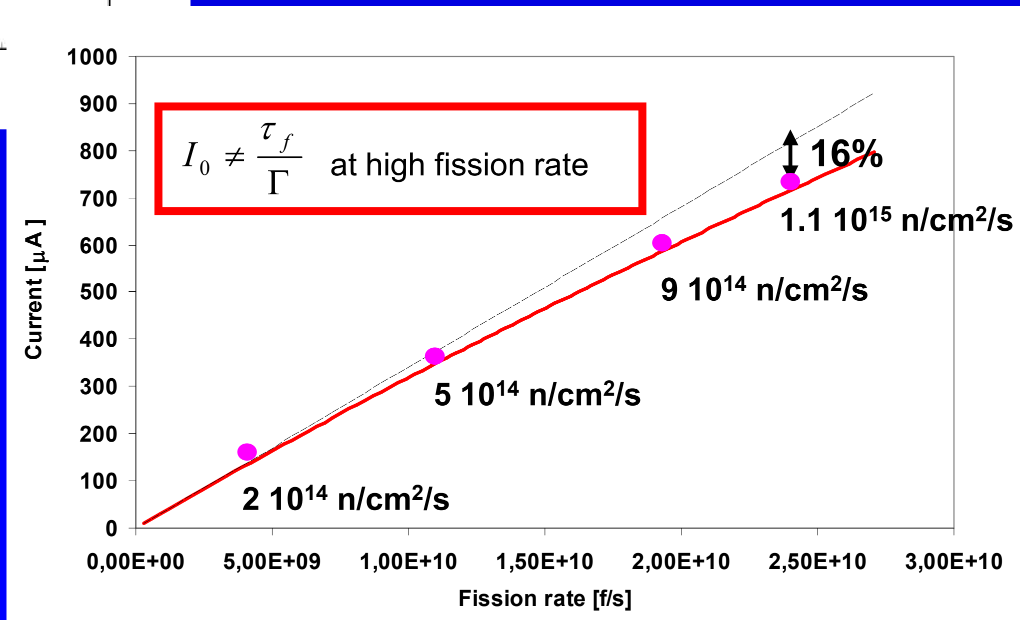
- Compensated Fission chambers (FC)
  - Minor Actinide
  - ^{235}U (reference)
  - Background



- Advantages
  - same ionisation gas
  - small inter-electrode gap to reduce space charges

Response of FC in high fluxes

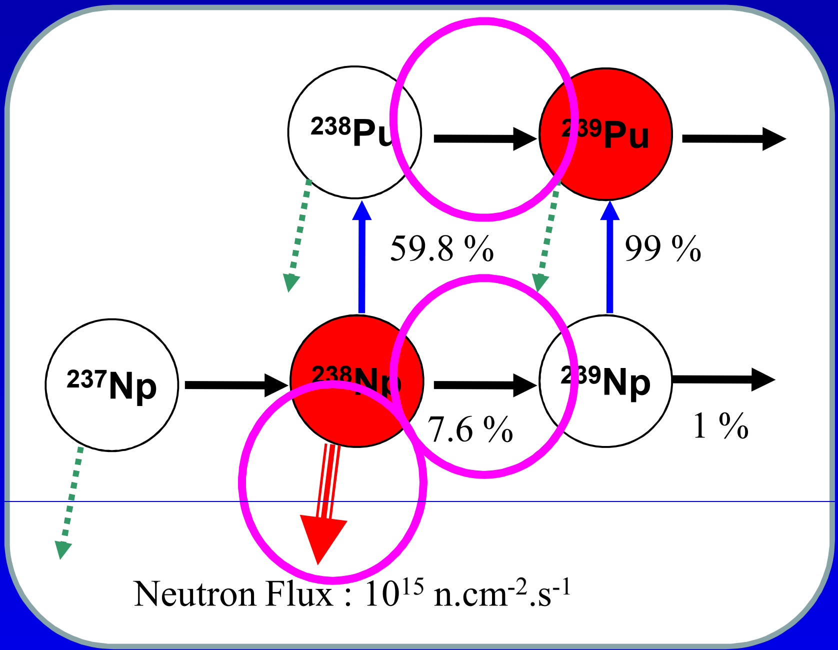




S. Chabod et al,
Modelling of fission chambers in current mode – analytical approach,
NIM A 566 (2006) 633-653

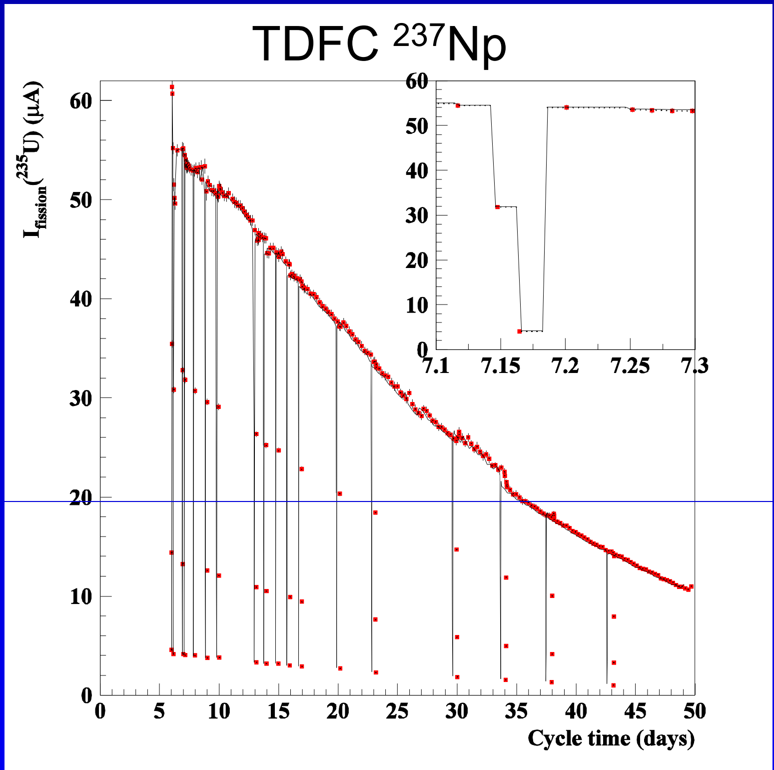
A. Letourneau et al.,
Recent developments on micrometric fission chambers for high neutron fluxes, ANIMMA09 conf proceedings,
to be published in IEEE.

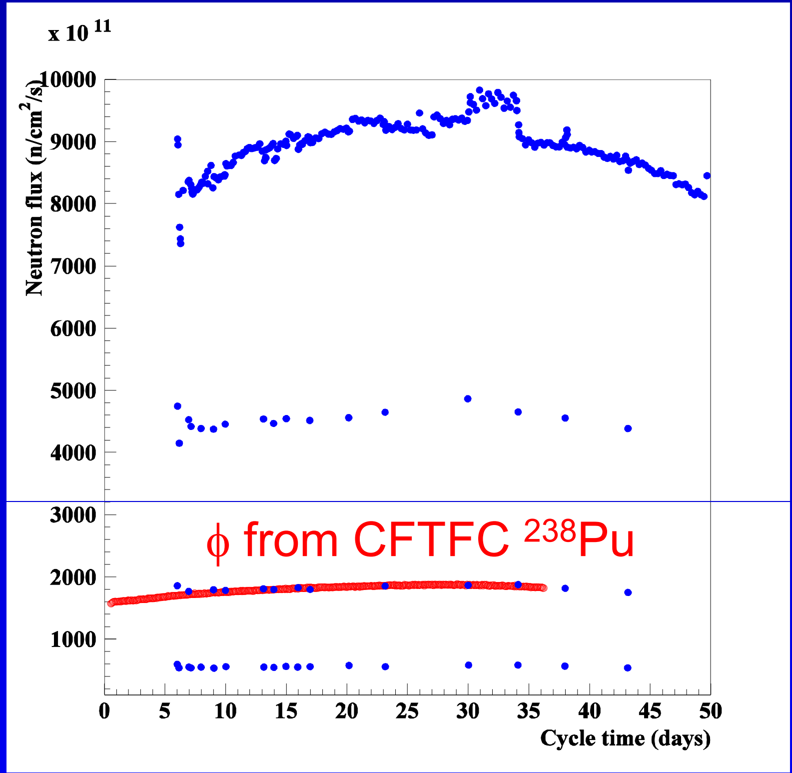
$^{238}Np(n,f)$ and $^{238}Pu(n,\gamma)$



- TDFC ^{238}Pu irradiated in 75 cm
  - (42.6±1.6) μg of ^{238}Pu (4.978% of ^{239}Pu)
  - (4.48±0.01) μg of ^{235}U
- TDFC ^{237}Np irradiated in 25 cm
  - (42±1.3) μg of ^{237}Np
  - (2.64±0.01) μg of ^{235}U

Neutron flux reconstruction

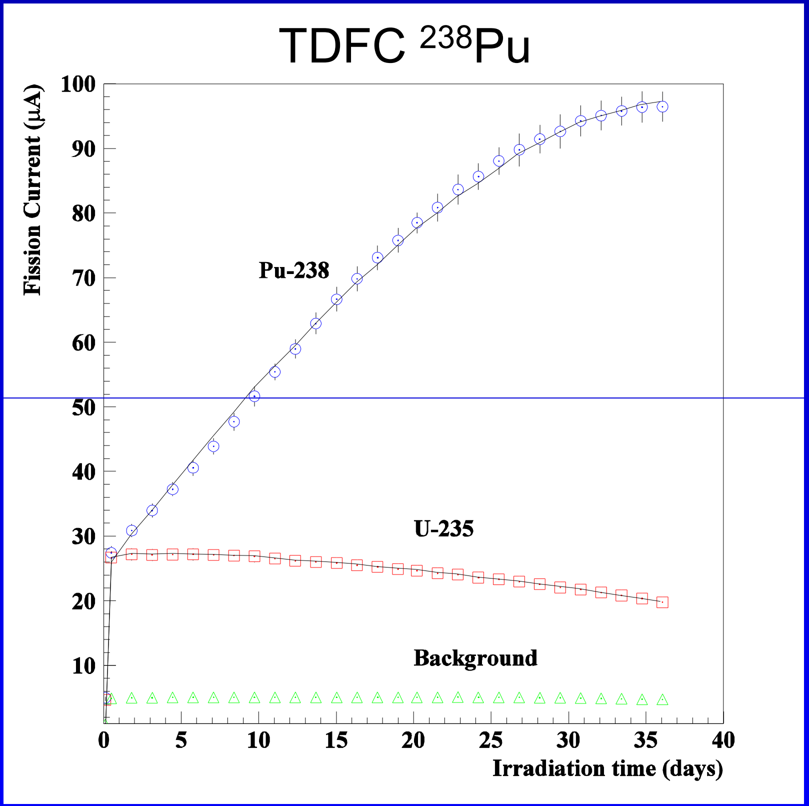


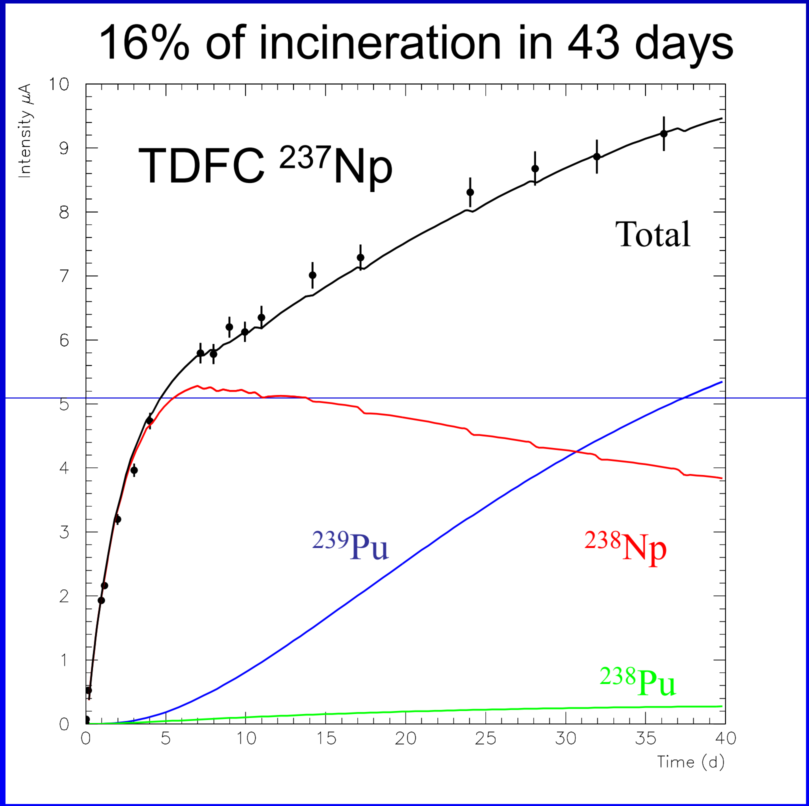


$$I(t) \propto \langle \sigma_f \rangle \phi(t)\, e^{-\int_0^t \langle \sigma_f + \sigma_c \rangle \phi(t)\, dt}$$

$$\Phi(t+dt) = \frac{I_{U5}(t+dt)}{I_{U5}(t)} \frac{\Phi(t)}{1 - \overline{\sigma_a}\Phi(t)dt}$$

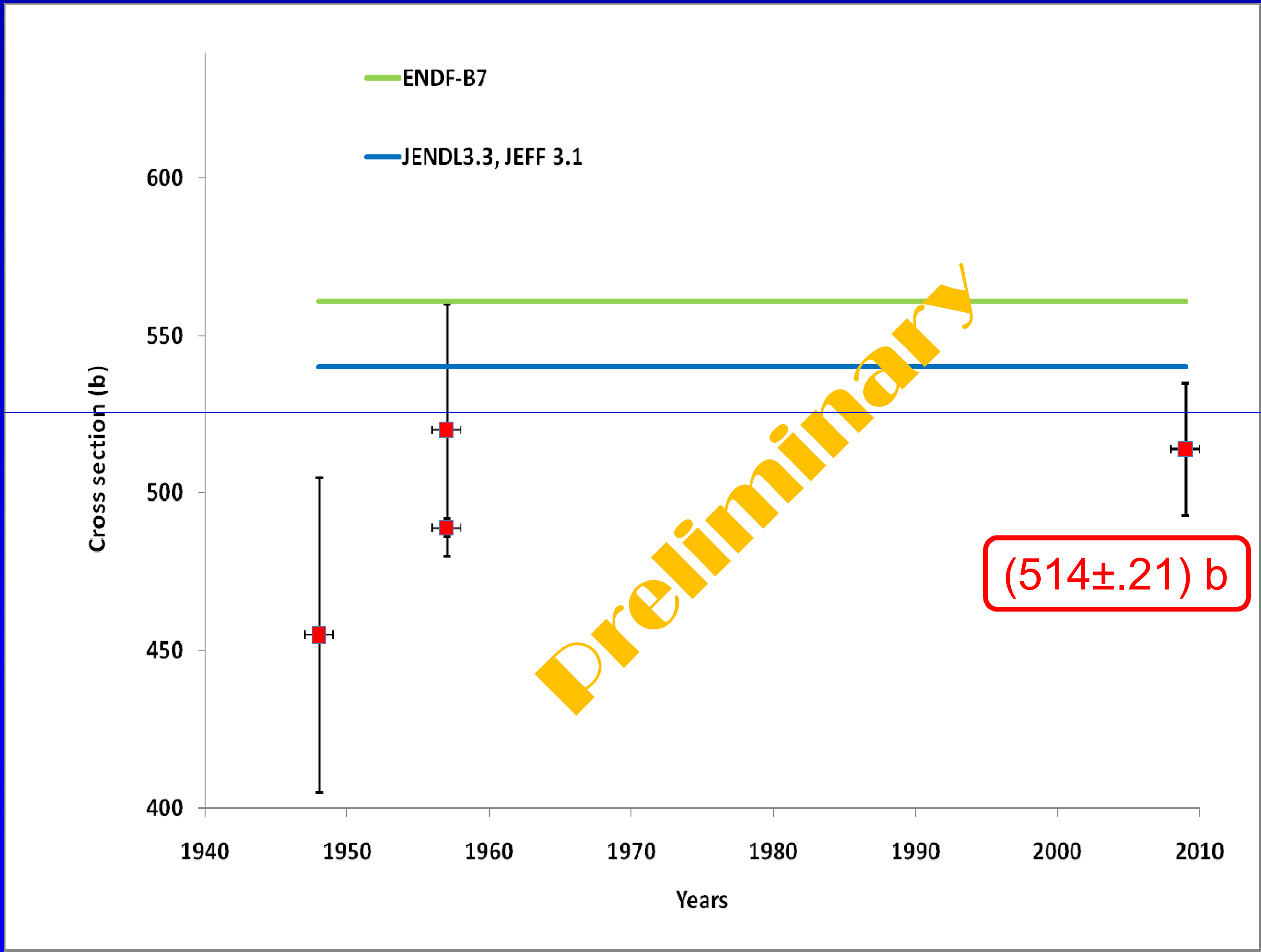
Actinide evolution



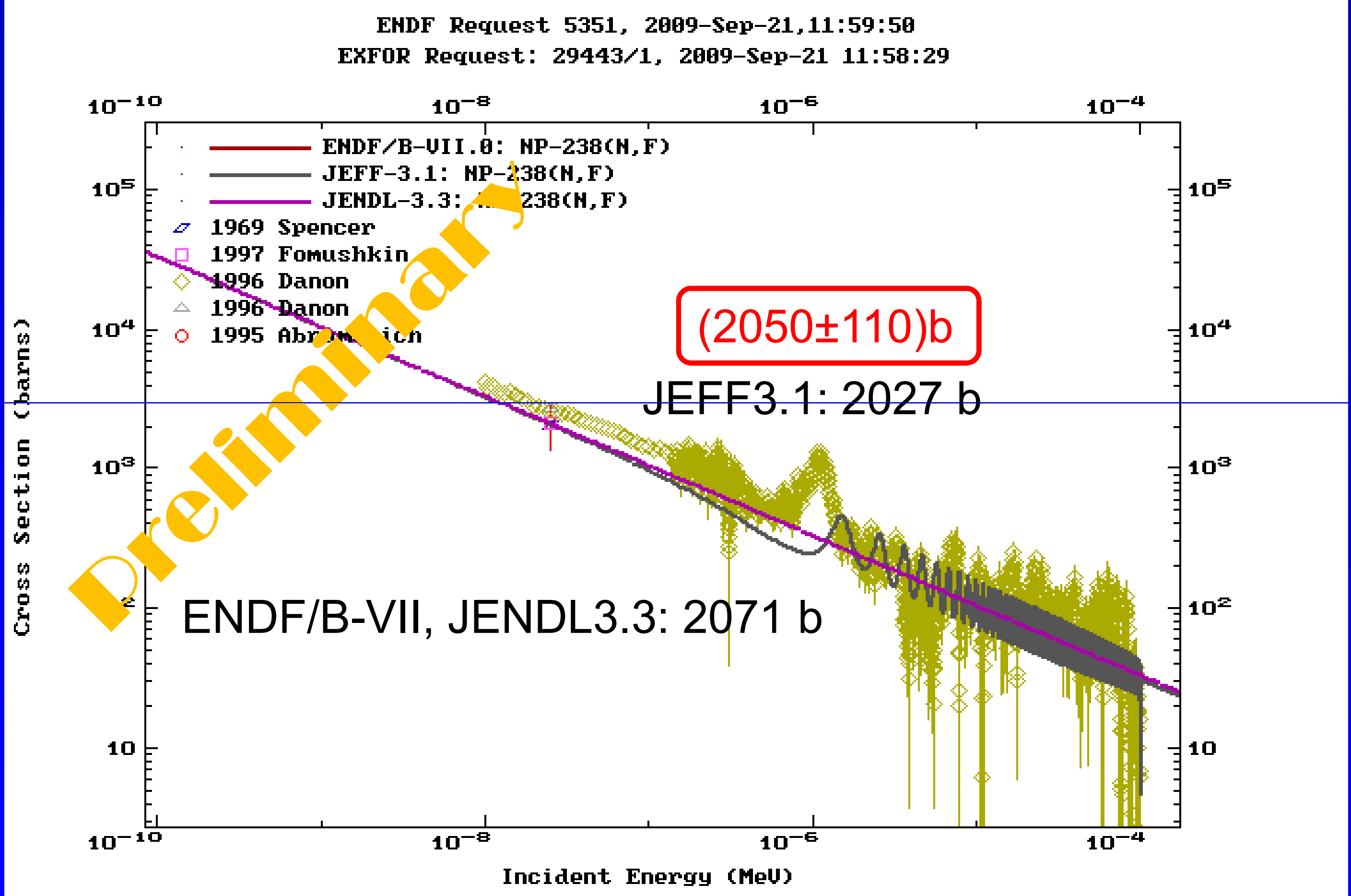


- Data were fitted with the MERCS code

25.3 meV $^{238}Pu(n,\gamma)$ value

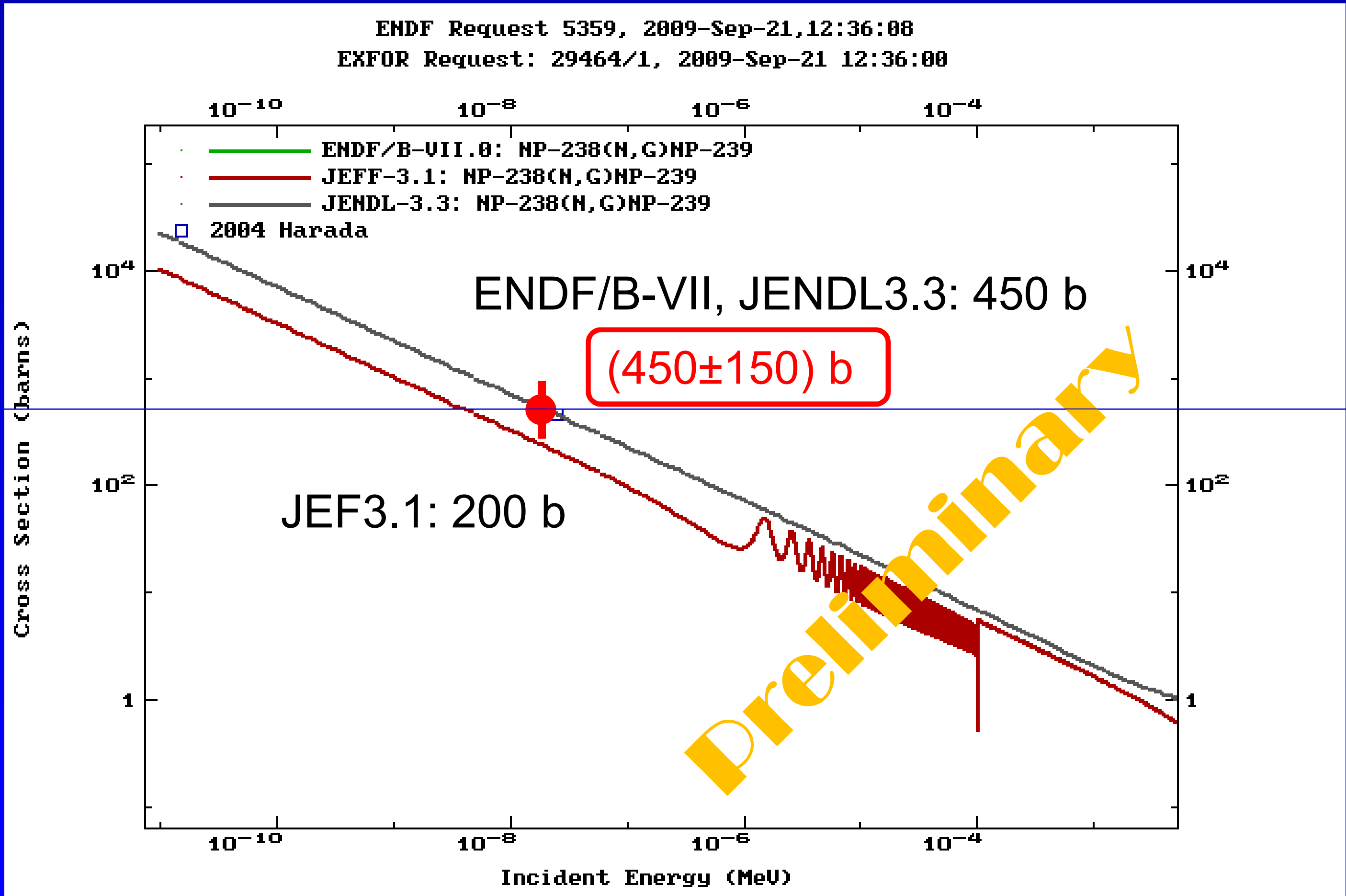


25.3 meV $^{238}Np(n,f)$ value



A. Letourneau et al., to be submitted to Nuclear Physics A

25.3 meV $^{238}Np(n,\gamma)$ value



Summary

- Efficient set-up and methods to measure slow neutron-induced:
  - Capture cross sections:
    - ^{232}Th, ^{233}Pa, ^{234}U, ^{237}Np, ^{238}Pu, ^{242}Pu, ^{241}Am, $^{242gs\text{-}m}Am$, ^{243}Am, ^{242}Cm, ^{244}Cm
  - Fission cross sections:
    - ^{238}Np, $^{242gs\text{-}m}Am$ and ^{245}Cm
  - β-decay half-life and/or γ-ray intensities:
    - ^{238}Np, $^{244m\text{-}gs}Am$
- In progress:
  - Capture:
    - ^{248}Cm, ^{249}Cf, ^{250}Cf, ^{251}Cf
- All the data are not yet published but in EXFOR

Involved people

E. Dupont, A. Letourneau, S. Panebianco, Ch. Veyssière
CEA/DSM/IRFU
L. Oriol, F. Chartier
CEA/DEN
P. Mutti
Institut Laue Langevin
I. AlMahamid
WADSWORTH Center, Laboratory of Inorganic and nuclear chemistry