The European Commission’s science and knowledge service

Joint Research Centre

European Commission

Characterisation of Nuclear Materials by using Neutron Resonance Analysis

C. Paradela

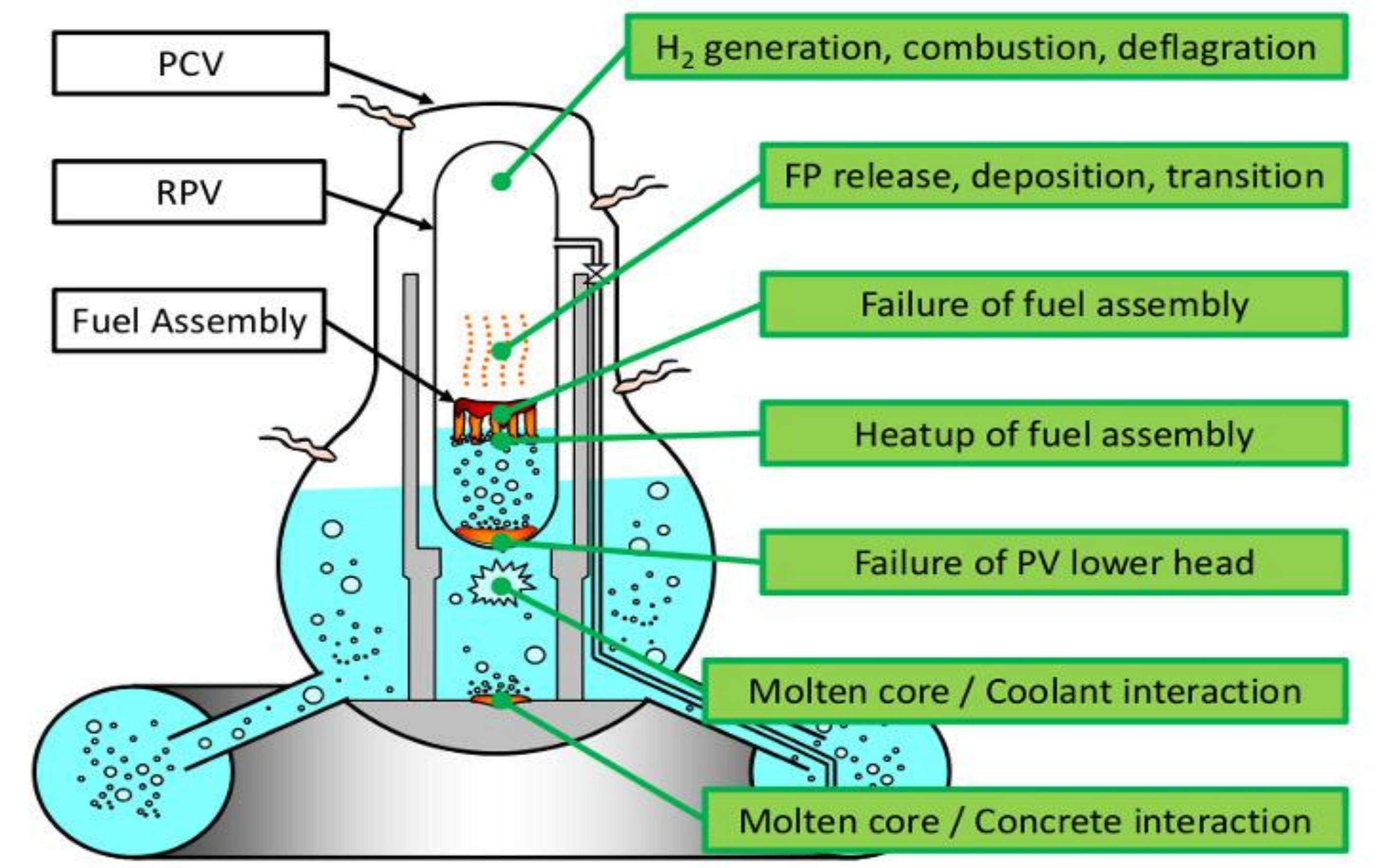
EuNPC 2018, Bologna, September 2nd-7th 2018

Content

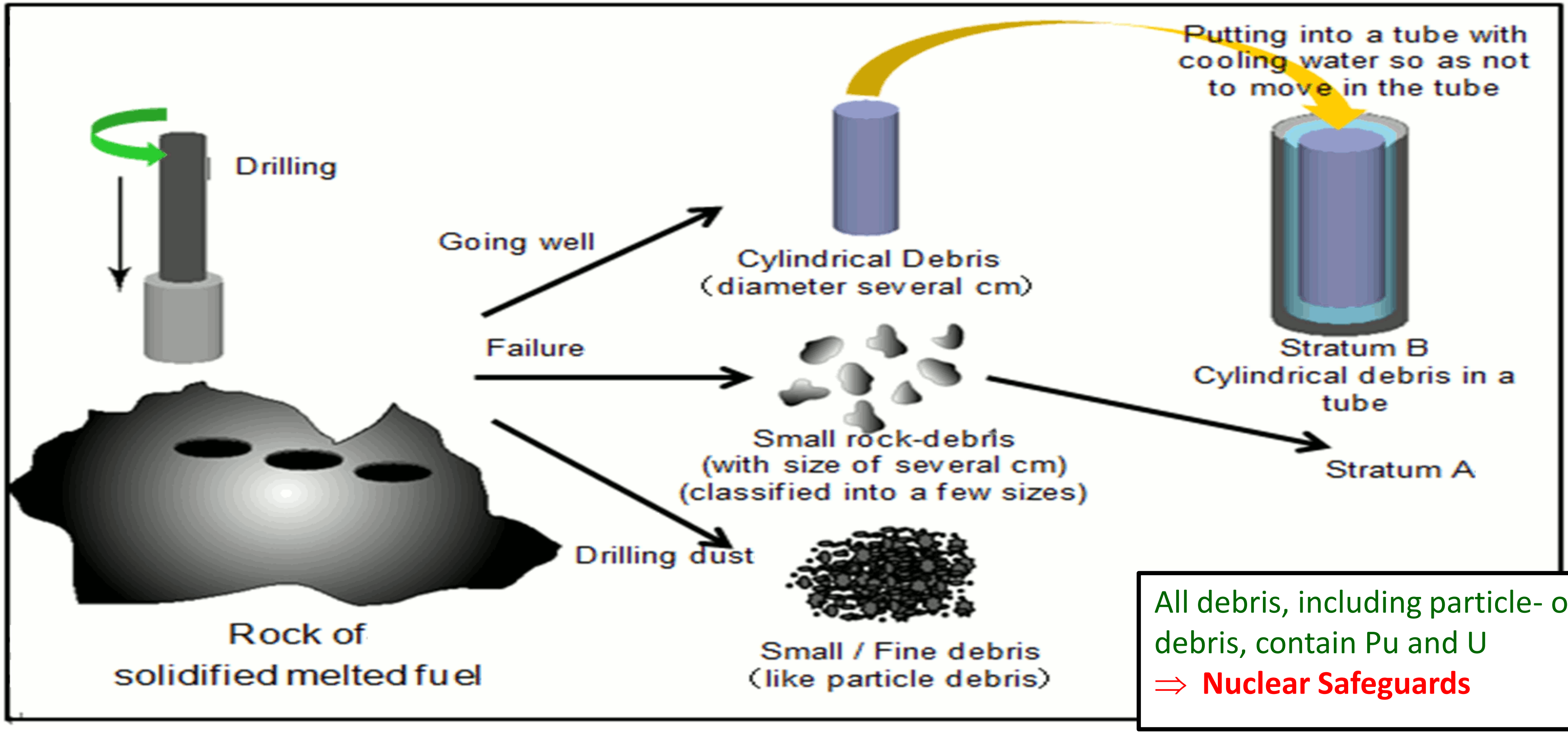
- Context
  - Special Nuclear Materials (SNM) characterisation
- Method
  - Measurement principle
  - Testing
- Characterisation of reference samples (CBRN standards)
  - Pu samples
  - U samples

Fukushima NPP decommissioning

- Following a severe earthquake in 2011, a tsunami destroyed the power supply to the Fukushima Daiichi nuclear power plant
- As a consequence, the nuclear fuel melted and mixed with coolant and structural materials
- During decommission, molten fuel must be removed from the reactor taking into account nuclear safeguards



Quantification of fissile material in fuel debris



All debris, including particle- or rock- like debris, contain Pu and U

⇒ **Nuclear Safeguards**

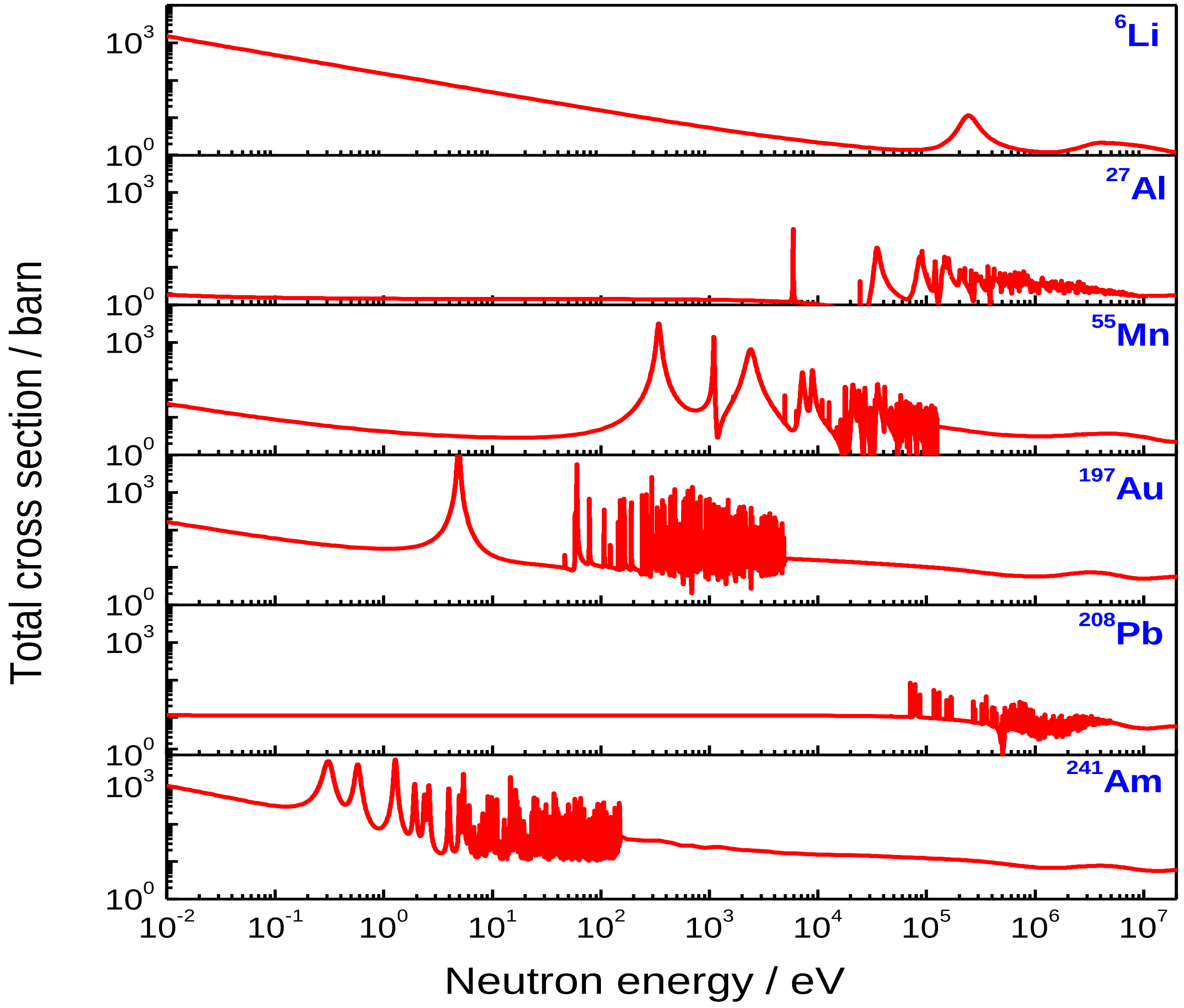
Nuclear physics for archeological studies

- Neutron resonance techniques were used for studying archeological artifacts at GELINA

- Neutron Resonance Capture Analysis (NRCA): based on the detection of γ-rays

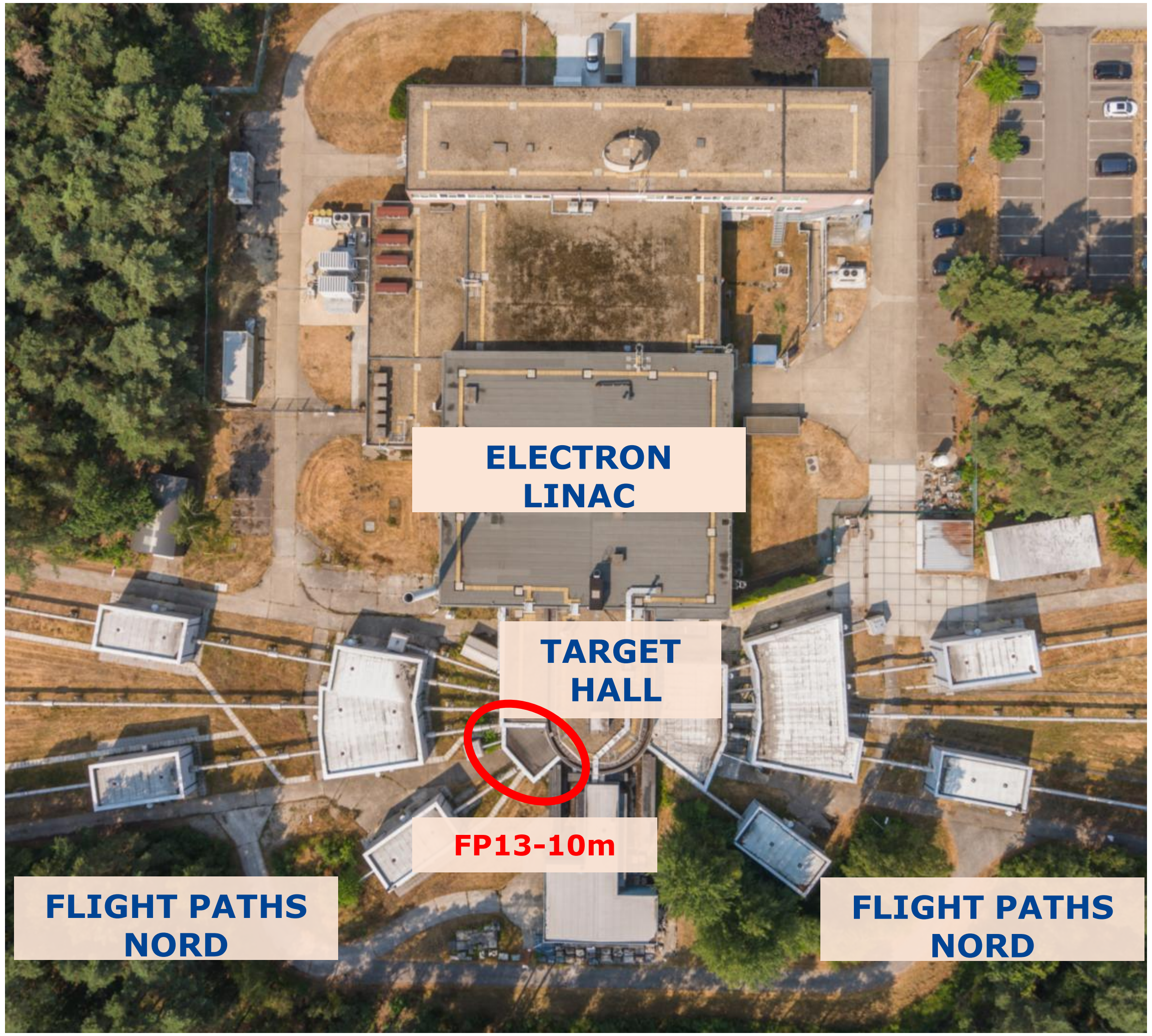
- Neutron Resonance Transmission Analysis (**NRTA**): based on transmission measurements

Neutron Cross Sections



- Probability of a neutron to undergo a reaction with a target nucleus
- Resonances appear at energies, which are specific for each nuclide
- Resonances can be used as fingerprints to determine the elemental composition

GELINA - overview



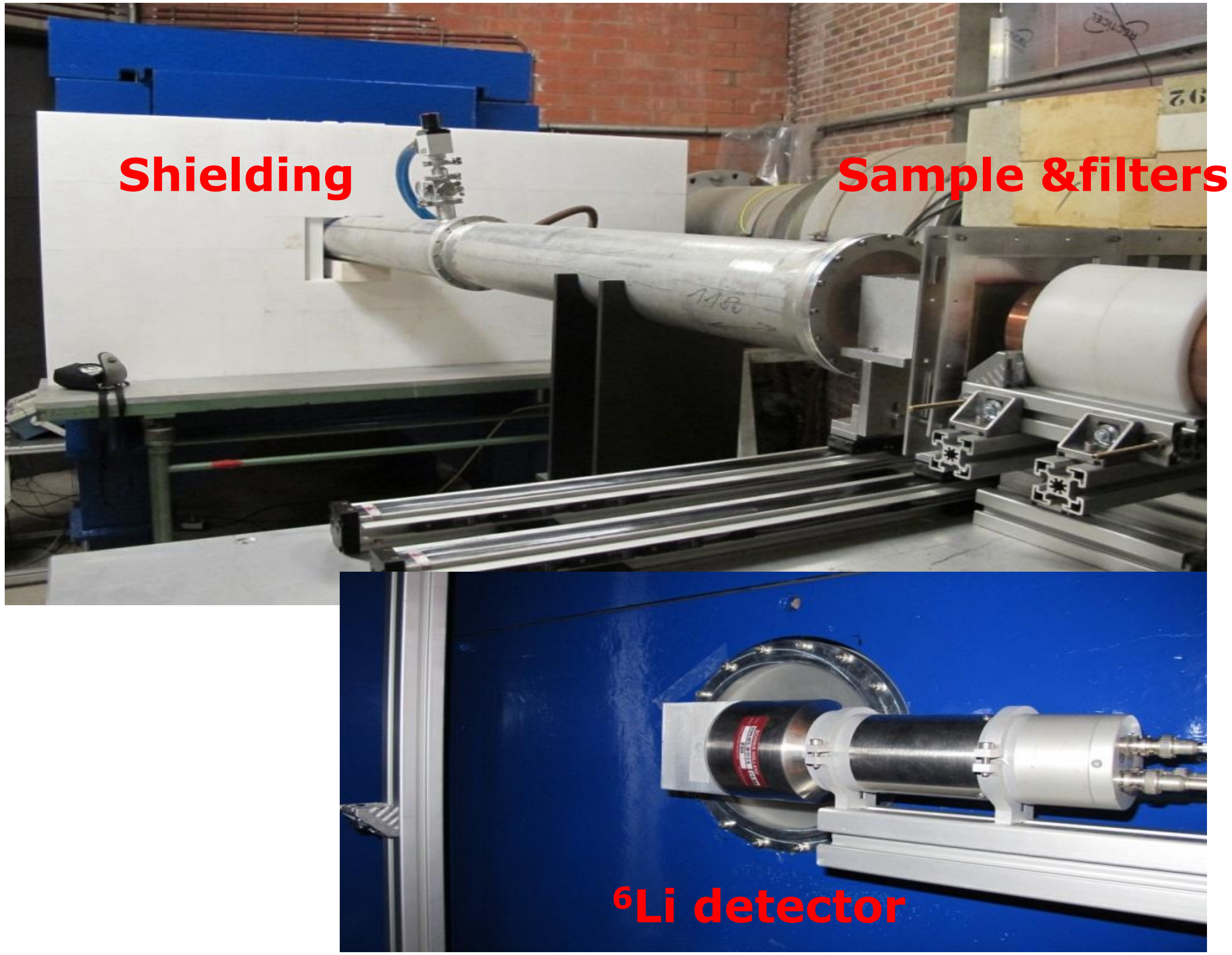
- Pulsed white neutron source

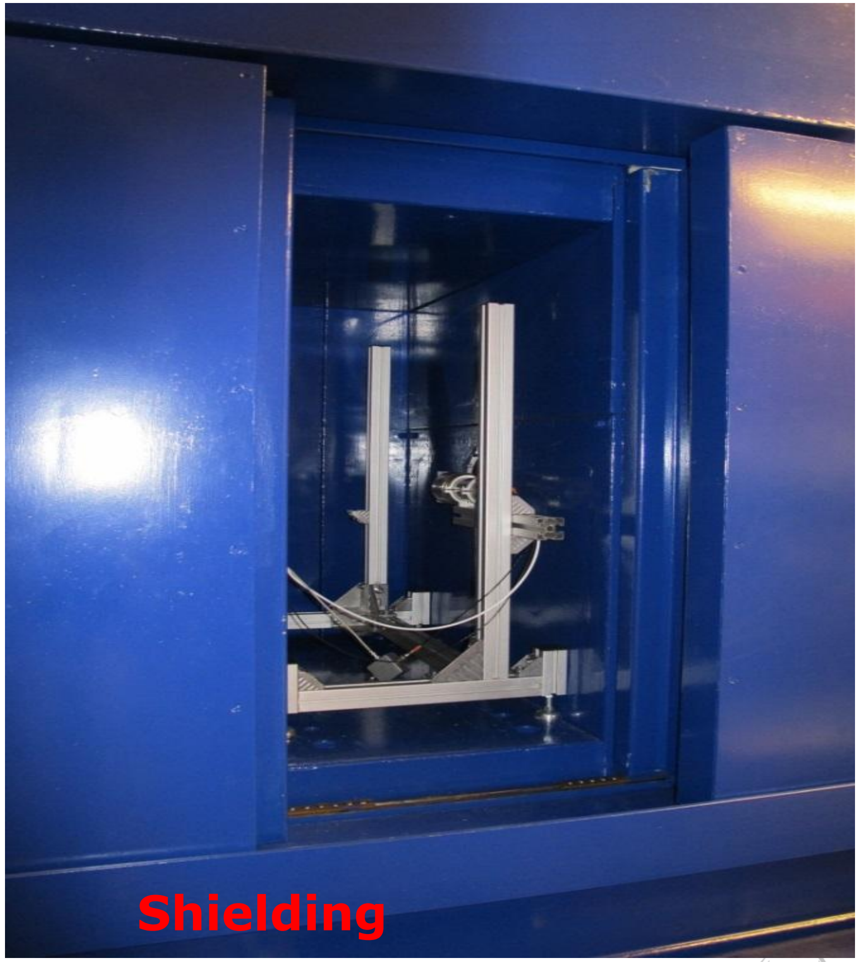
  (10 meV < E_n < 20 MeV)

- Pulse frequency 50Hz – 800 Hz
- Neutron energy : time – of – flight (TOF)
- Multi-user facility: 12 flight paths
  (10 m - 400 m)
- Measurement stations with special equipment to perform:
  - Total cross section measurements
  - Partial cross section measurements

European Commission

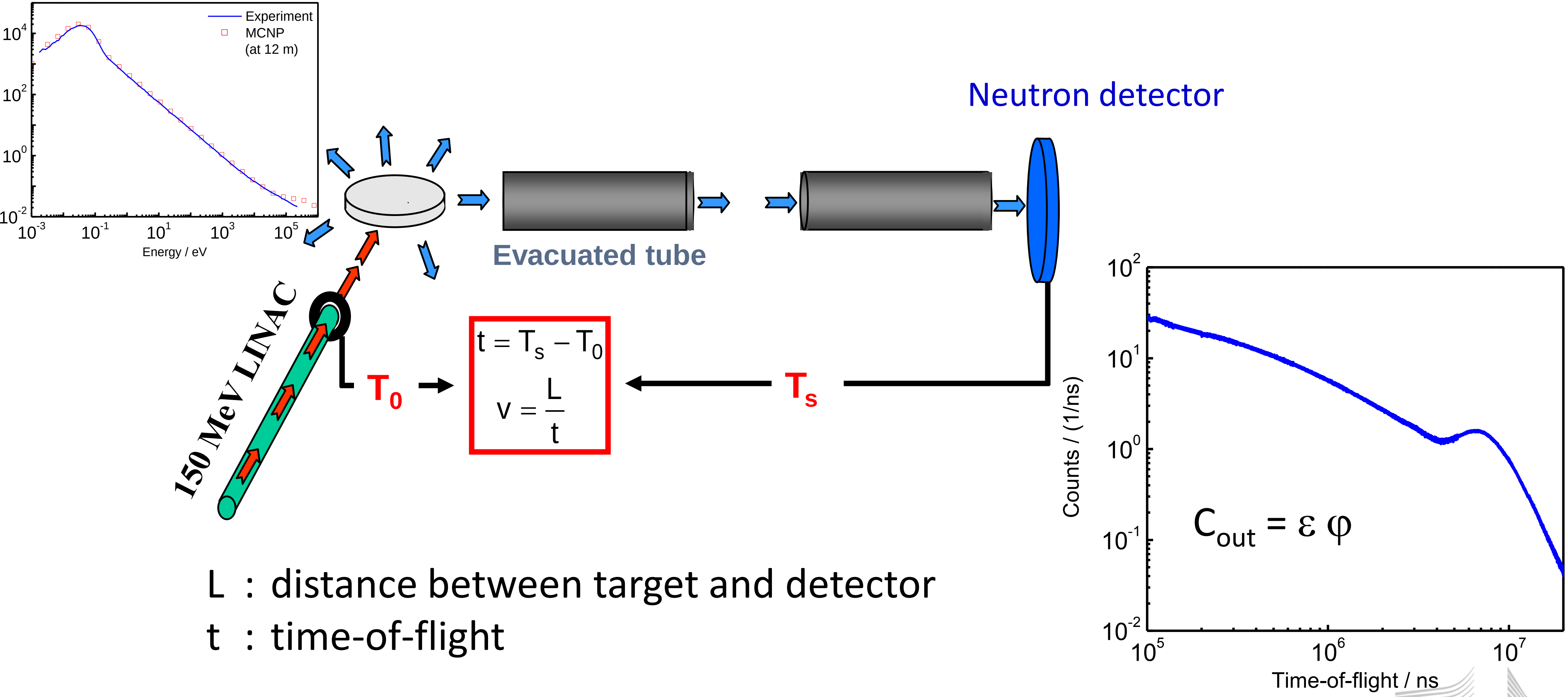
GELINA - NRTA station (FP13)





European Commission

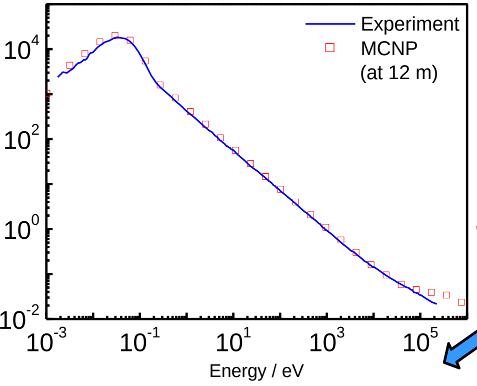
ToF - principle

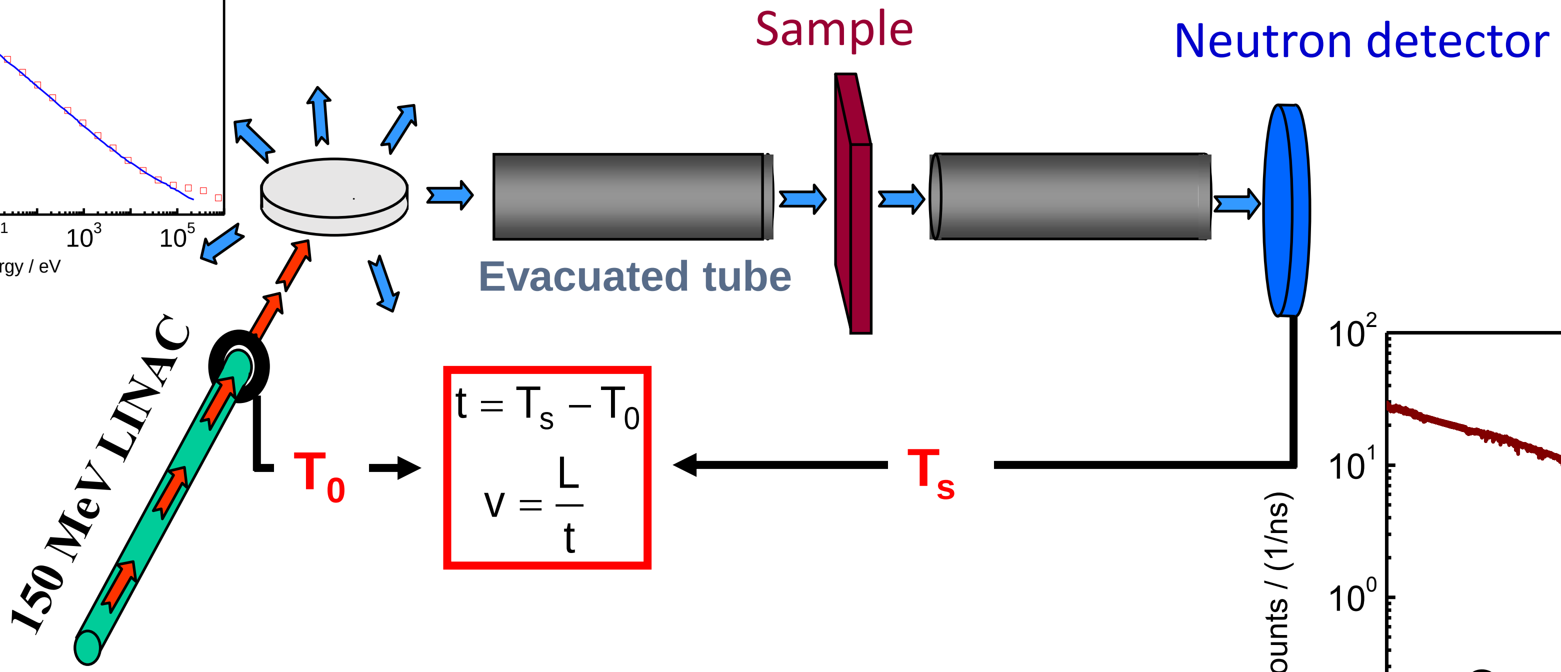


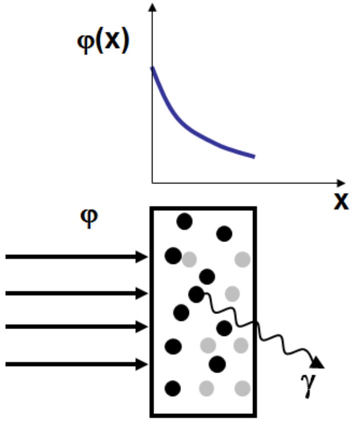
L : distance between target and detector

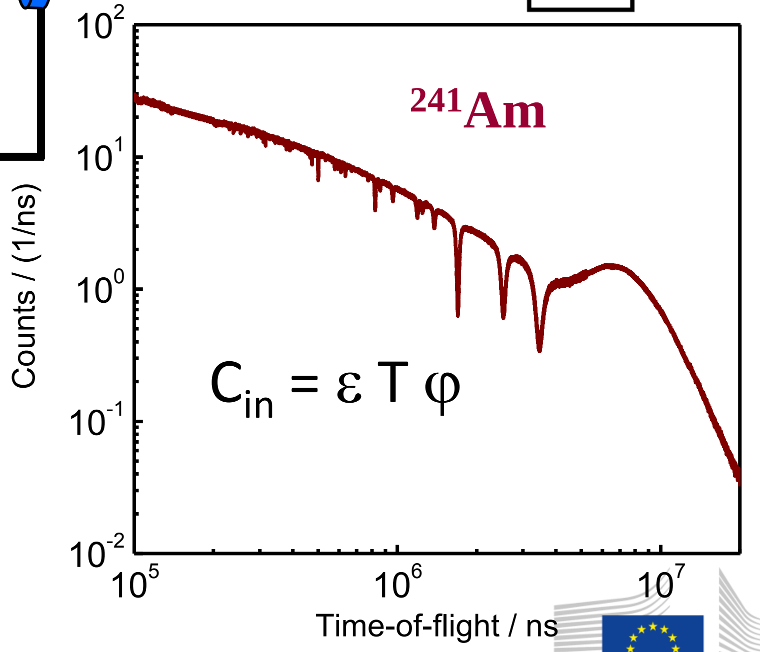
t : time-of-flight

ToF - principle







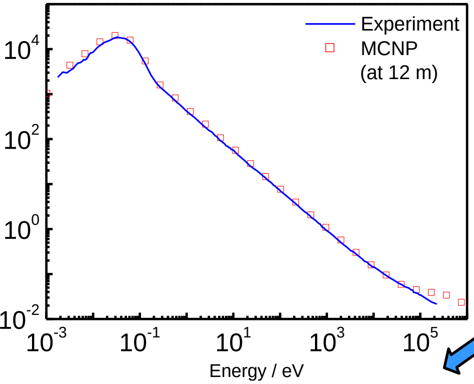


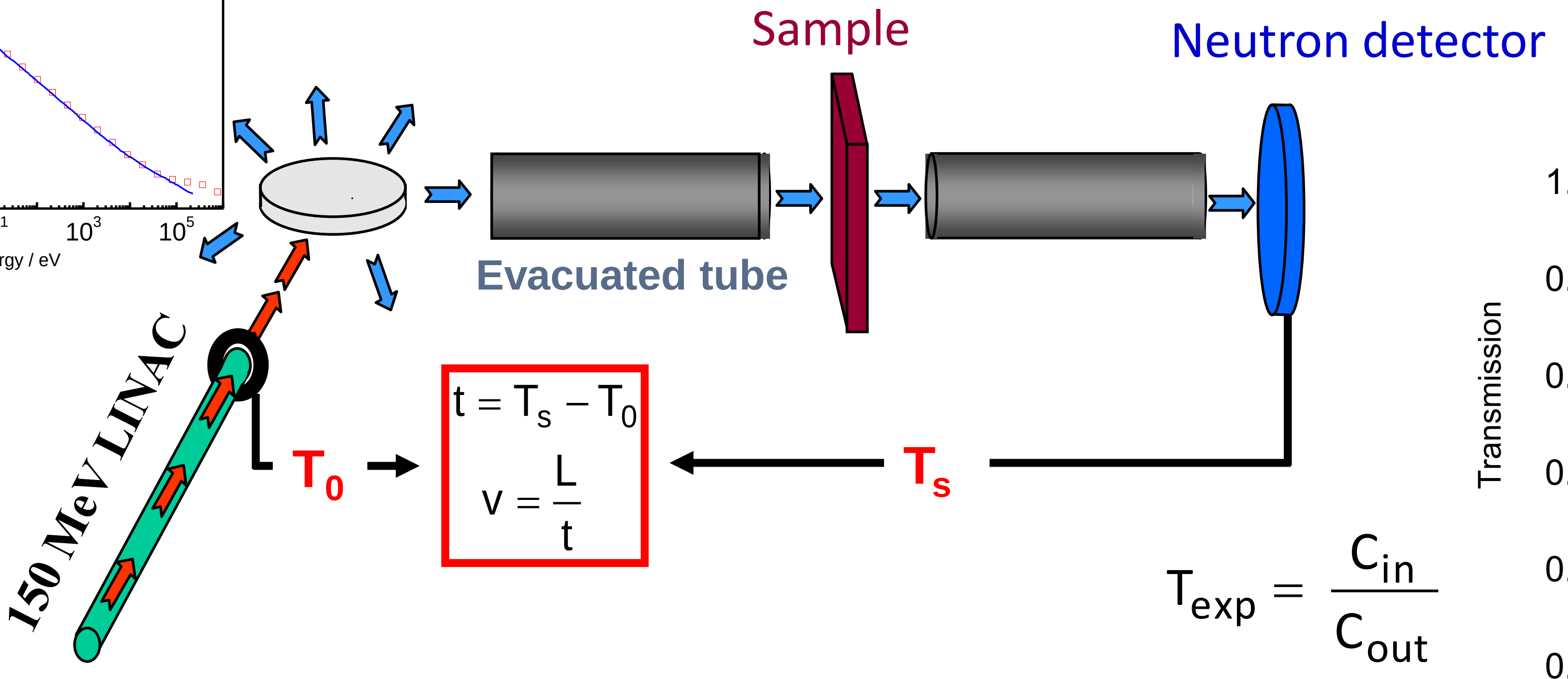
L : distance between target and detector

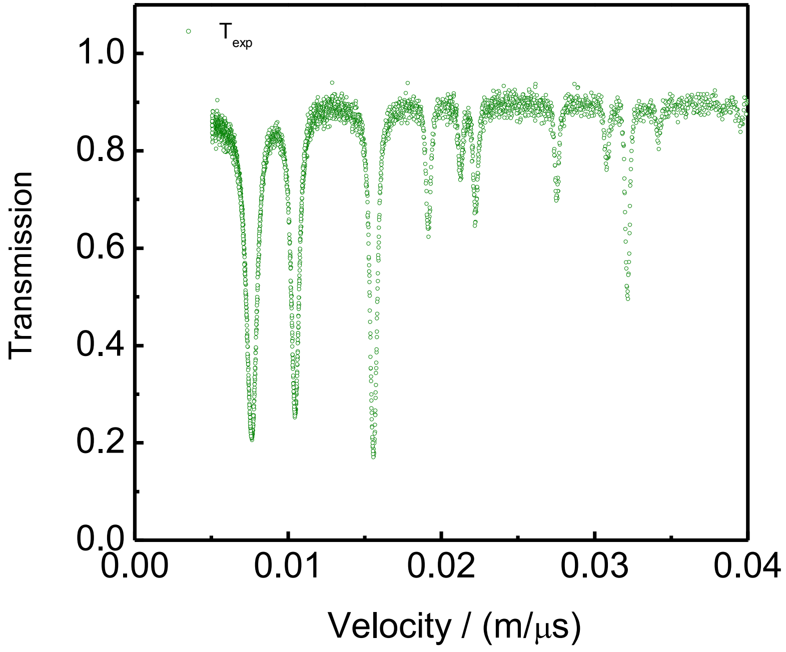
t : time-of-flight

European Commission

ToF - principle







$$\Rightarrow \quad E = mc^2(\gamma - 1) \cong \frac{1}{2}mv^2$$

$$T \cong e^{-n\sigma_{tot}}$$

NRTA

$$T_{exp} = \frac{C_{in}}{C_{out}}$$

$$T = e^{-n\,\bar{\sigma}_{tot}}$$

Resonance Shape Analysis: REFIT

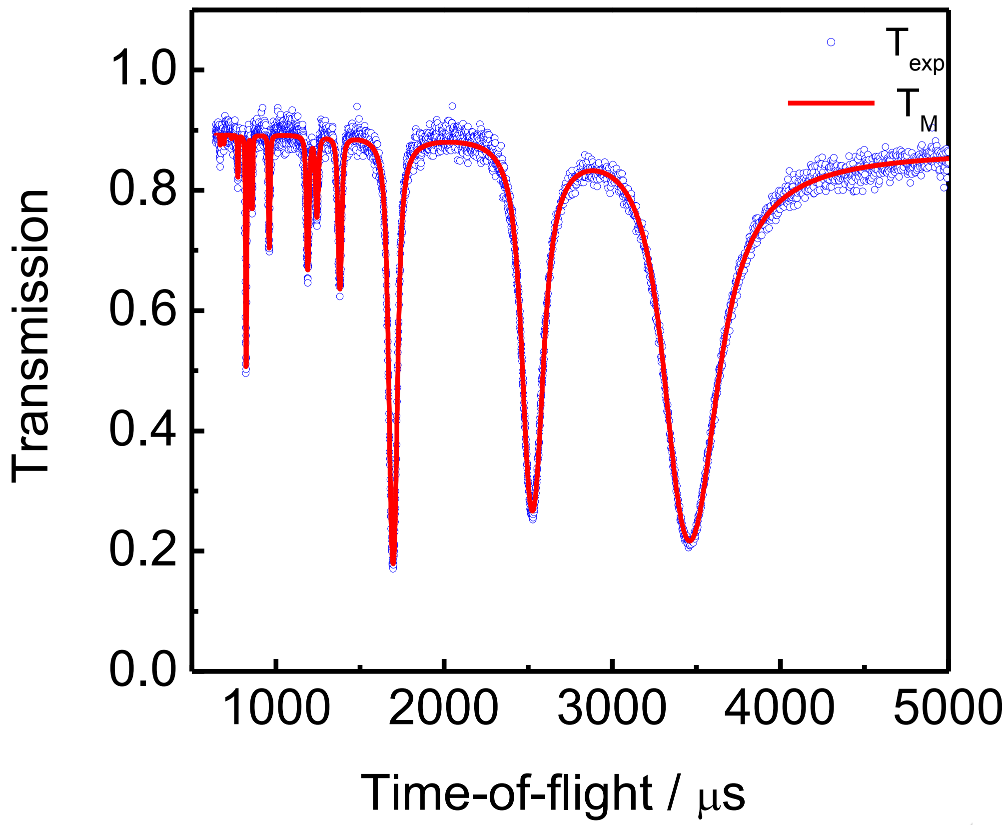
$$\chi^2(n) = \left(T_{exp} - T_M(t,n)\right)^T V_{T_{exp}}^{-1} \left(T_{exp} - T_M(t,n)\right)$$

$$T_M(t) = \int R(t, E)\, T(E)\, dE$$

$$T(E) = e^{-n\,\bar{\sigma}_{tot}(E)}$$

n : areal density

total number of nuclei per unit area



CBNM standard: Pu

PuO$_2$ Reference samples

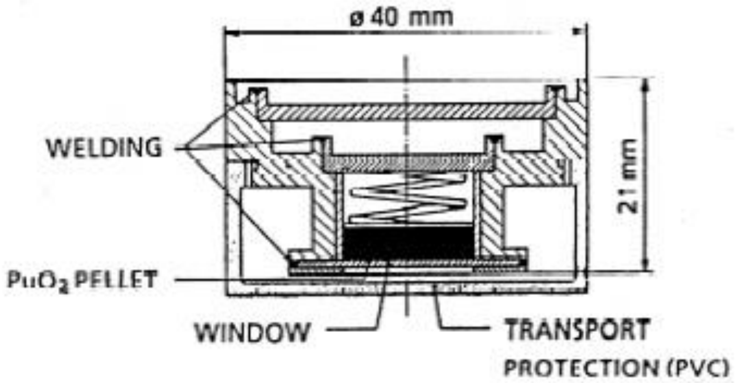
- Certified Reference Material for γ-ray spectrometry
- Different ^{239}Pu enrichment
- Presence of ^{241}Am from ^{241}Pu decay

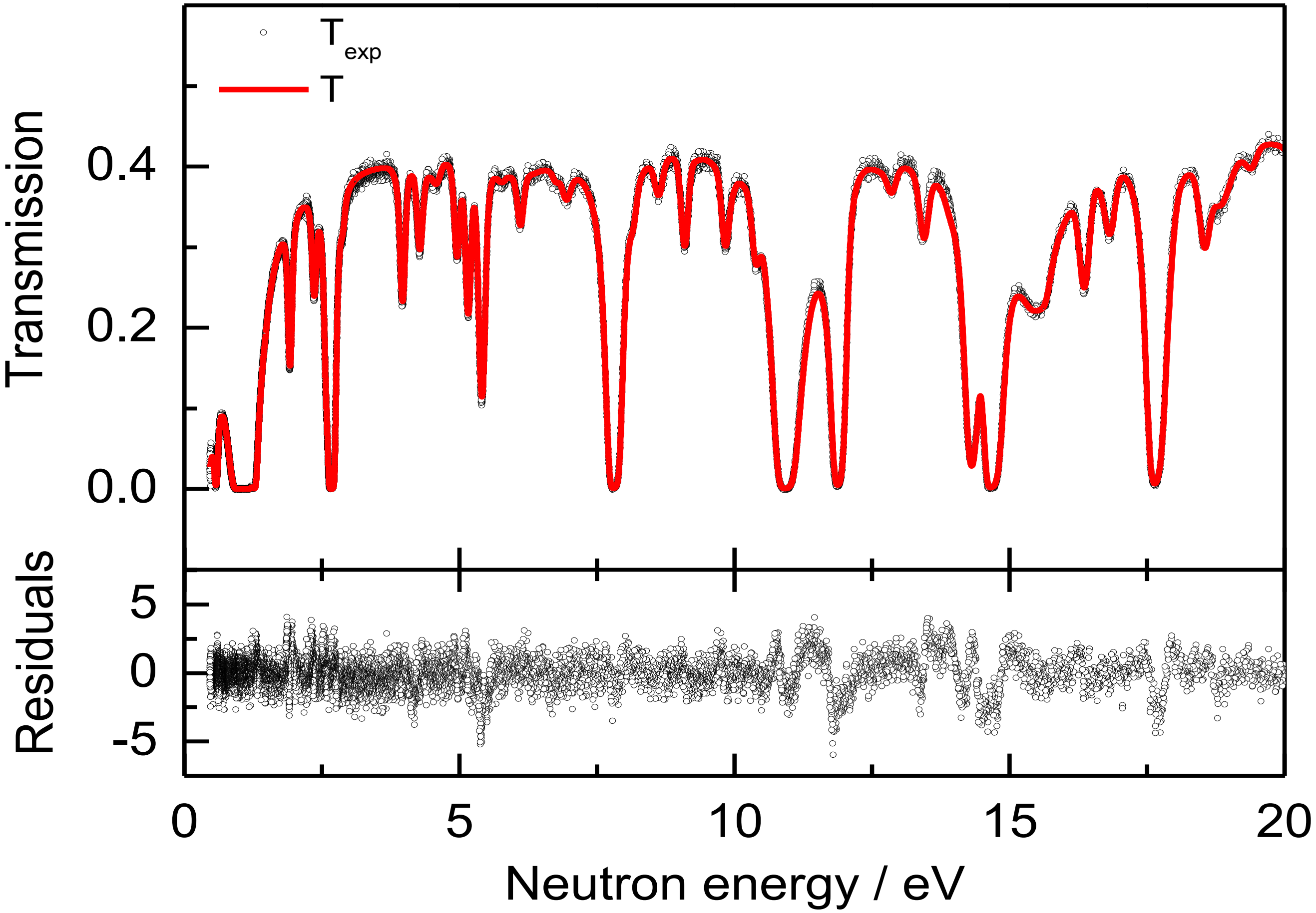
Plutonium Isotopic Abundance on June 20, 1986

Atom Percent

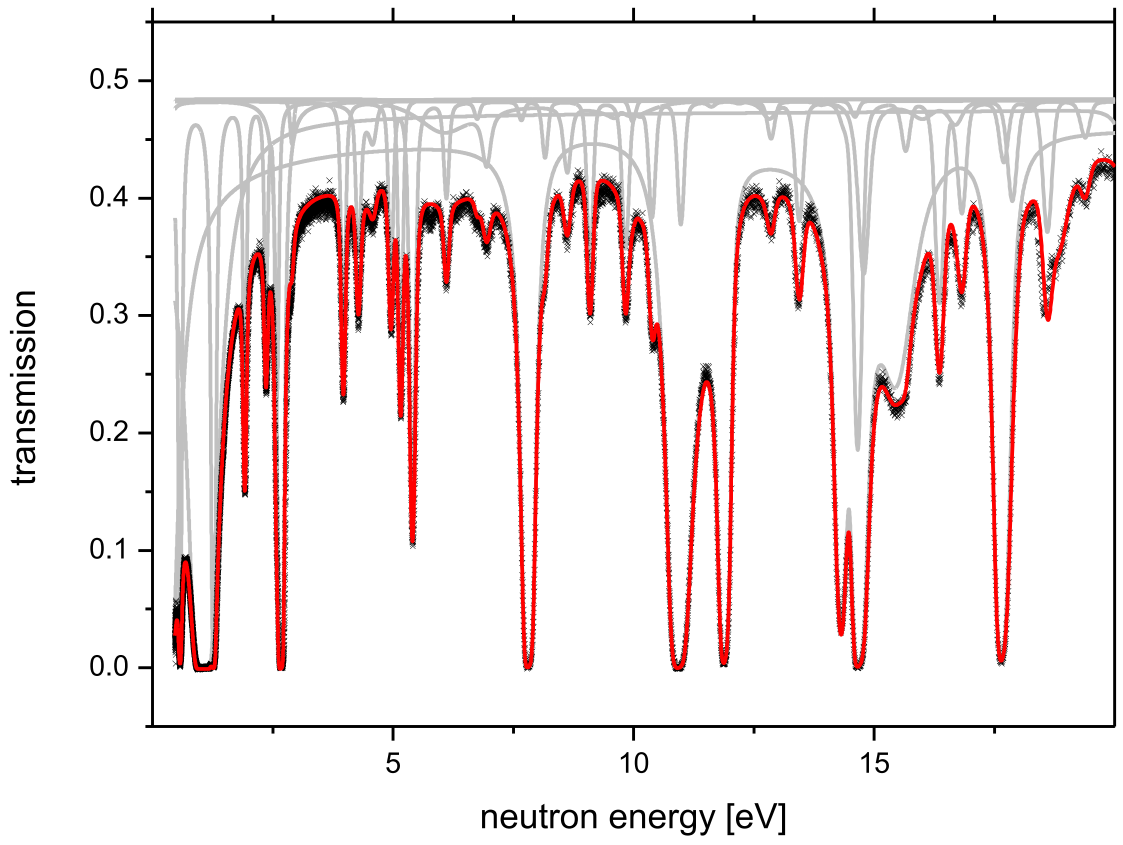
Material	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am/Pu
CBNM Pu 93	0.0117	93.4392	6.2886	0.2215	0.0390	0.1039
CBNM Pu 84	0.0706	84.3985	14.1578	1.0197	0.3534	0.2157
CBNM Pu 70	0.8506	73.4248	18.2445	5.4257	2.0544	1.1624
CBNM Pu 61	1.2045	62.6562	25.3526	6.6376	4.1491	1.4362

CBNM standard: Pu



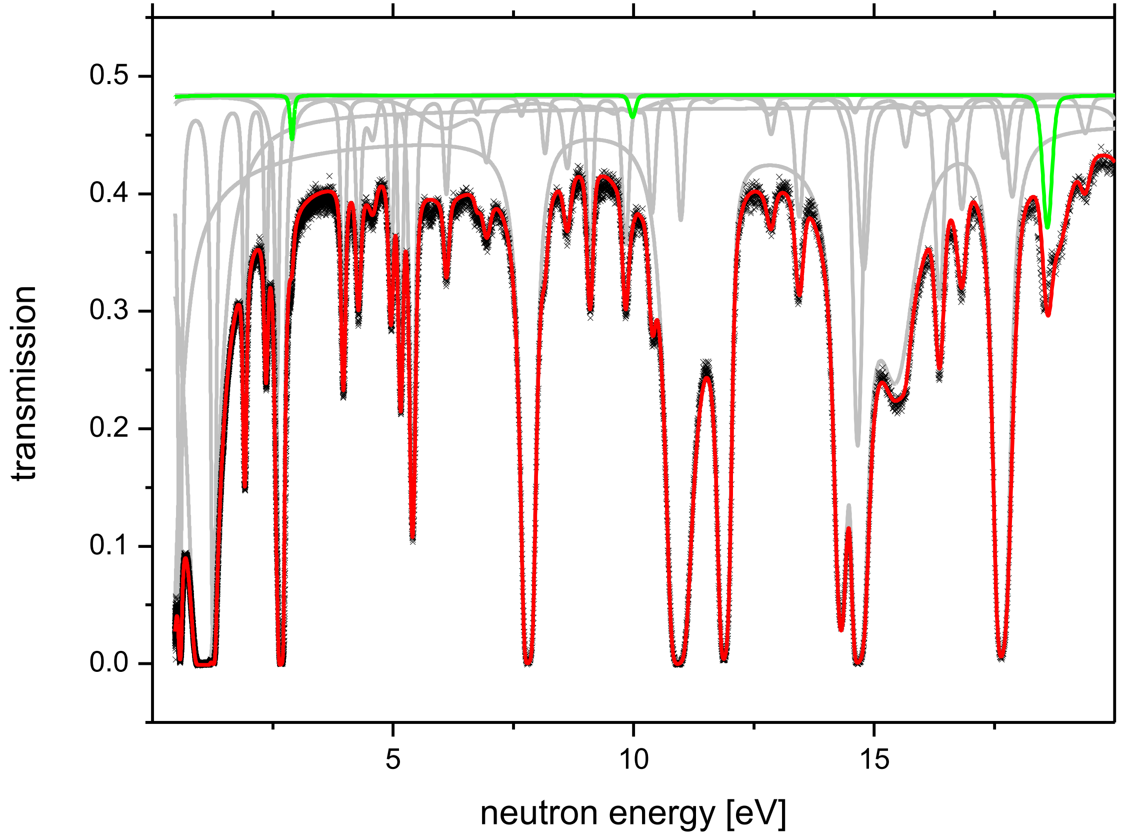


CBNM standard: Pu - Contributions



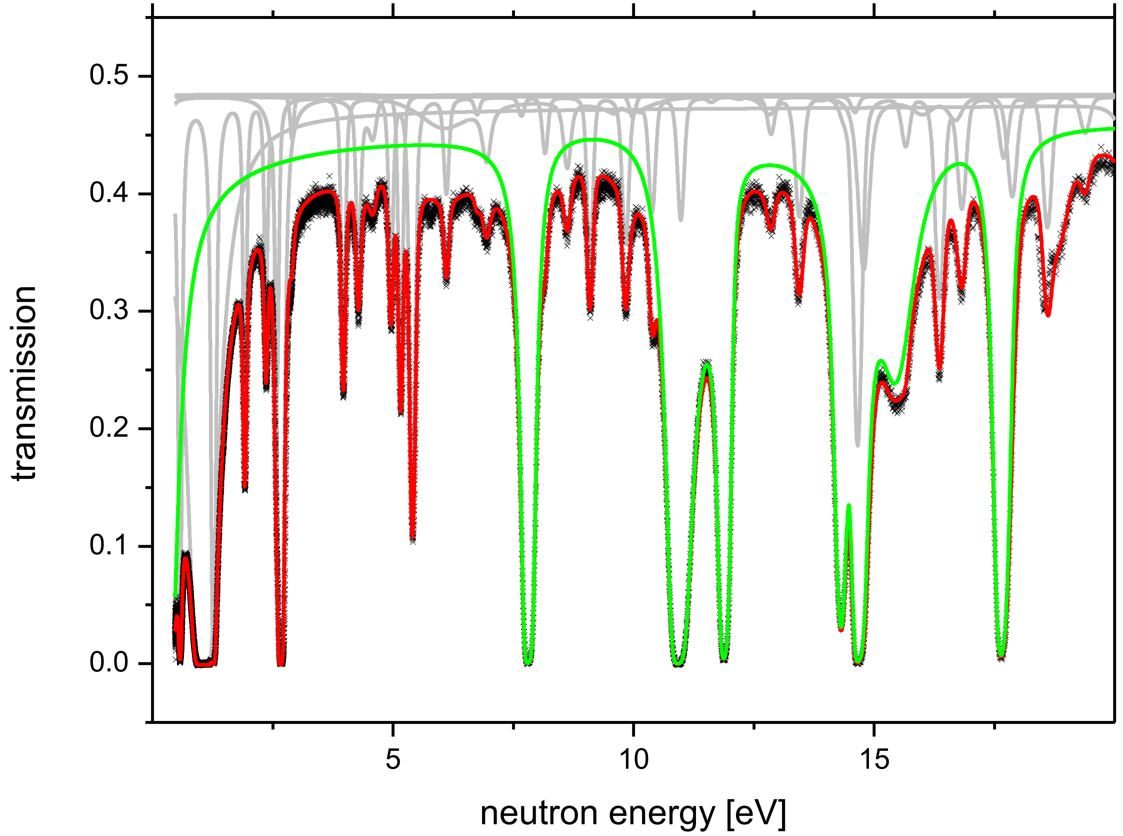
- Pu-238
- Pu-239
- Pu-240
- Pu-241
- Pu-242
- Am-241

CBNM standard: Pu - Contributions



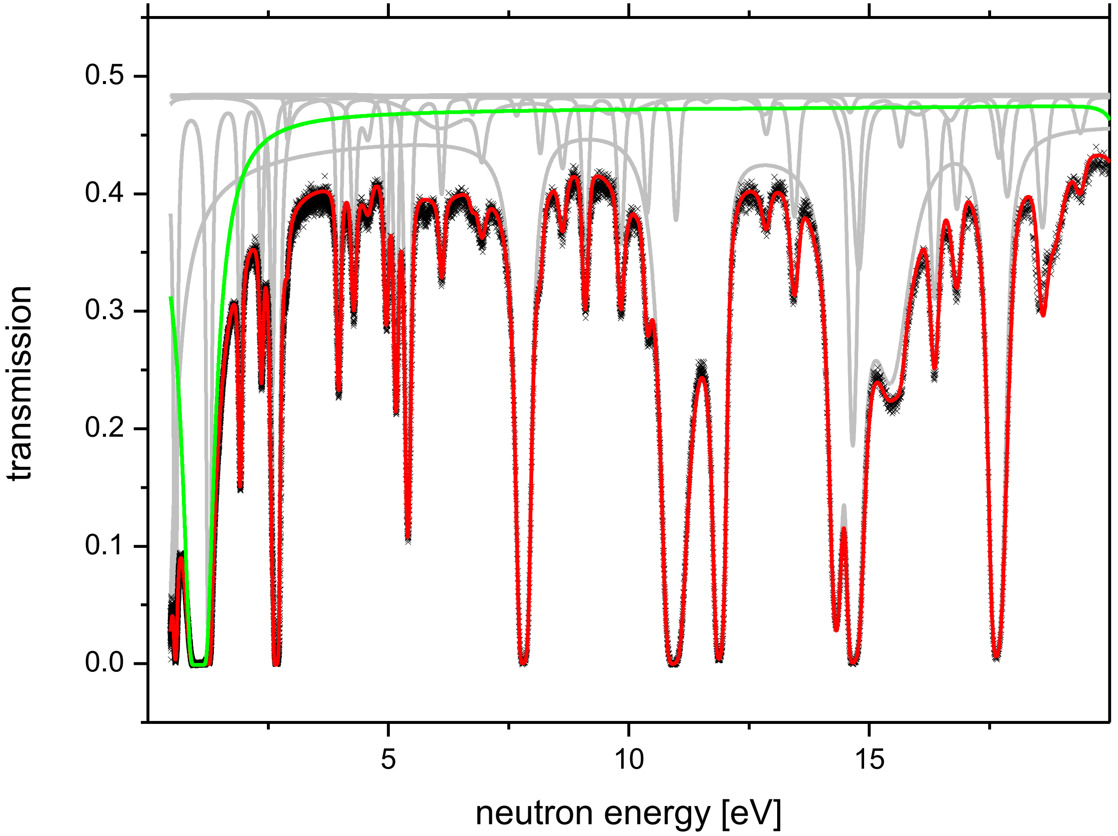
- Pu-238
- Pu-239
- Pu-240
- Pu-241
- Pu-242
- Am-241

CBNM standard: Pu - Contributions



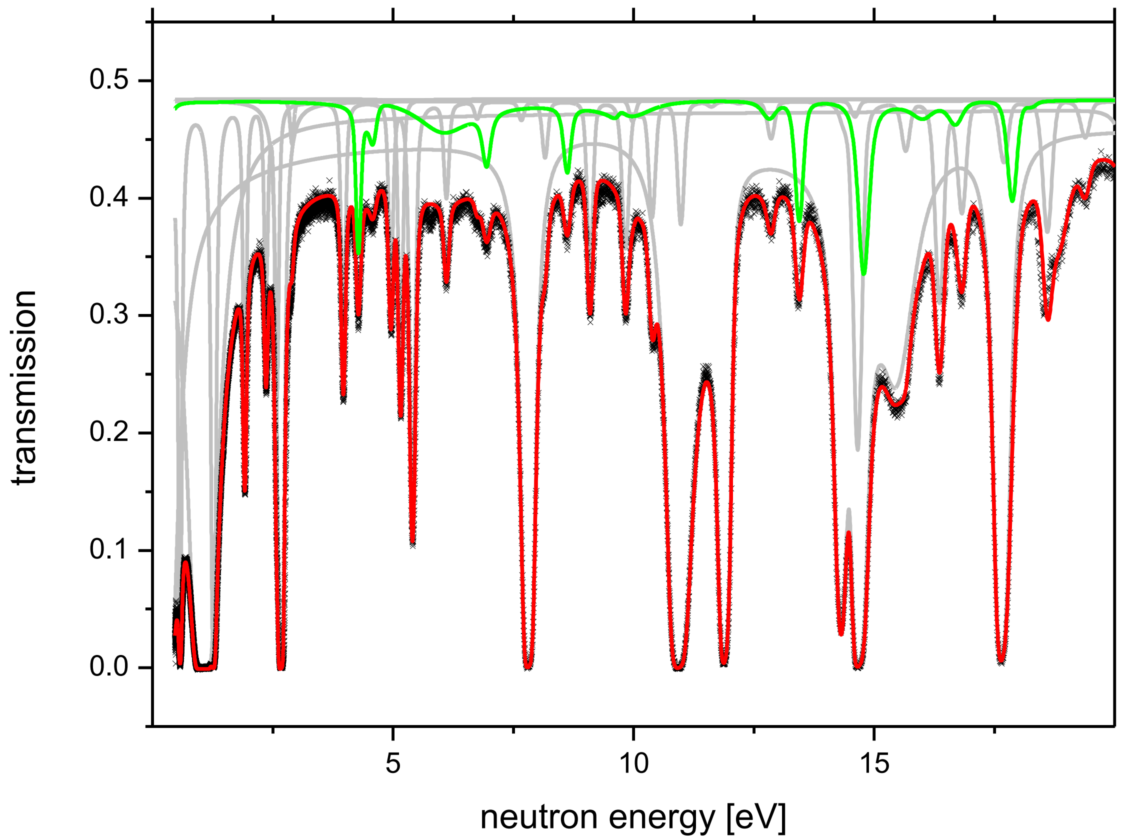
- Pu-238
- Pu-239
- Pu-240
- Pu-241
- Pu-242
- Am-241

CBNM standard: Pu - Contributions



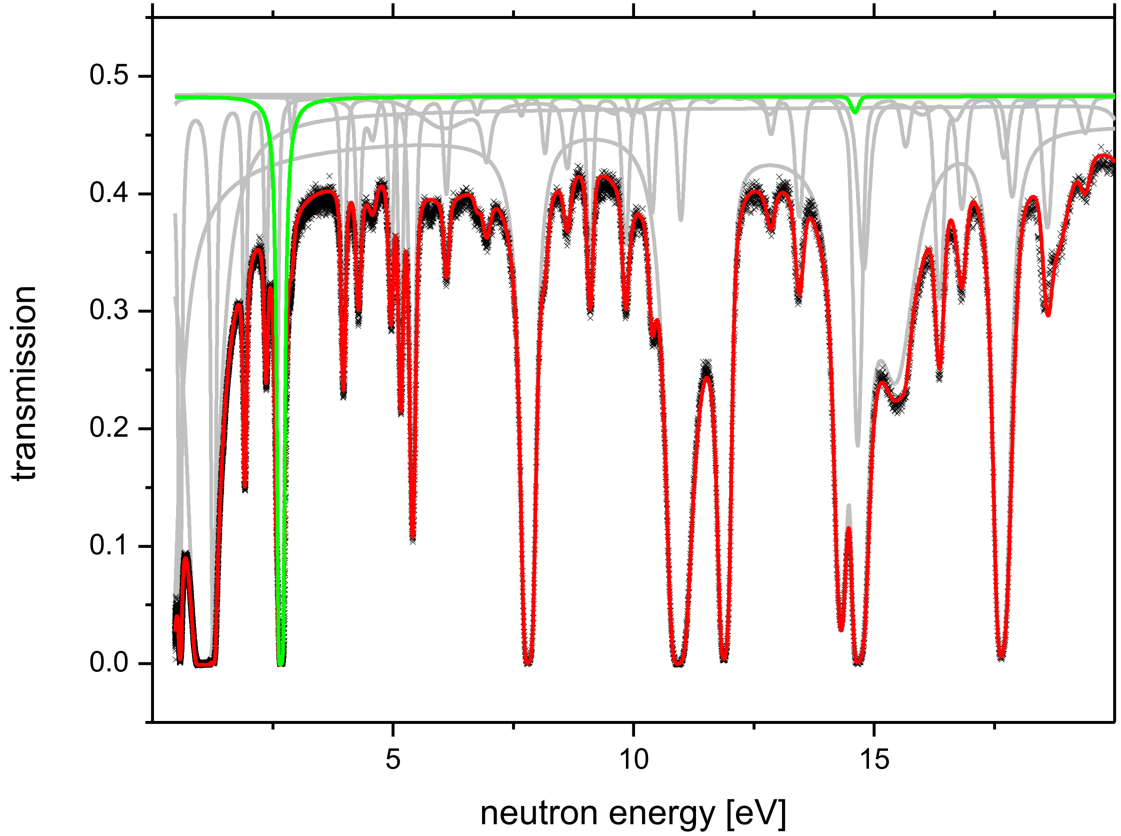
- Pu-238
- Pu-239
- Pu-240
- Pu-241
- Pu-242
- Am-241

CBNM standard: Pu - Contributions



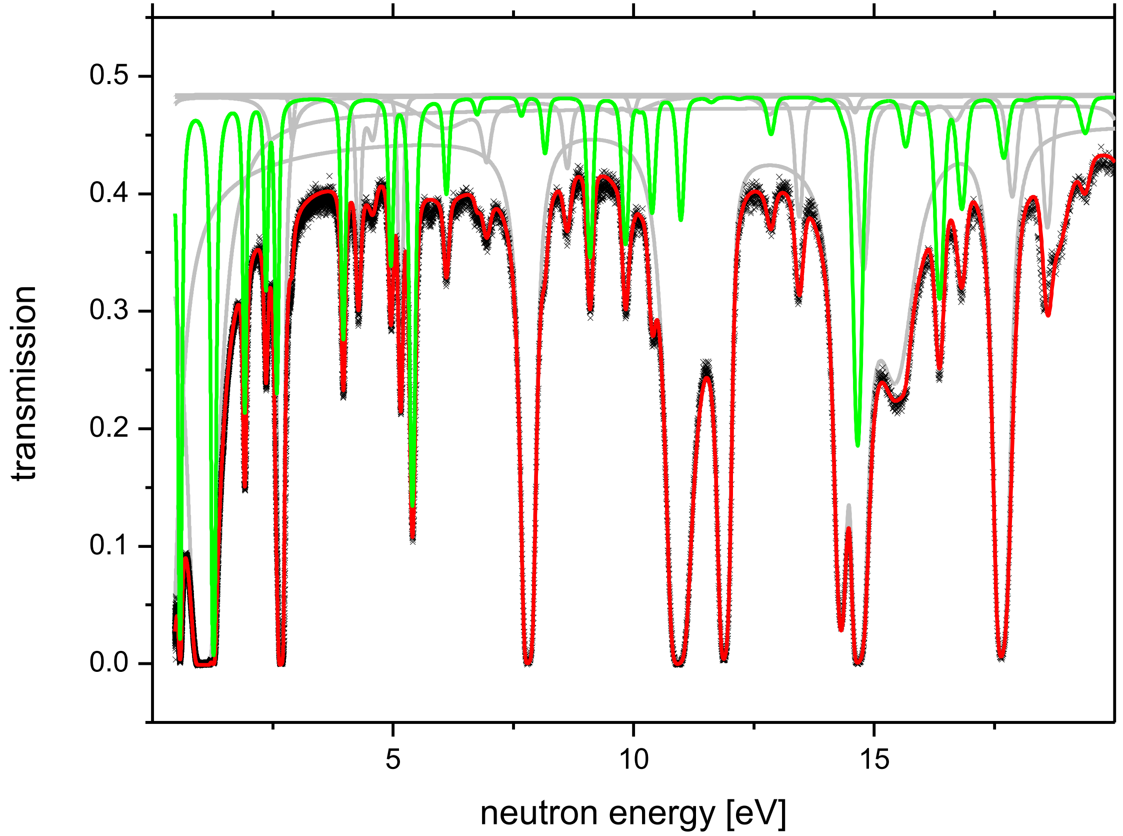
- Pu-238
- Pu-239
- Pu-240
- Pu-241
- Pu-242
- Am-241

CBNM standard: Pu - Contributions



- Pu-238
- Pu-239
- Pu-240
- Pu-241
- Pu-242
- Am-241

CBNM standard: Pu - Contributions



- Pu-238
- Pu-239
- Pu-240
- Pu-241
- Pu-242
- Am-241

Experiments at GELINA

PuO_2 Reference samples

NRM 271 Pu 61

Pu-isotope	Declared %*	NRTA	Ratio
Pu-238	0.95174	0.979±0.018	1.029
Pu-239	62.6025	62.54±0.1	0.999
Pu-240	25.3526	26.25±0.02	1.039
Pu-241	1.5641	1.574±0.008	1.007
Pu-242	4.1489	3.983±0.008	0.960
Am-241	6.2870	6.316±0.008	1.005

*Estimation considering half lives and ^{241}Am production

European Commission

Experiments at GELINA

PuO_2 Reference samples

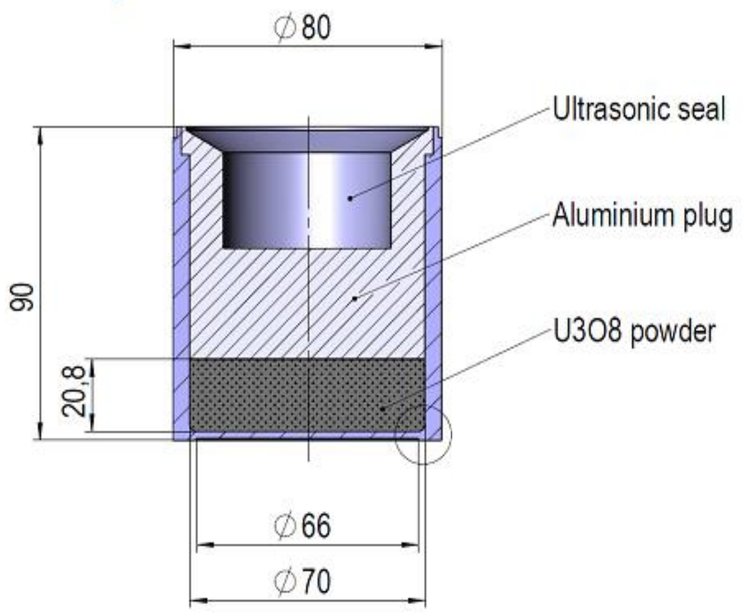
NRM 271 Pu 61

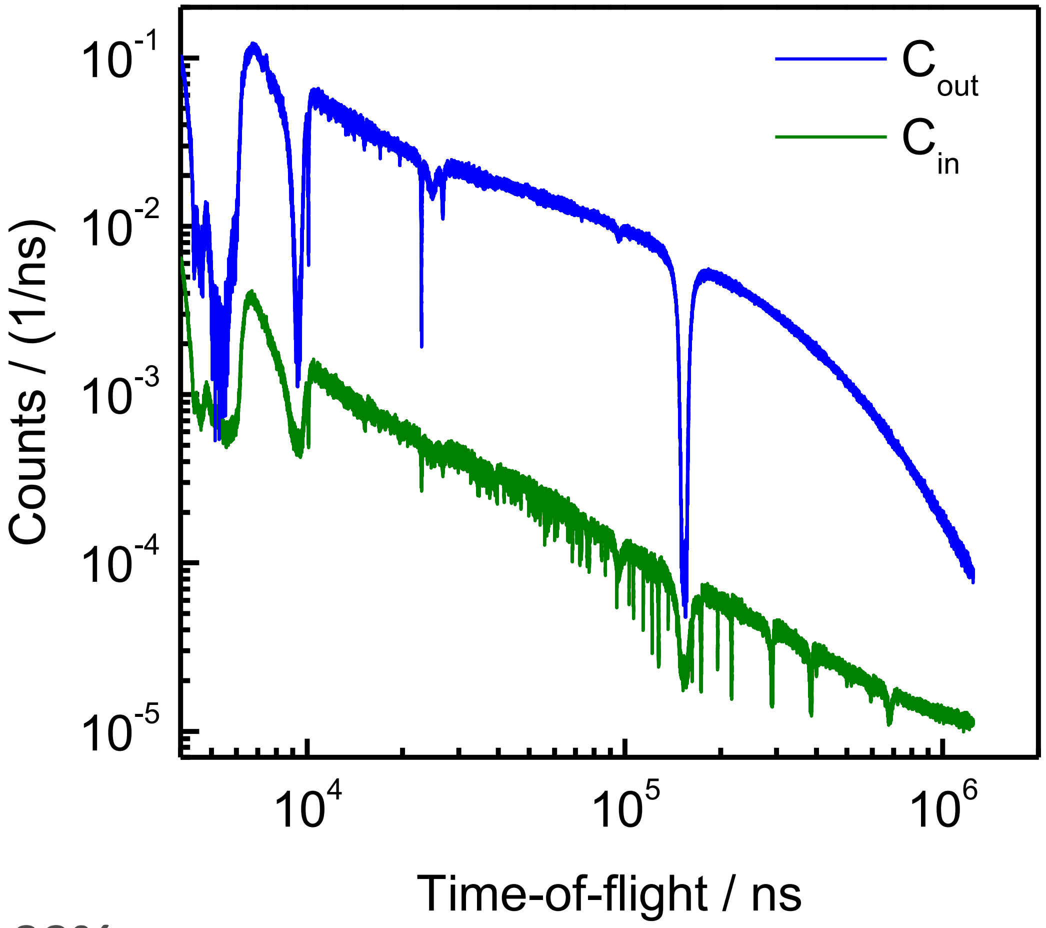
Pu-isotope	Declared %*	NRTA	Ratio
Pu-238	0.95174	0.979±0.018	1.029
Pu-239	62.6025	62.54±0.1	0.999
Pu-240	25.3526	26.25±0.02	1.039
Pu-241	1.5641	1.574±0.008	1.007
Pu-242	4.1489	3.983±0.008	0.960
Am-241 (ENDF/B-VIII.0)	**6.2870**	**7.525±0.008**	**1.197**

*Estimation considering half lives and ^{241}Am production

CBNM standard: U

U_3O_8 reference sample
EC NRM 171





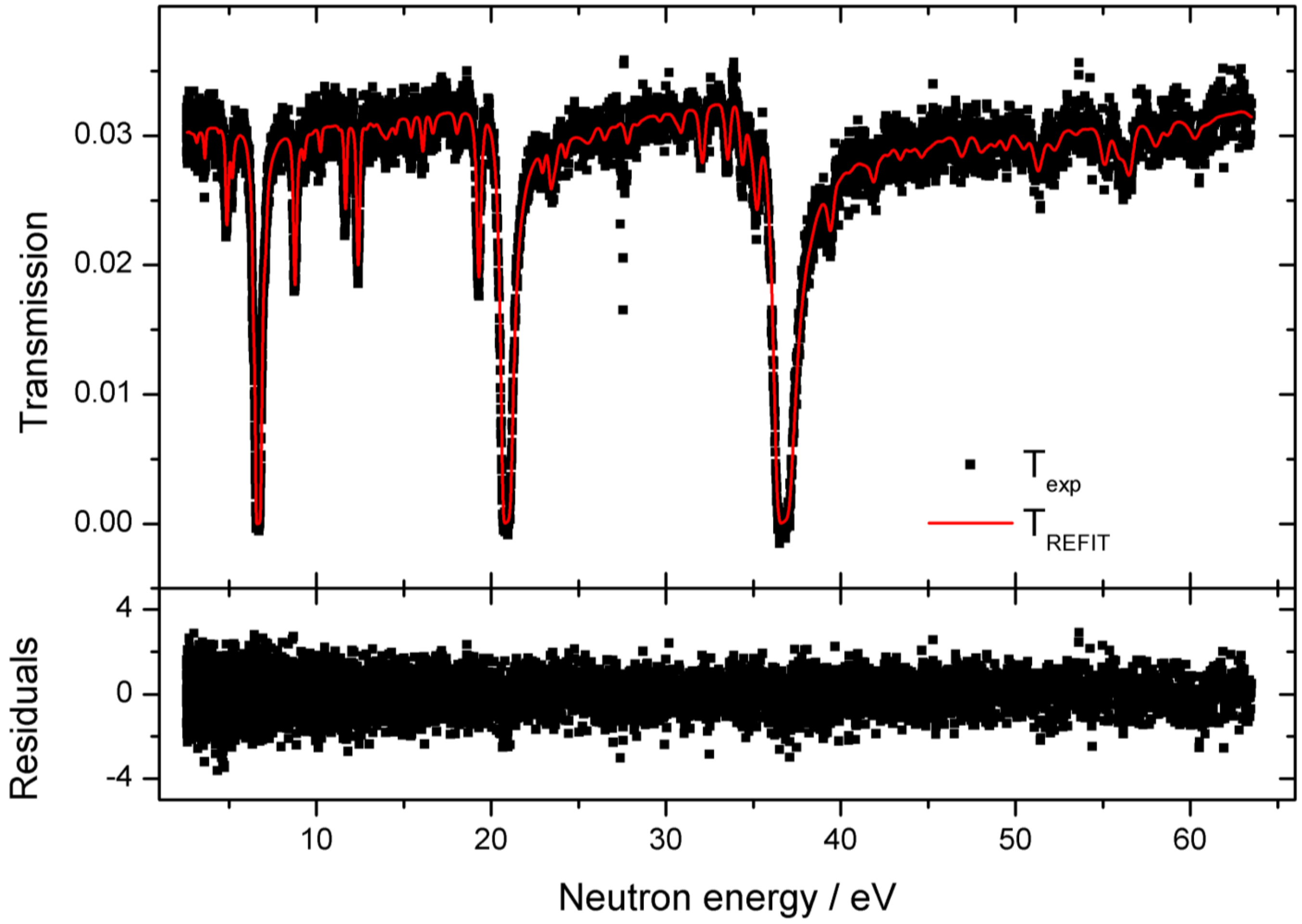
- Strong impact of matrix material
- **Beam attenuation due to matrix ~ 99%**

CBNM standard: U

U_3O_8 reference sample
EC NRM 171

Fit for $^{235,238}U$ areal density
+

$$n_X\ \sigma_{tot,X}(E) = a_X + \frac{b_X}{\sqrt{E}}$$



CBNM standard: U

Recent ENDF/B-VIII.0 evaluation for U-235.
NRTA uncertainties only due to counting statistics.

NRM 171	U5/U8 (declared)	U5/U8 (NRTA)	Ratio
031	0.003206	0.00313 (33)	0.976 (99)*
070	0.007209	0.00812 (80)	1.127 (125)*
194	0.019664	0.02012 (23)	1.023 (12)
295	0.029857	0.03012 (28)	1.009 (9)
445	0.045168	0.04627 (68)	1.024 (15)

*Measurements with reduced irradiation time

European Commission

Summary

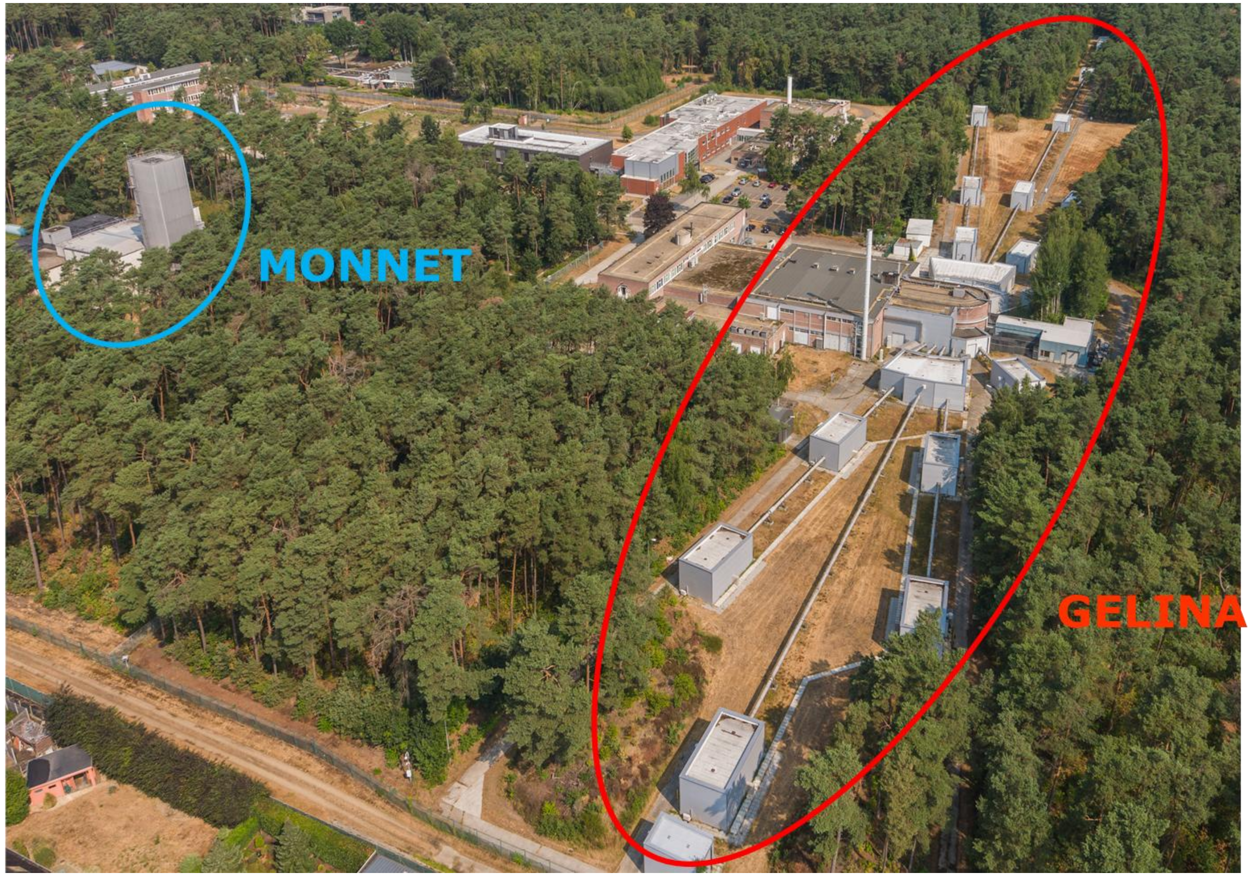
- NRTA is a powerful non-destructive analysis technique
  - Based on well-established methods for cross section measurements
  - Applicable for high radioactive material
  - Absolute method, does not require calibration measurements with representative samples
  - Method validated at GELINA for short flight paths (poor energy resolution)
  - Accurate analytical method (uncertainty < 3% whenever accurate cross sections are available) even at poorer resolution station (short flight path station)
- $\Rightarrow$ Various applications: nuclear safeguards, security, forensics …

General Remarks

Project on characterisation of spent fuel:
together with SCK-CEN, SKB, …

EUFRAT:
open access to facilities at JRC GEEL

ENEN+:
funding scheme for training (bachelor, master, PhD students and postdocs)



Any questions?

You can find me at carlos.PARADELA-DOBARRO@ec.europa.eu

European Commission