Neutron Capture and Waste Transmutation

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Layout of the lectures

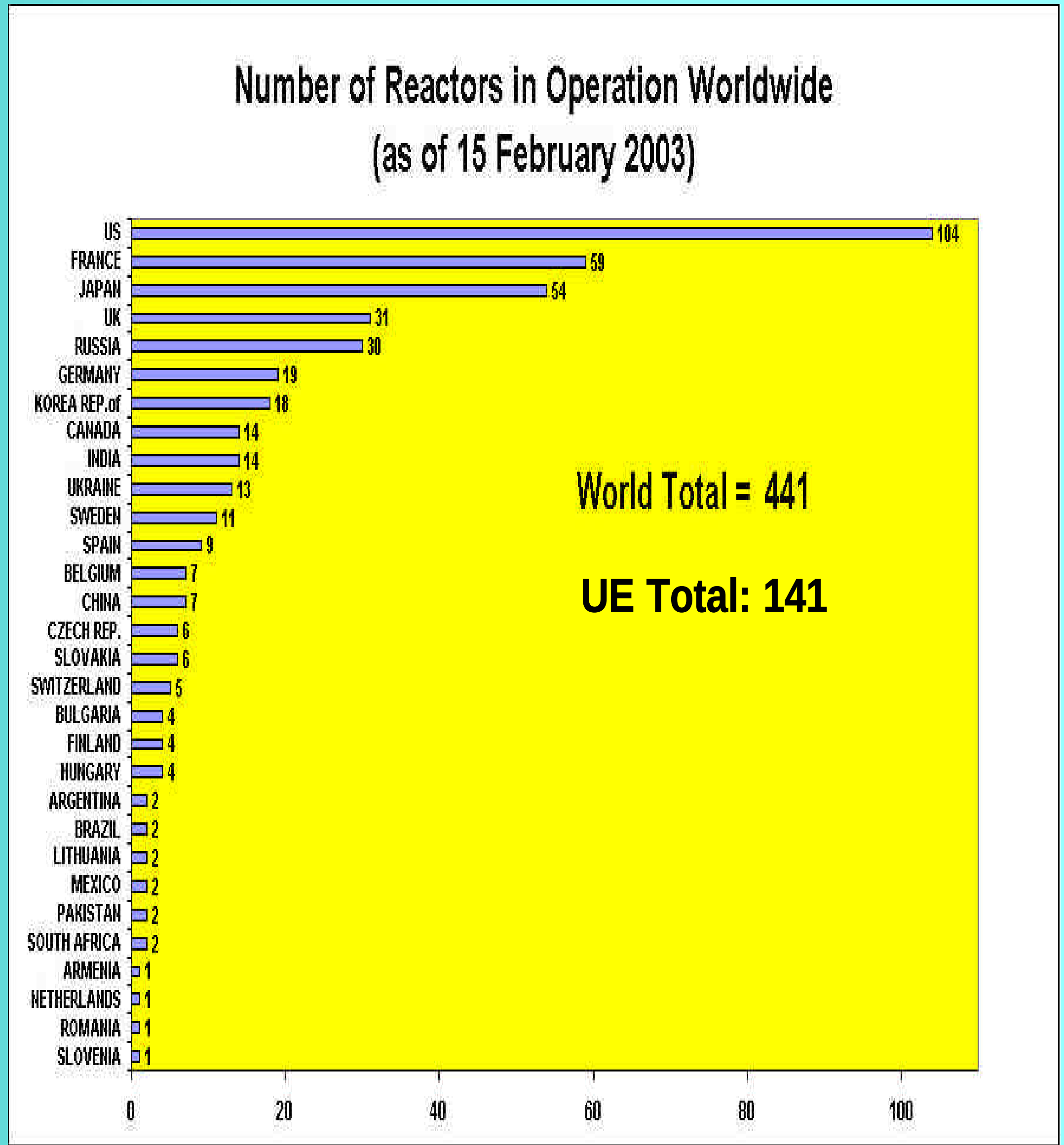
- Transmutation of radioactive waste, ADS & Nuclear Data (☞ J. Benlliure)
- Neutron capture: theory and practice

Nuclear Waste, Transmutation, ADS & Nuclear Data

Nuclear power plants in the world:

- **441 plants in operation,**
32 under construction

http://www.iaea.org



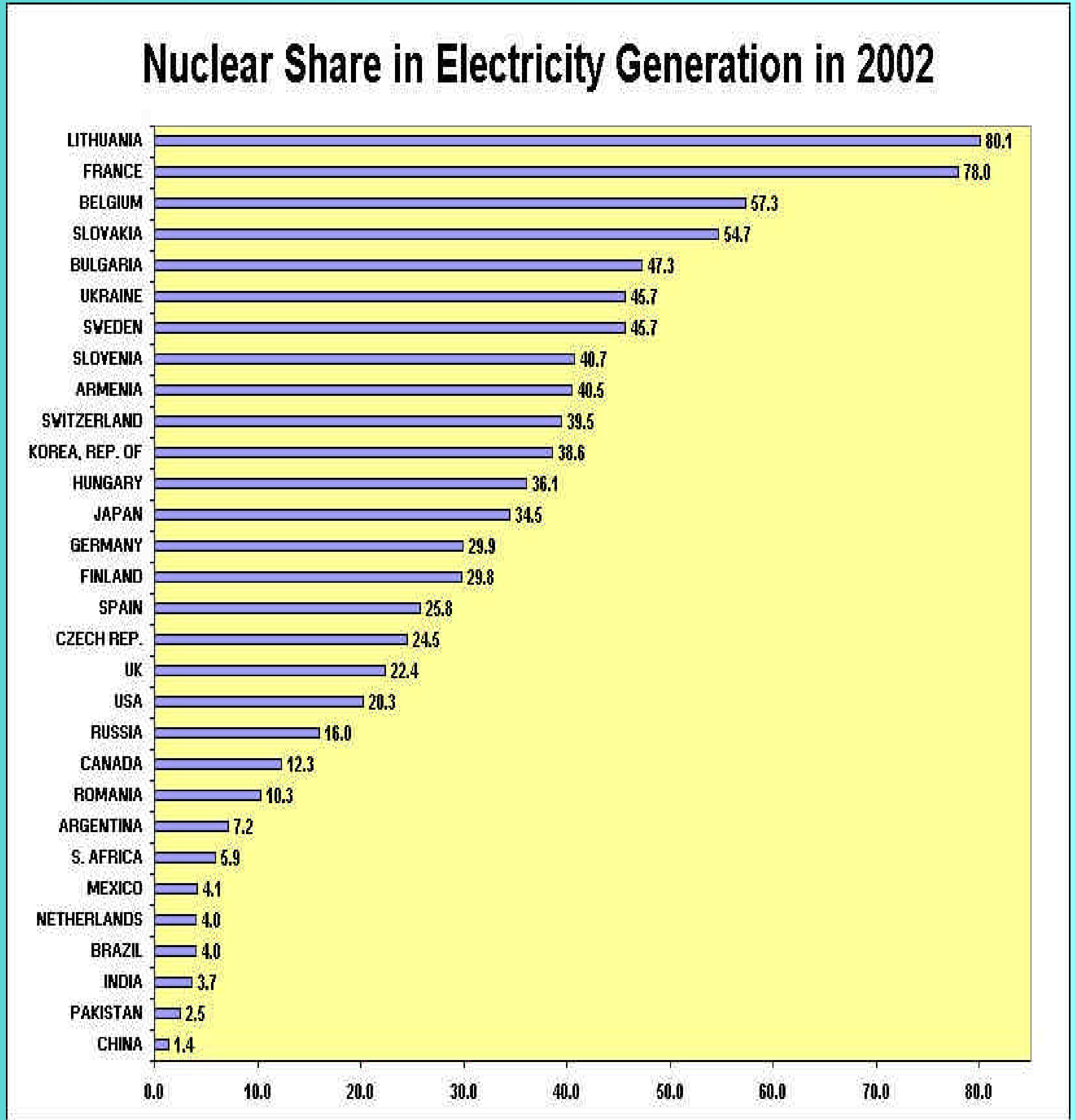
Nuclear electricity generation:

Total:

- **Installed: 359 GW(e)**
- **Produced: 2574 TWh** (81.7% availability)

Share:

- **World average: 17.3%**
- **EU average: 33.6%**



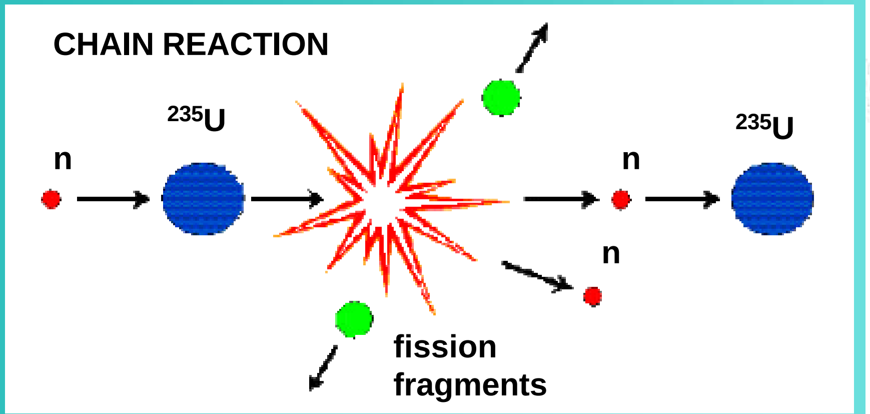
Nuclear Fission Reactors

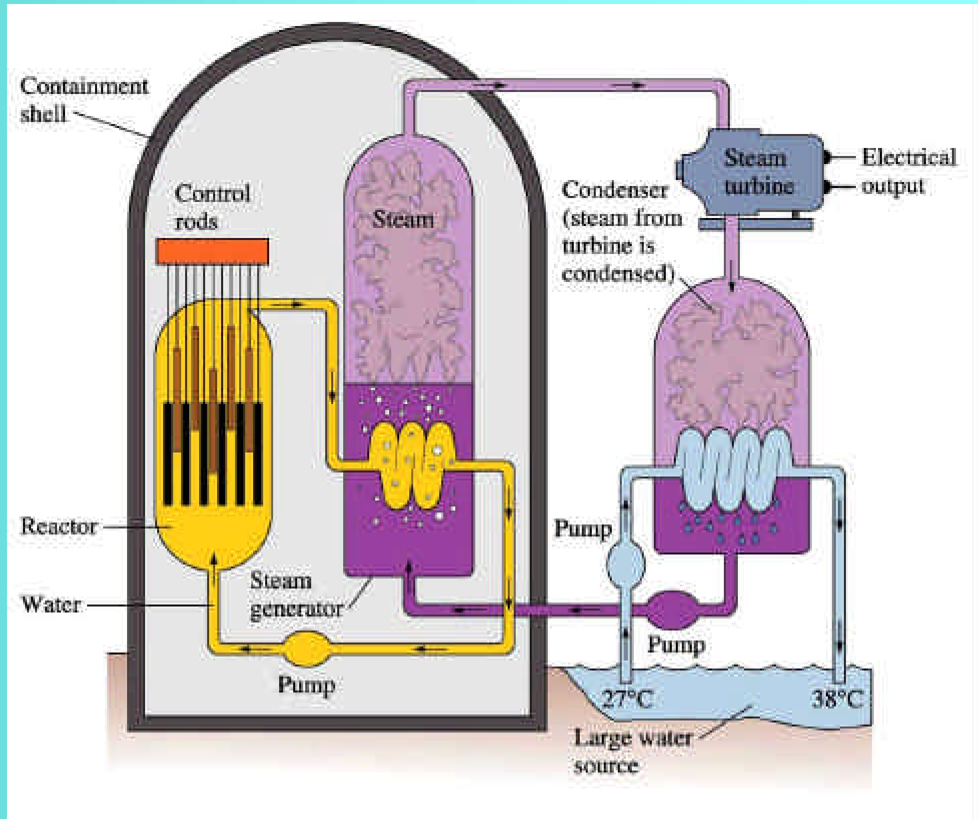
Most reactors in use are of Light Water Reactor type, either Boiling Water Reactors or Pressurized Water Reactors:

Fuel: UO_2 , 2- 4% ^{235}U

Moderator-Coolant: H_2O

n-spectrum: thermal





Reactors operate at CRITICAL POINT (neutron balance) which must be kept: REACTOR CONTROL

Nuclear waste generation:

The loaded fuel is transformed during the energy generation process:

n + ^{235}U ® 3n + ^{134}I + ^{99}Y; FISSION

^{134}I(52min) ® ^{134}Xe.

Fission Product

^{99}Y(1.5s) ® ^{99}Zr(2s) ® ^{99}Nb(15s) ® ^{99}Mo(66h) ® ^{99}Tc(2.1×10^{5}y)

n + ^{238}U ® ^{239}U; CAPTURE

^{239}U(23.5min) ® ^{239}Np(2.4d) ® ^{239}Pu(2.4×10^{4}y)

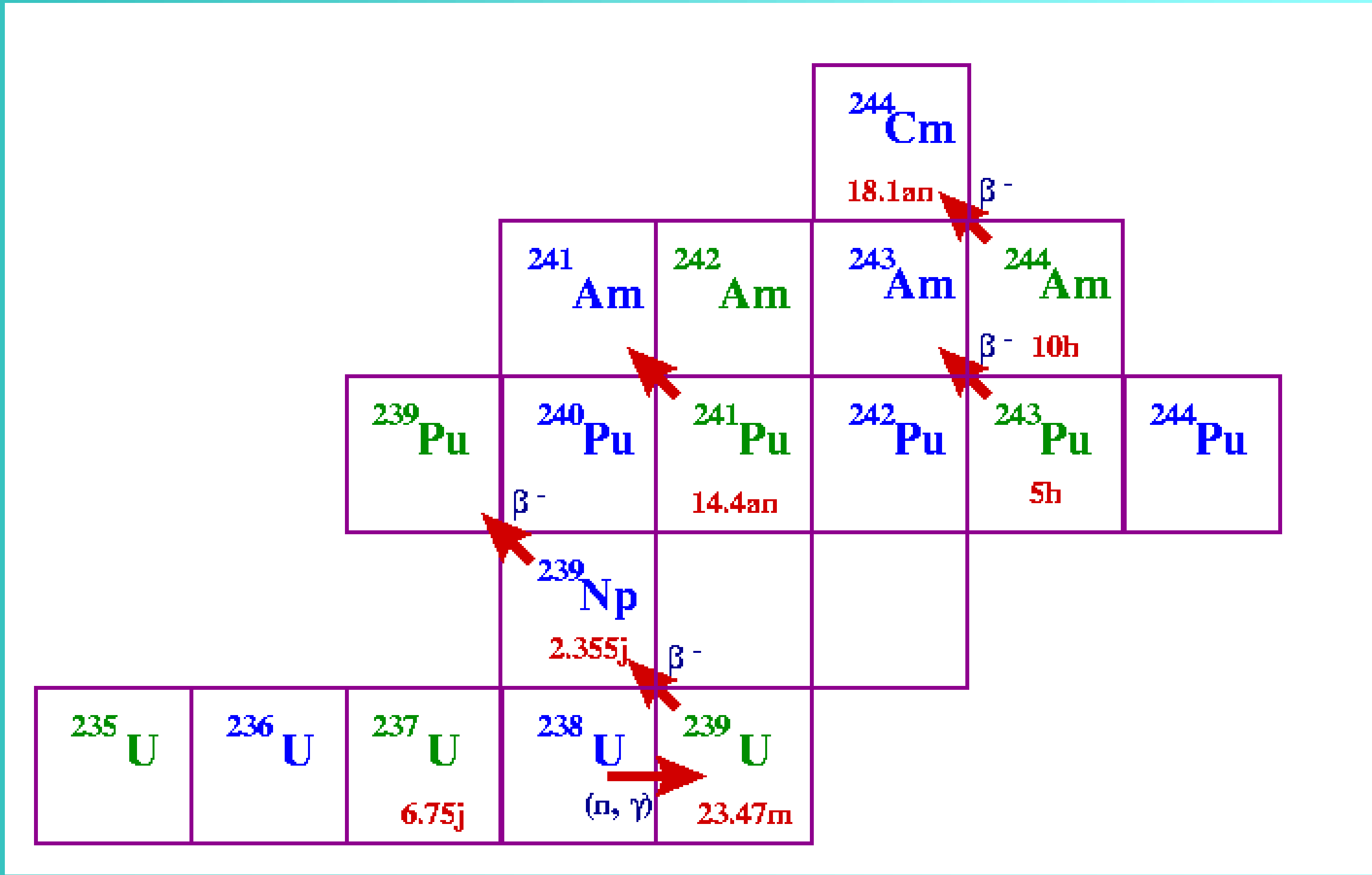
TRans-Uranics

n + ^{239}Pu ® ^{240}Pu(6.5 ×10^{3}y); n + ^{240}Pu ® ^{241}Pu(14.4y) ® ^{241}Am(432y)

Minor Actinides

Actually a complex set of reactions will take place…

Partial reaction chain of the U-Pu cycle:



As a consequence an inventory of long lived highly radioactive isotopes builds up in a reactor ∀ High Level Waste

A 1GWe (3GWth) Pressurized Water Reactor, producing 7TWh (80% avail.), burns 1 ton/year fissile material.

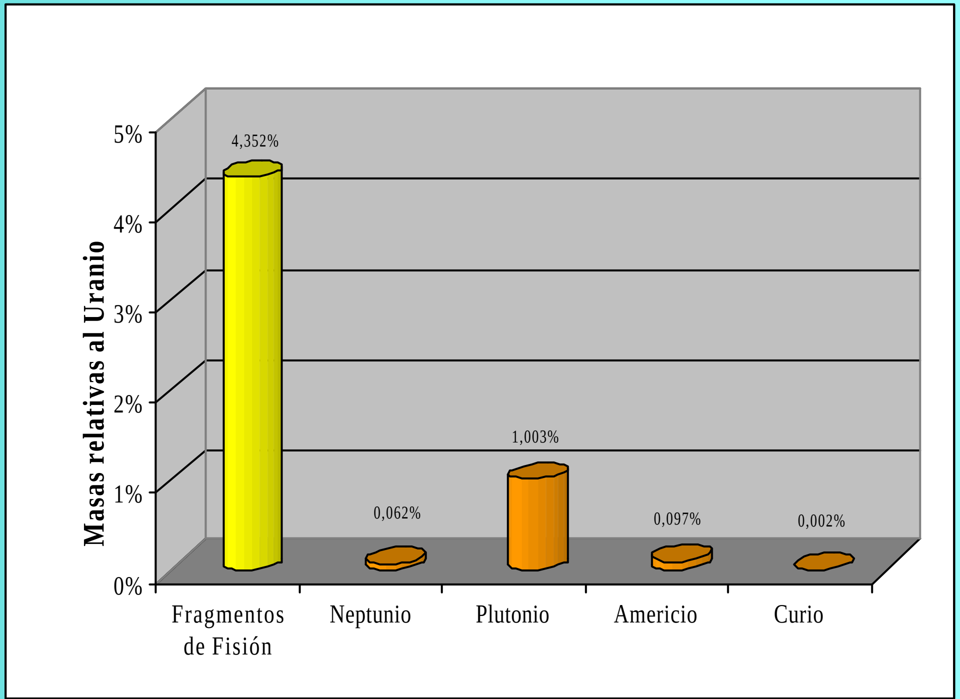
Loaded with 27.3 Ton of 3.5% enriched UO2 (954kg ^{235}U) produces after a burn-up of 33GWd/ton (~1 year):

266 kg Pu (156kg ^{239}Pu)

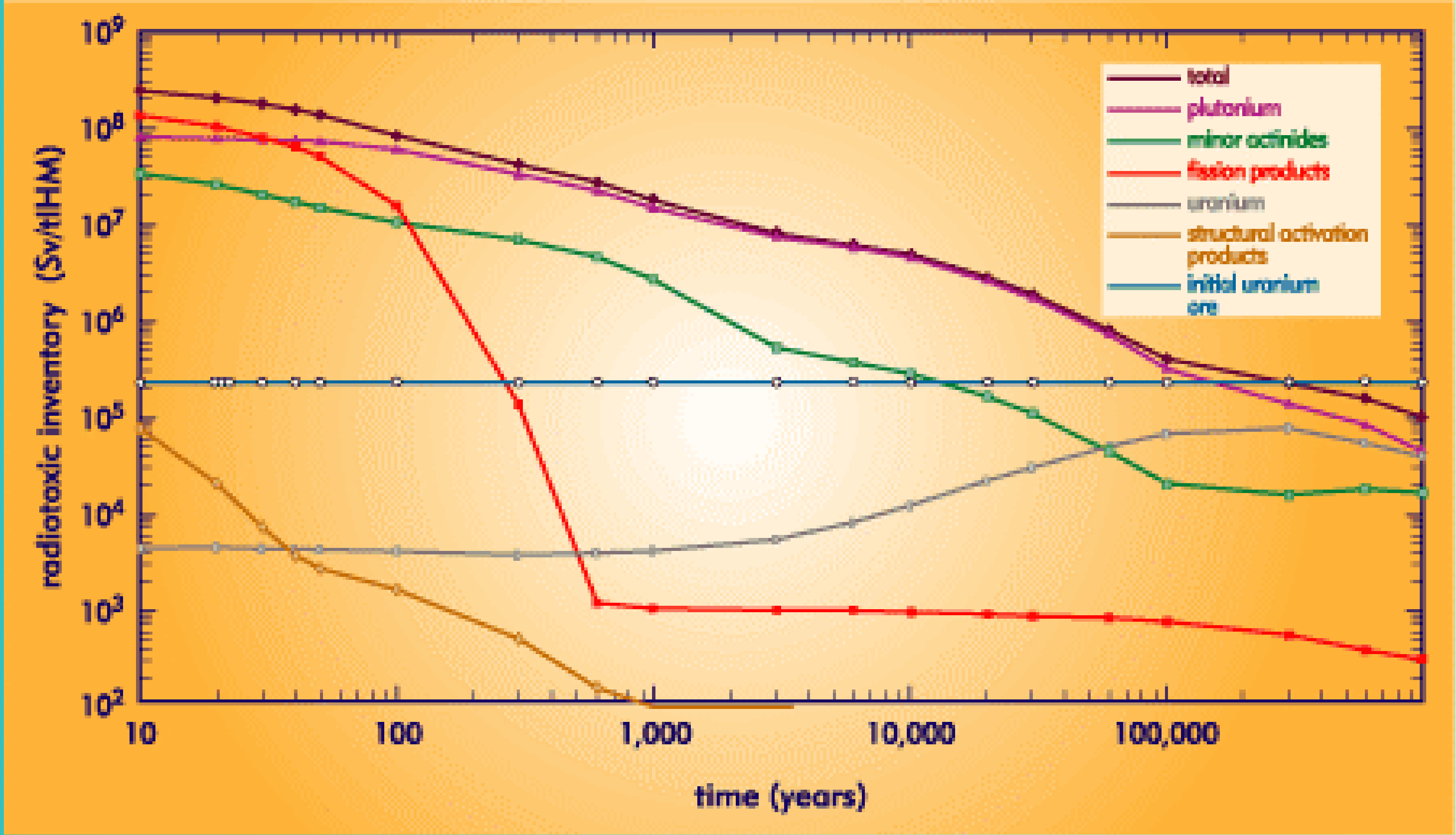
20 kg MA

946 kg FP (63kg long-lived FP)

and still contain 280kg ^{235}U plus 111kg ^{236}U



The hazard they represent can be measured by the evolution of their radio-toxicity:



One possible strategy to reduce the hazard of HLW in the long term is provided by transmutation (and/or incineration):

Actinides:

- **Fission:**

  n + ^{243}Am(7.4×10^{3}y) ® FF's + n's

- **Capture (+ Decay) + Fission:**

  n + ^{243}Am ® ^{244}Am(10.1h) ® ^{244}Cm;

  n + ^{244}Cm(18.1y) ® FF's + n's

Long-lived FP:

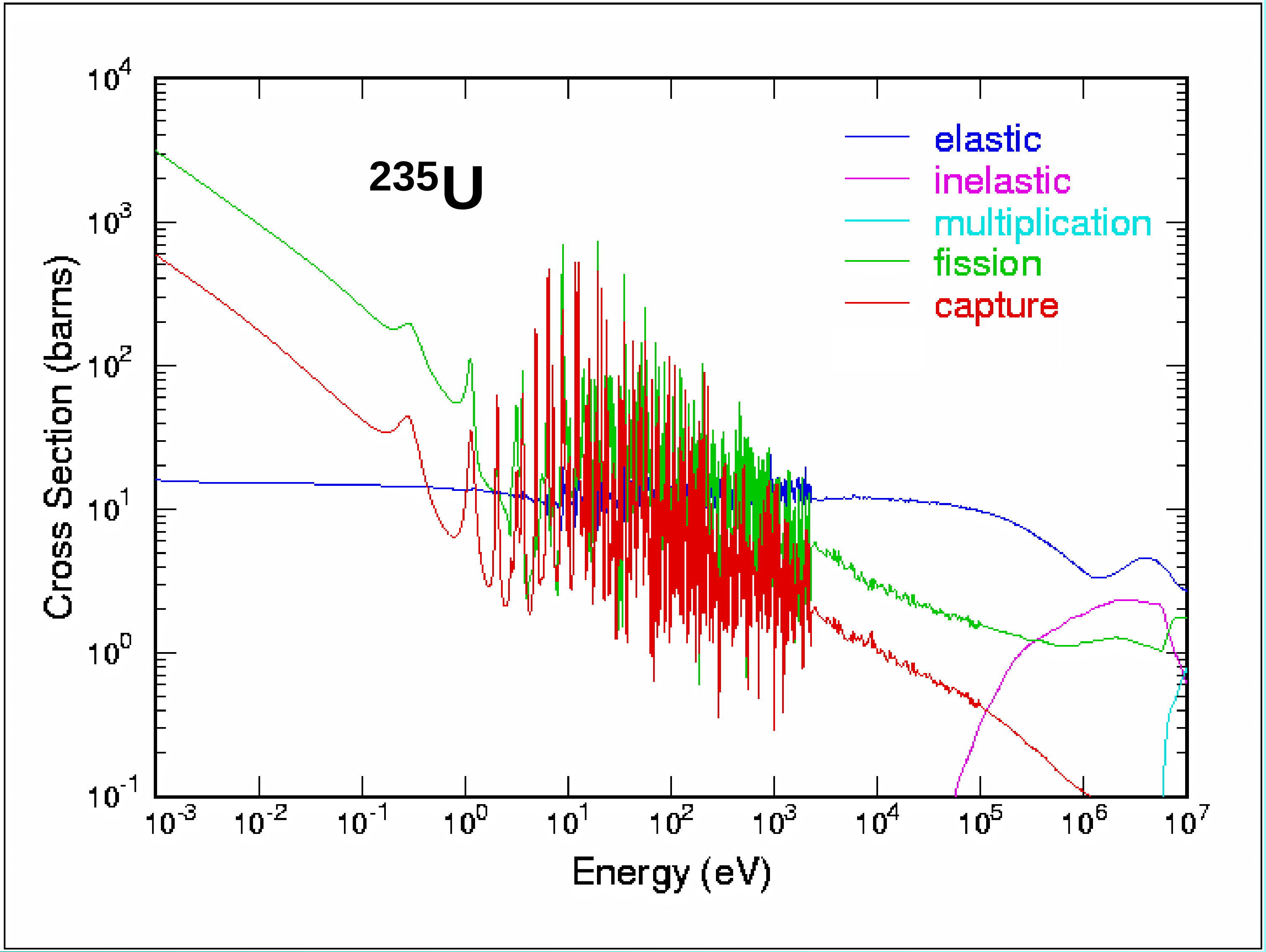
- **Capture:**

  n + ^{99}Tc(2.1×10^{5}y) ® ^{100}Tc(15.8s) ® ^{100}Ru.

Is it possible to use the same reactions that create the waste to destroy it?

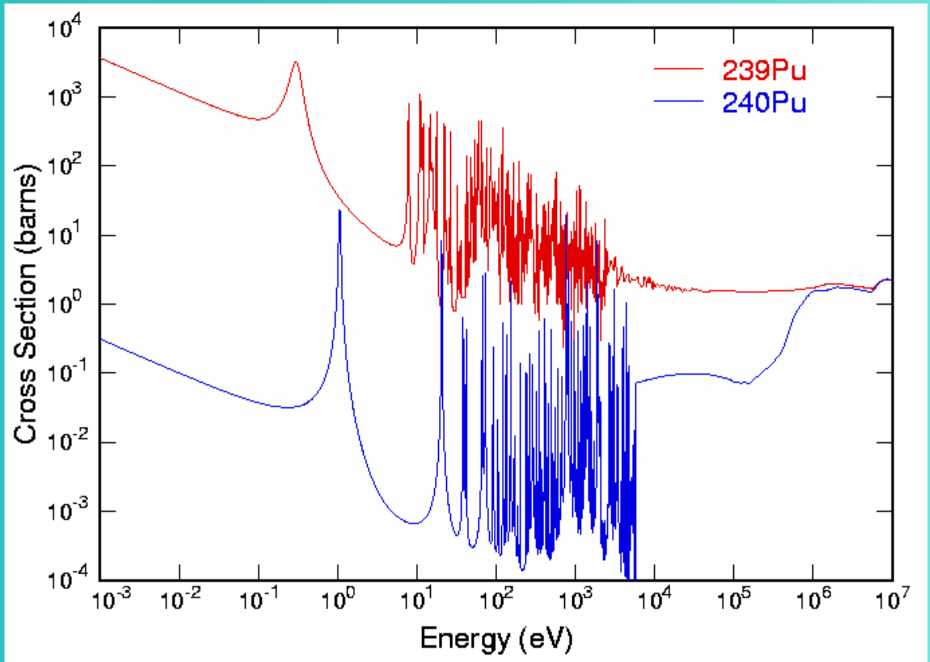
The key point is the separation of the different components from spent fuel (or partitioning)

Neutron induced reactions: strong energy dependence …

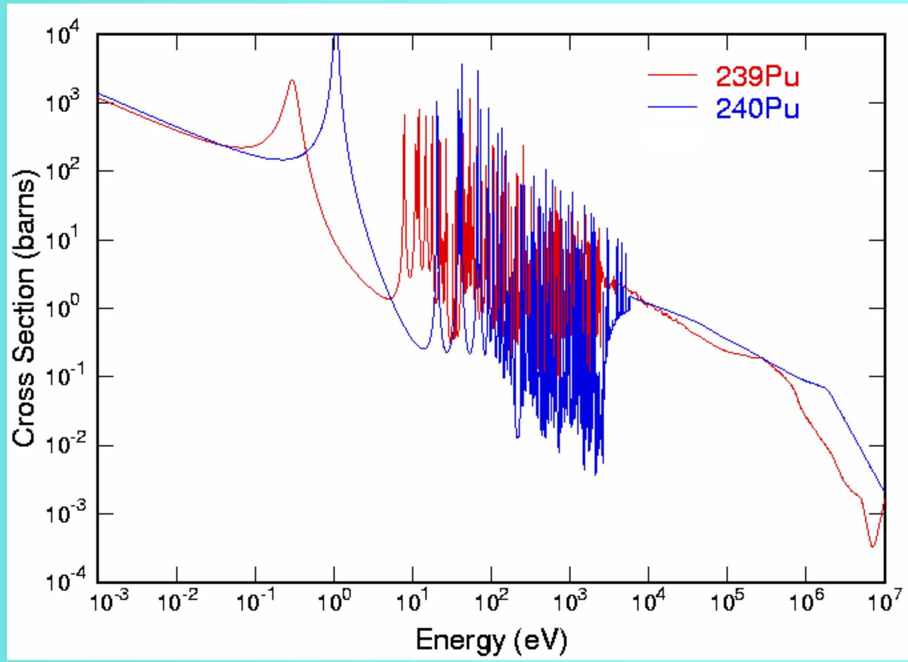


…and isotope dependence:

Fission:

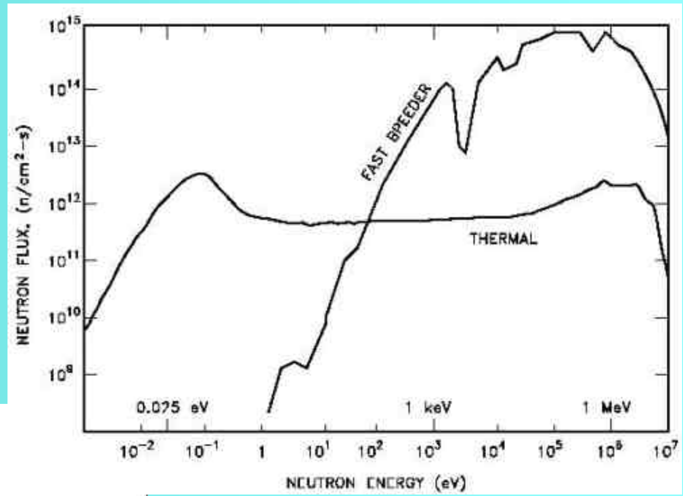


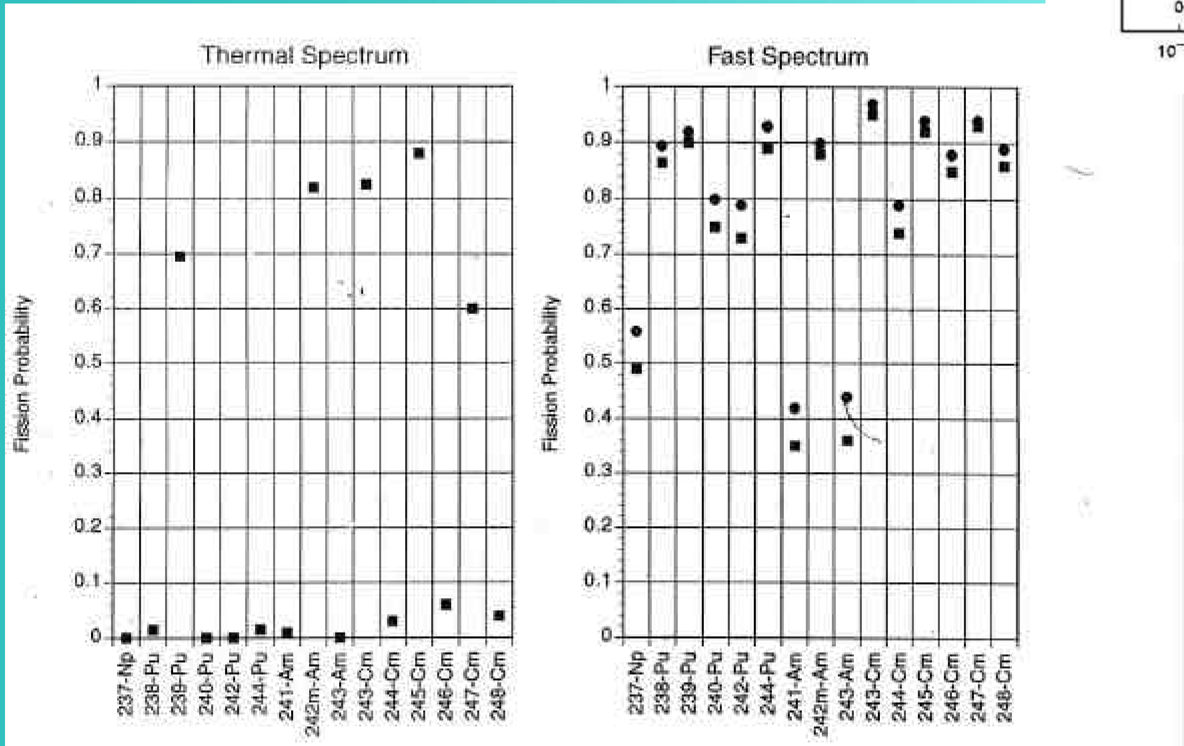
Capture:



Therefore there are several possible (from physics point of view) solutions to the problem of "burning" HLW

For example, the **burning of TRU** is favoured by **“fast” n spectrum** ...



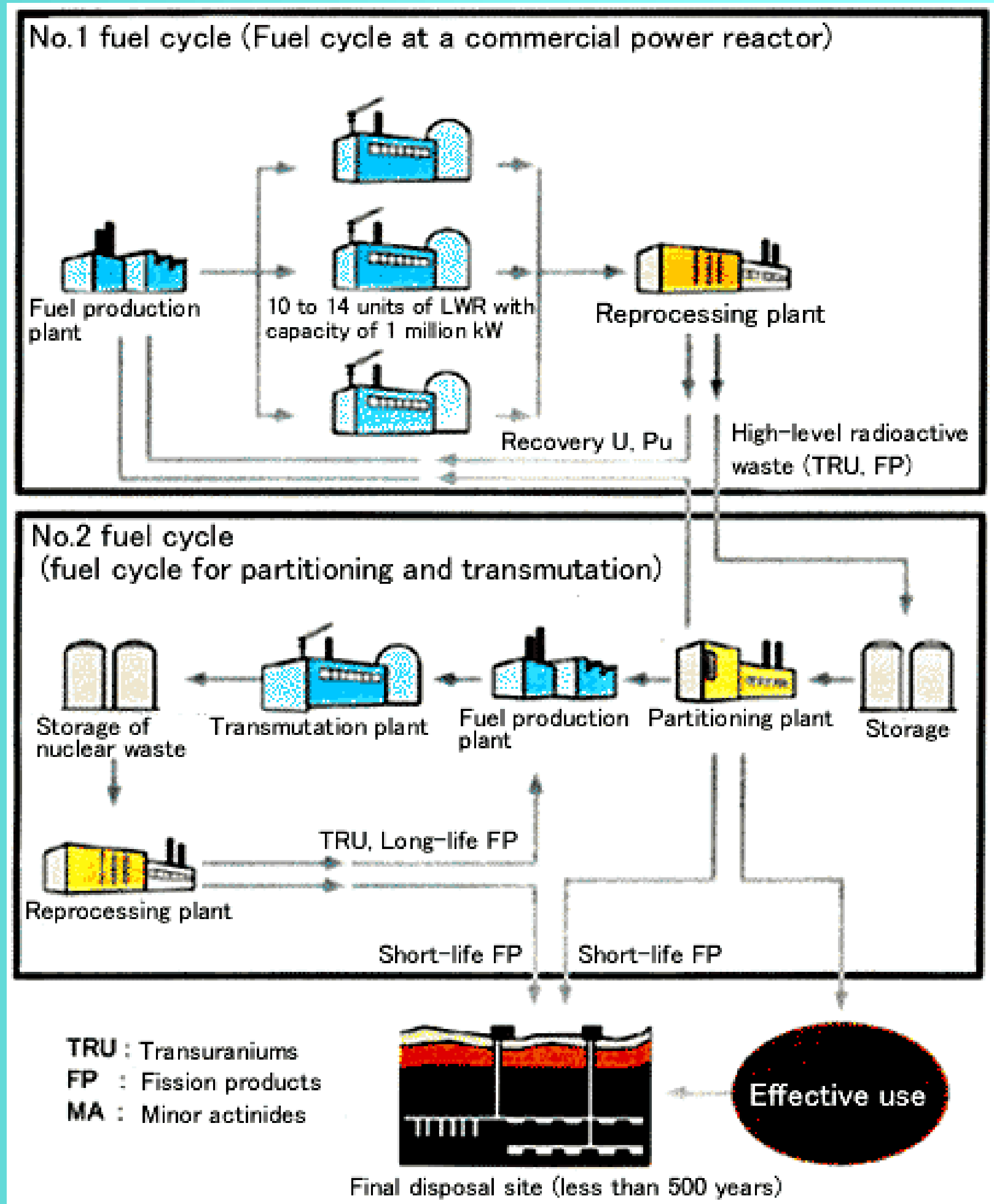


Only Actinides
T > 10 years are shown

In any case a surplus of n are needed to transmute …

The scientific considerations, together with technological, military and political considerations has lead to the proposal of different schemes for the management of HLW.

A currently considered scheme is **the double strata scenario:** ☞

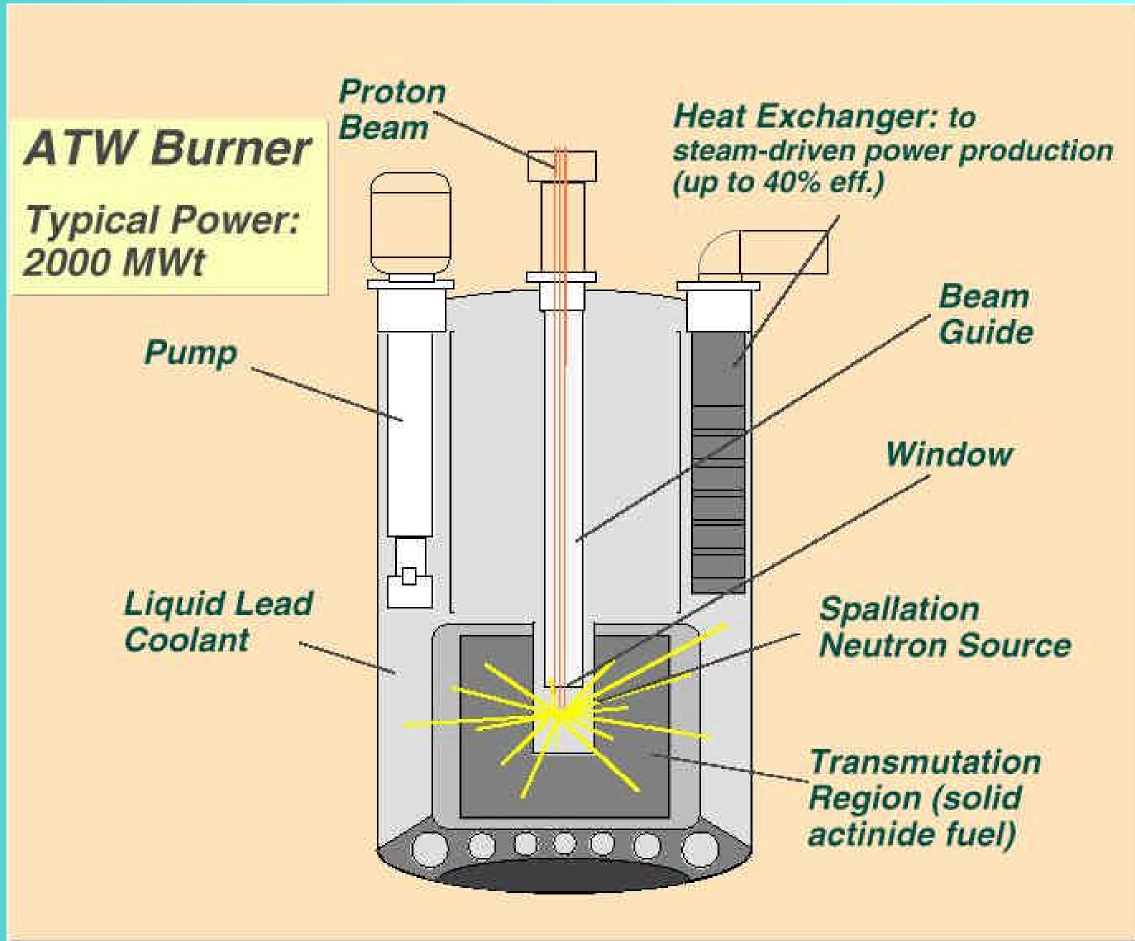


Accelerator Driven Systems:

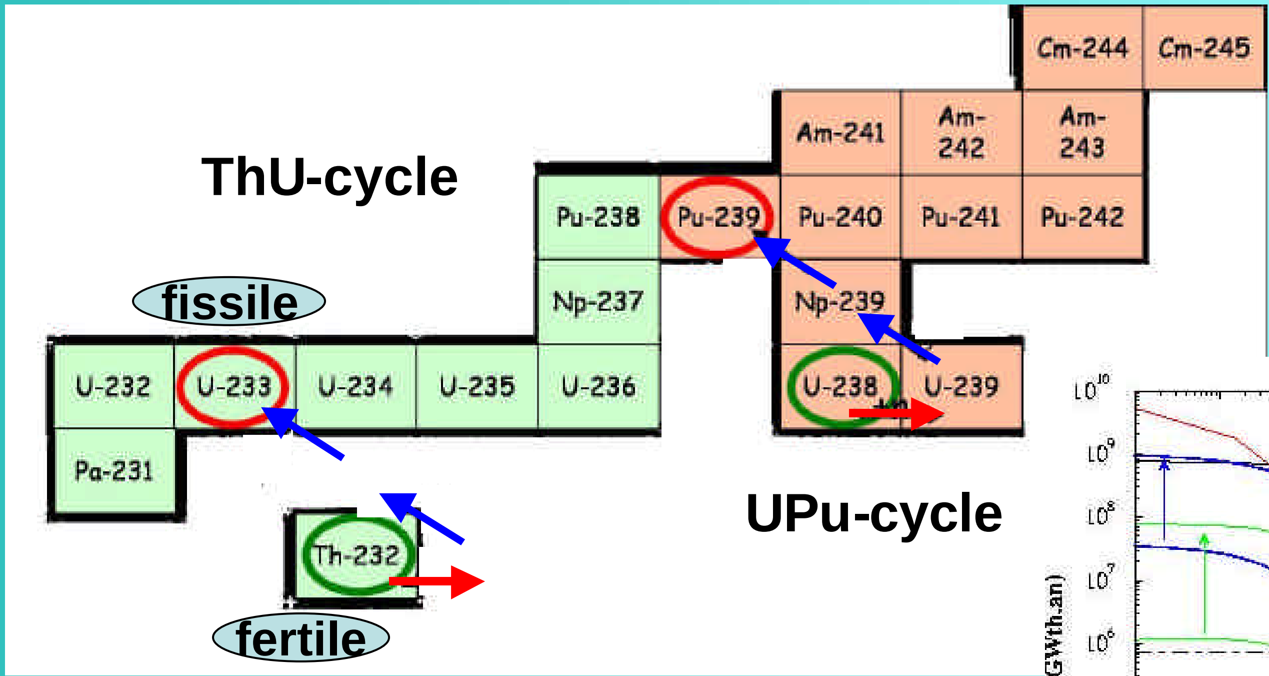
A nuclear reactor with a subcritical core which uses an accelerator to produce the neutrons necessary to maintain the chain reaction:

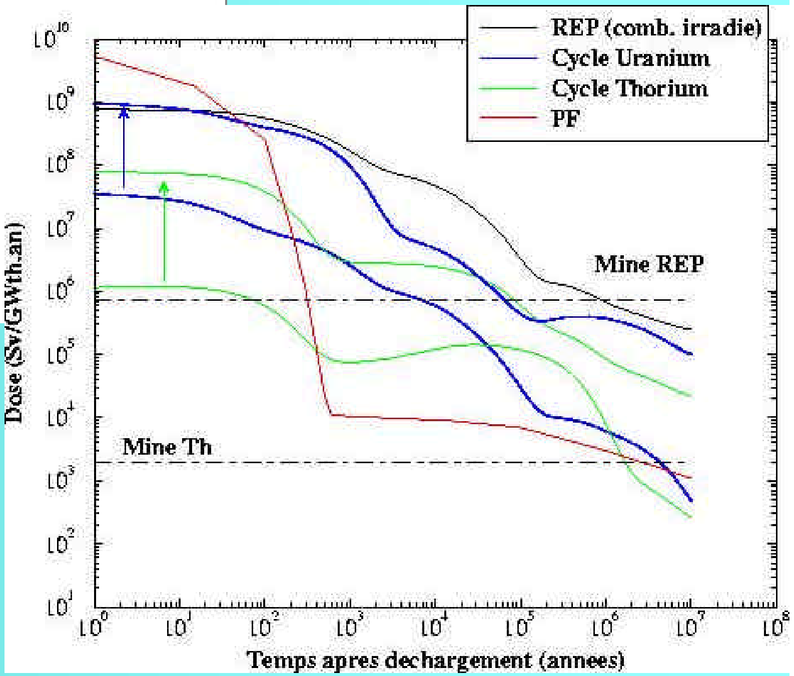
⇨ **Advantage: less dependence on fuel composition**

For example: a very high intensity ~1 GeV proton accelerator producing neutrons by spallation on a molten Pb or Pb/Bi target which acts also as the coolant, with solid actinide fuel.



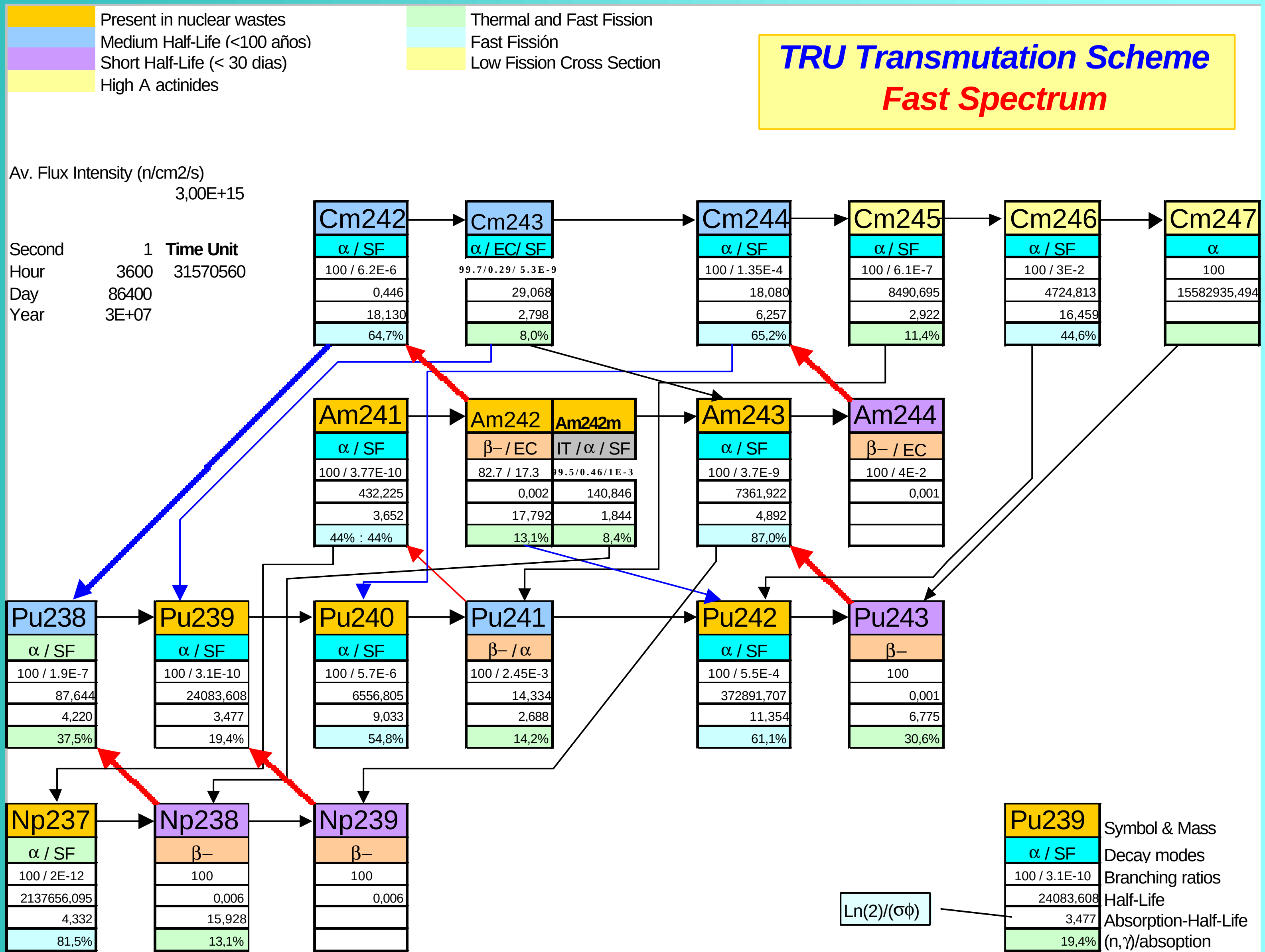
An ADS can also be utilized for energy production and if based on Th-U fuel cycle, will generate a reduced amount of TRU ...



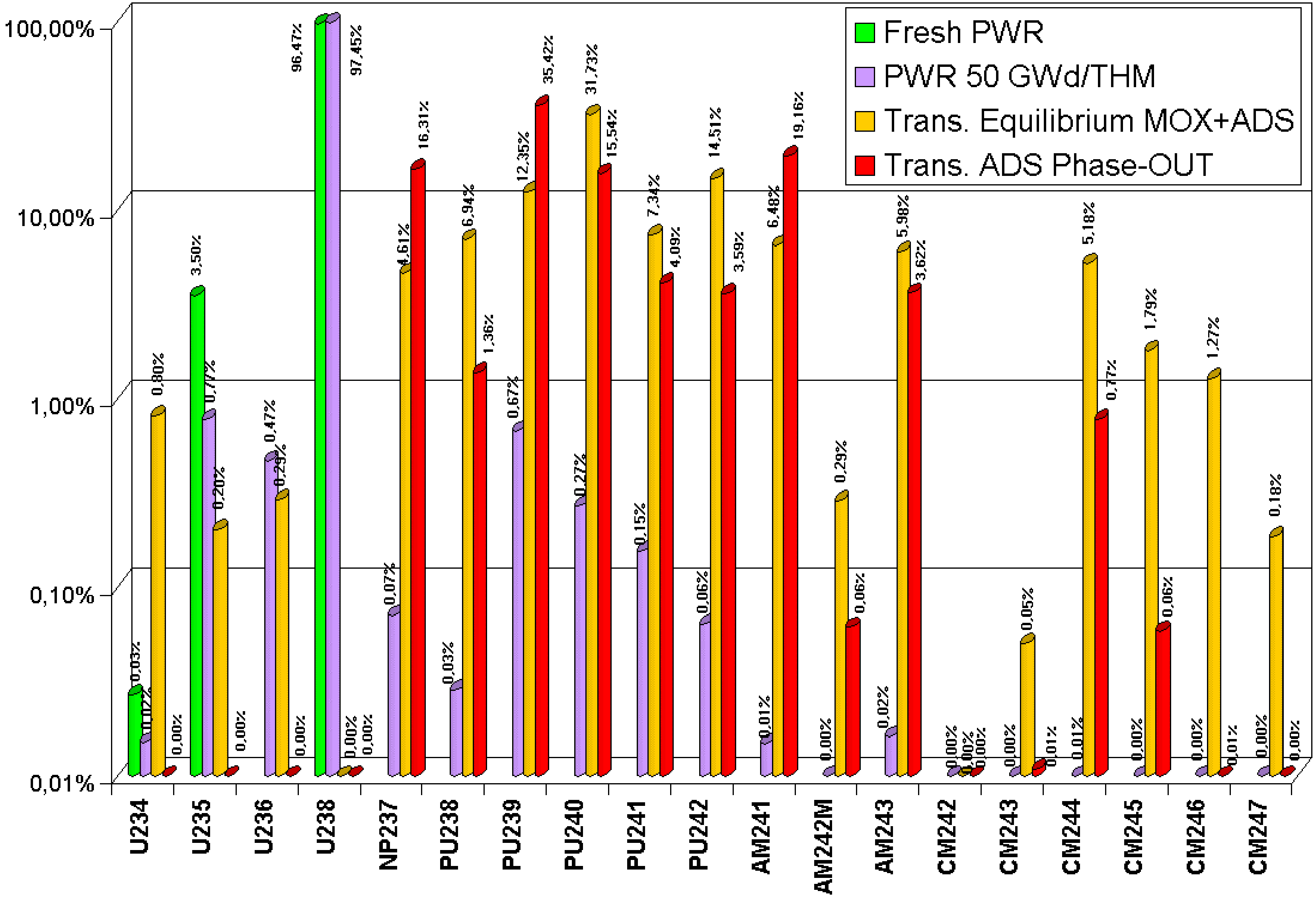


⇨ **Energy Amplifier**

Transmutation of TRU: set of reactions



Transmutation of TRU: new fuel compositions



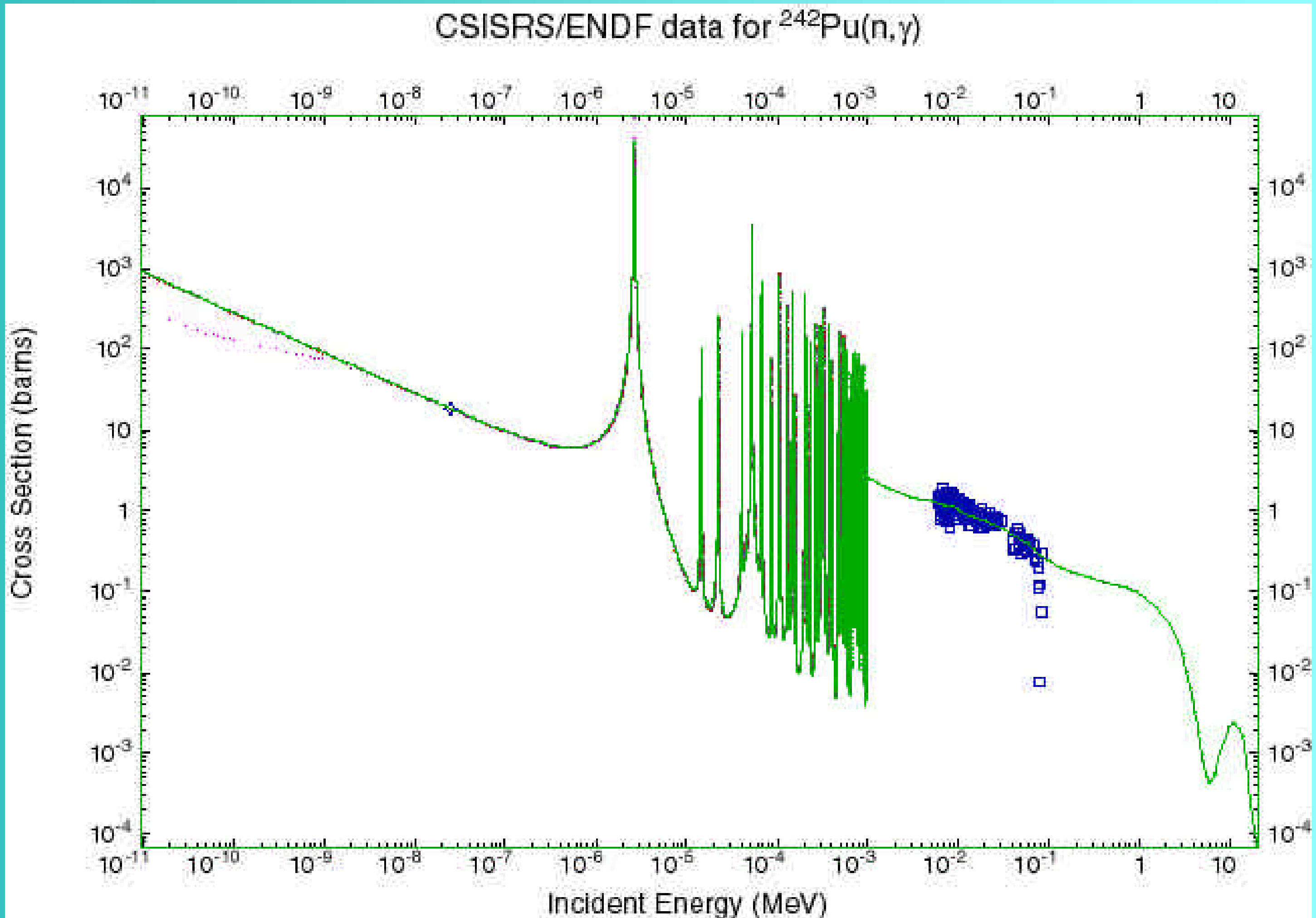
Challenges in the field of ADS:

- **Waste separation (or partitioning) methods**
- **Design of a high power accelerator, the spallation target and their coupling**
- **Study of reactor core behaviour and transmutation rates**

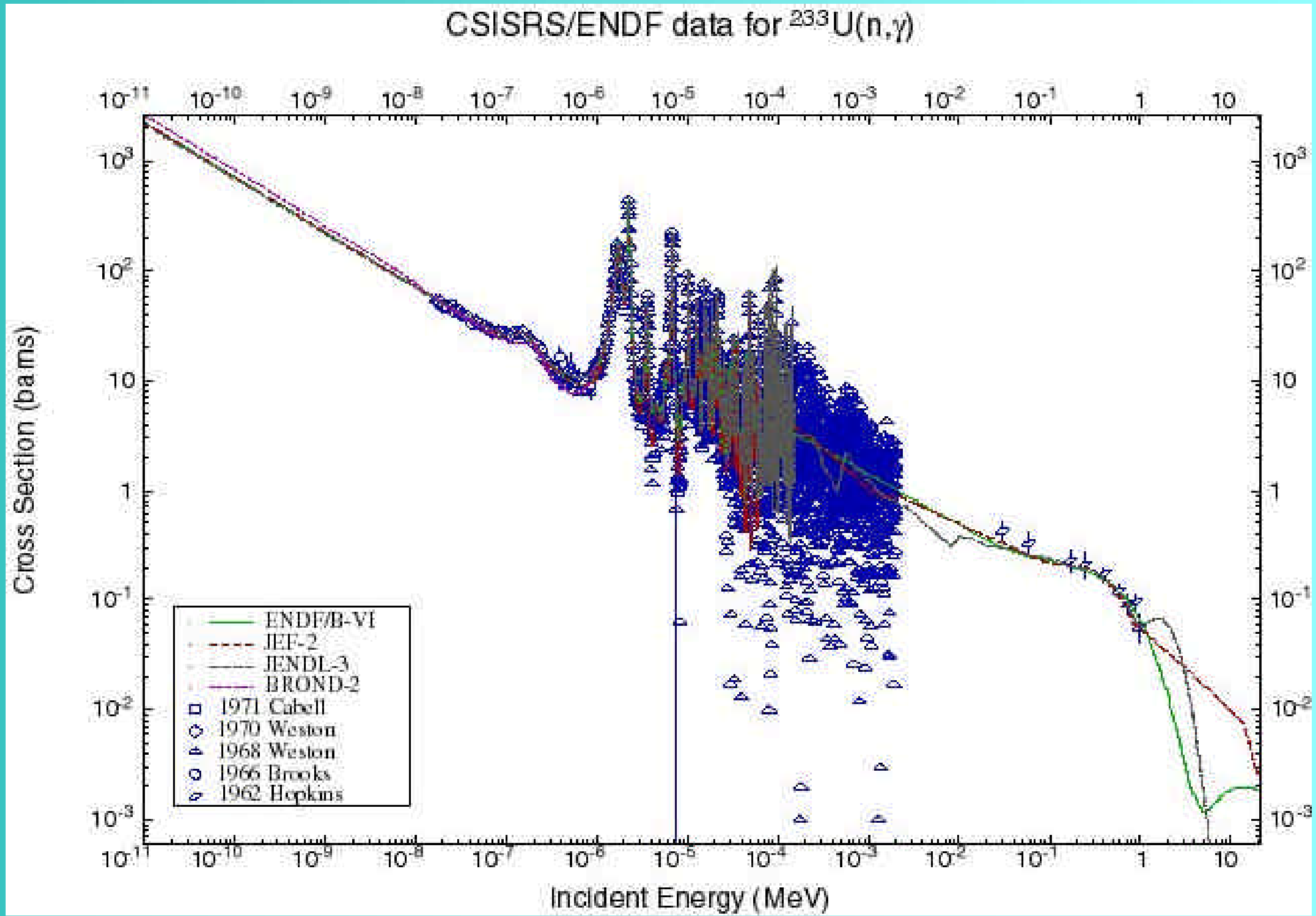
Need for new or improved accuracy nuclear data:

- **proton spallation reaction: n yield, residues (☞ J. Benlliure)**
- **neutron induced reactions: fission, capture, (n,xn),… on actinides, fission products and structural materials**

An example of poorly known reaction…



…another example of not so poorly known



OBJECTIVES of the "*n_TOF-ND-ADS*" EC Project

- Measure with a precision of few % the appropriate **capture**, **fission** for elements with relatively well known cross-sections (^{197}Au, $^{24\text{-}26}$Mg, ^{207}Pb, ^{56}Fe, ^{235}U and ^{238}U) though their knowledge at high energies is still limited.
- Determine with a precision of few % the **capture cross sections** for the isotopes, relevant to the Th-cycle: **^{232}Th, ^{231}Pa, ^{233}U, ^{234}U, ^{236}U**.
- Determine with a precision of few % **the capture cross sections** for the transuranic isotopes: **^{237}Np, ^{240}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{245}Cm.**
- Determine with a precision of few % the **capture cross sections** of specific LLFF as **^{151}Sm, ^{99}Tc, ^{129}I, ^{79}Se, ^{93}Zr** and further on **^{205,206,207}Pb and ^{209}Bi**.
- Determine with a precision of few % the **fission cross sections** of: **^{232}Th, ^{231}Pa, $^{233\text{-}236}$U, ^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm and ^{245}Cm**.
- Precise measurement of **(n,xn) cross sections** using also activation techniques of: **^{233}U, ^{232}Th, ^{231}Pa, ^{239}Pu, ^{241}Pu, ^{241}Am, ^{243}Am, ^{237}Np and ^{207}Pb.**
- Measure the **total cross sections** of: **^{237}Np, ^{129}I, ^{239}Pu and ^{240}Pu**.
- Measure capture and fission cross sections at **given neutron energies with mono-energetic beams** of the isotopes: **^{232}Th, ^{233}U, ^{237}Np, ^{241,243}Am and ^{99}Tc, ^{129}I, ^{79}Se, ^{151}Sm, ^{137}Cs**.

Neutron radiative capture: Theory and practice

Neutron reactions at low energies

A neutron is absorbed to form a "compound nucleus":

$$n + {}^{A}Z \rightarrow {}^{A+1}Z^{*}$$

which lives for a short time and decays:

${}^{A+1}Z^{*} \rightarrow n + {}^{A}Z$ (elastic)

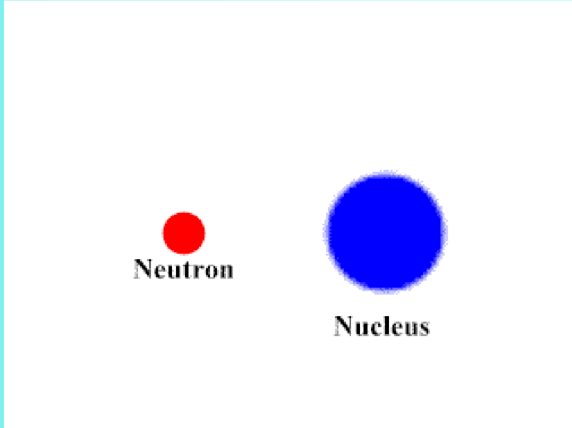
${}^{A+1}Z^{*} \rightarrow {}^{A+1}Z^{*\prime} + \gamma$ (radiative capture)

${}^{A+1}Z^{*} \rightarrow {}^{A1}Z_1^{*} + {}^{A2}Z_2^{*} + xn$ (fission)

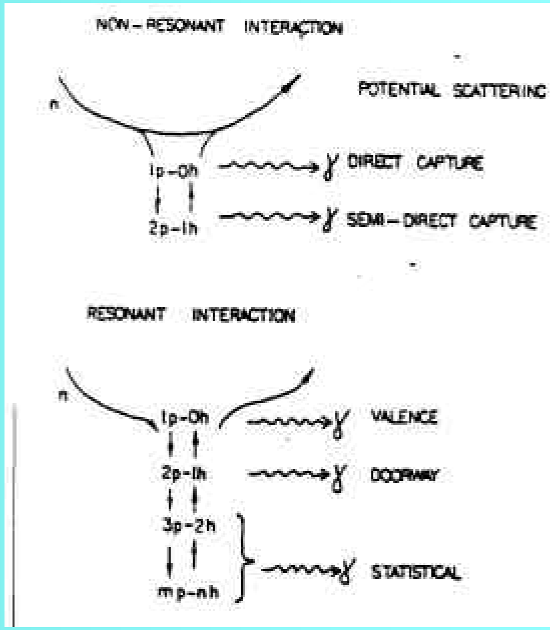
${}^{A+1}Z^{*} \rightarrow n + {}^{A}Z^{*}$ (inelastic)

${}^{A+1}Z^{*} \rightarrow {}^{A+1-x}Z^{*} + xn$ (n multiplication)

…



Other contributions:



The CN formation probability is higher for certain neutron energies E_n corresponding to quasi-bound or virtual states: **resonances**

$$E_R = S_n + \frac{A}{A+1} E_n$$

S_n : neutron separation energy of CN (<10MeV)
® level separation D_0 ~ 1 eV – 100 keV

Life-time « Energy-width: G

$$G = G_n + G_g + G_f + \ldots$$

$$(s = s_n + s_g + s_f + \ldots)$$

G ~ 1 meV – 100 keV

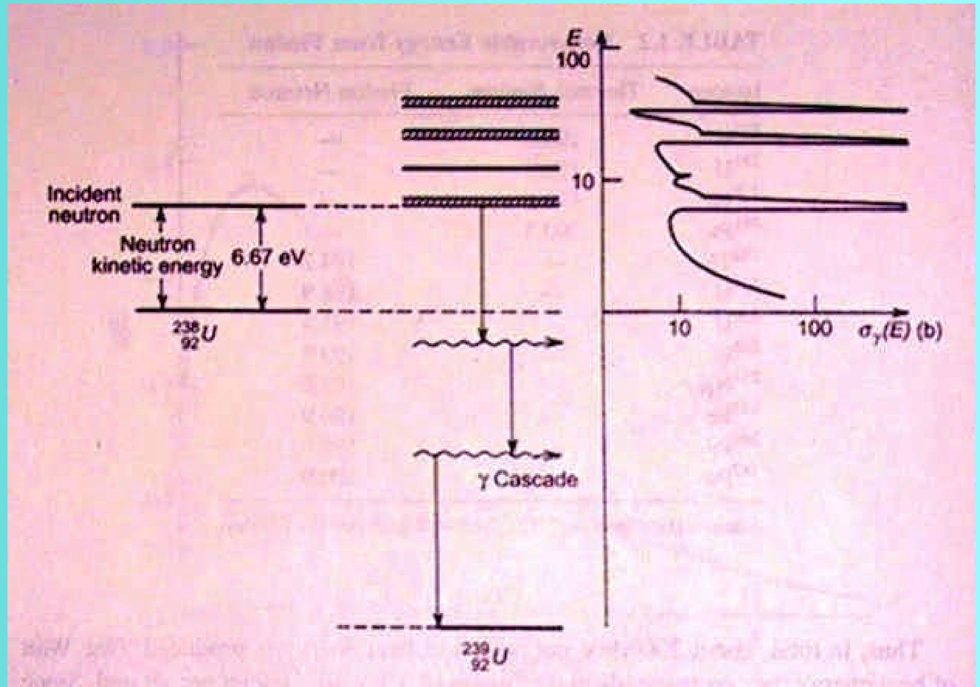


Fig. 1.12 Energy-level diagram for compound nucleus formation. (From Ref. 12; used with permission of Wiley.)

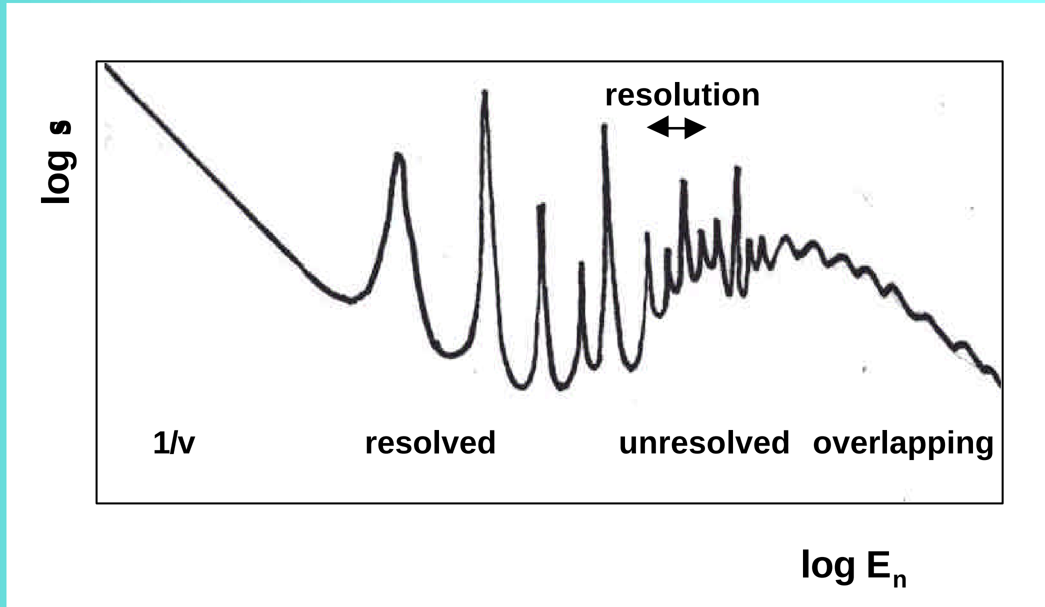
Shape of neutron cross-section

1/v: thermal

G < D0, G > DE: resolved resonance region (RRR)

G < D0, G < DE: unresolved resonance region (URR)

G > D_0 : overlapping resonances



- **In the RRR region, s is described using the R-Matrix formalism, in one of its usual approximations.**
- **In the URR region, average s are described by Hauser-Feshbach statistical theory**
- **It is a parametric approach since nuclear theory cannot predict the values.**
- **Experimental information is strictly necessary.**

Single Level Breit-Wigner Formalism: (n,g)

For ℓ-capture into an isolated spin **J** resonance at E_R:

$$s_g(E) = p ƛ^2 \, g_J \, \frac{G_n \, G_g}{(E-E_R')^2 + \frac{1}{4}G^2}$$

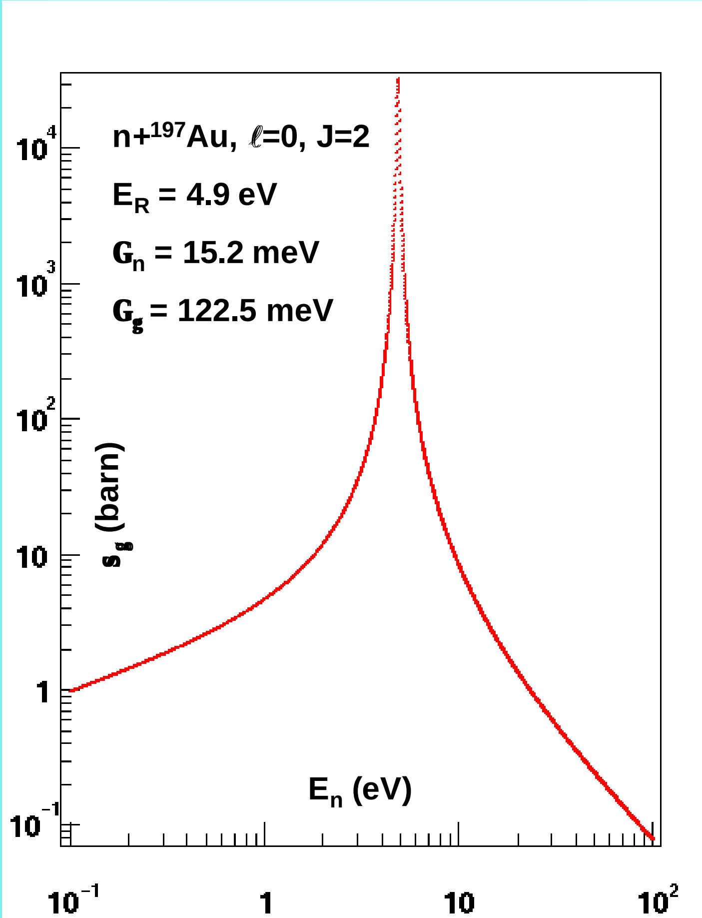
$ƛ = \hbar / \text{Ö}\overline{2mE}$ (neutron wavelength)

$$g_J = \frac{2J + 1}{2(2I + 1)}$$

(spin stat. factor; $|I - \ell \pm ½| \text{ £ } J \text{ £ } |I - \ell + ½|$)

$G(E) = G_n(E) + Gg + \ldots$;   (FWHM)

$G_n(E) = \text{Ö}\overline{E/E_R} \; G_n(E_R)$, ℓ=0



… and the channel radius R_c

SLBW formalism: elastic and capture cross sections

E L A S T I C

$$\sigma = \frac{4\pi}{k^2}\, g_J \left[\sin^2 \Phi_\ell + \frac{\Gamma_n}{\Gamma}\frac{1}{1+\xi^2}\cos 2\Phi_\ell + \frac{\Gamma_n}{\Gamma}\frac{\xi}{1+\xi^2}\sin 2\Phi_\ell \right]$$

(potential, resonant, interference)

C A P T U R E

$$\sigma_\gamma = \frac{4\pi}{k^2}\, g_J \frac{\Gamma_\gamma \Gamma_n}{\Gamma}\frac{1}{1+\xi^2}$$

$$k = \sqrt{2mE} / \hbar$$

$$g_J = \frac{2J+1}{2(2I+1)}$$

$$\xi = \frac{2}{\Gamma}(E - E_R')$$

$$\Gamma_n = \frac{P_\ell(E)}{P_\ell(E_R)}\Gamma_n(E_R)$$

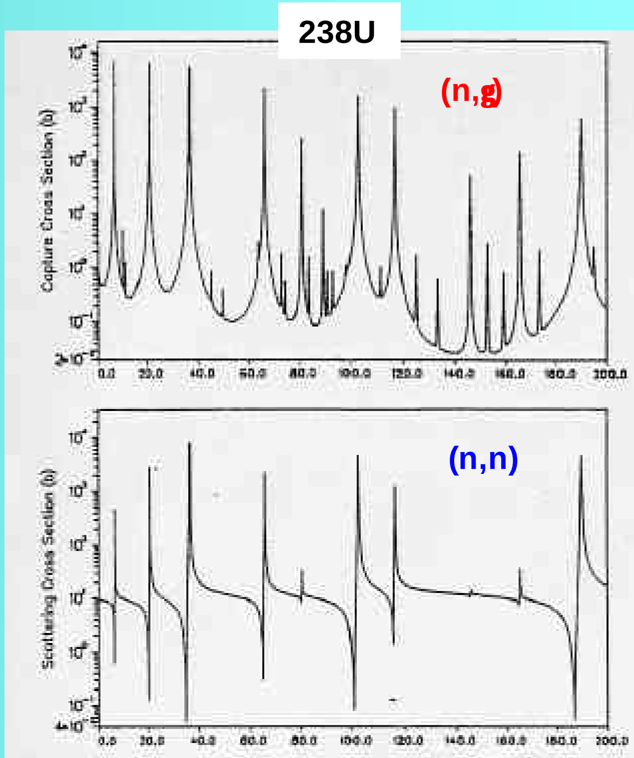
$$E_R' = E_R + \frac{S_\ell(E_R) - S_\ell(E)}{2P_\ell(E_R)}\Gamma_n(E_R)$$

$$P_0 = \Phi_0 = k R_C = \rho, \quad S_0 = 0$$

$$P_\ell = \rho^2 P_{\ell-1} / ((\ell - S_{\ell-1})^2 + P_{\ell-1}^2)$$

$$S_\ell = \rho^2(\ell - S_{\ell-1}) / ((\ell - S_{\ell-1})^2 + P_{\ell-1}^2) - \ell$$

$$\Phi_\ell = \Phi_{\ell-1} - \tan(P_{\ell-1} / (\ell - S_{\ell-1}))$$



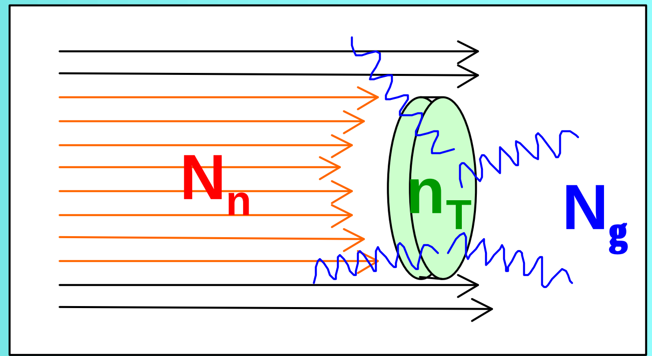
Neutron Reaction Data

- **From the analysis of experimental data on capture, total, fission, … cross-sections, resonance parameters are obtained for every nucleus.**
- **All the information is combined, cross-checked for consistency, etc, in a process called "evaluation" until a recommended set of parameters is obtained.**
- **This information is published in a Evaluated Nuclear Data File using an accepted standard format (ENDF-6)**
- **There exist several files:**
  - **BROND-2.2 (1993, Russia)**
  - **CENDL-3 (2002, China)**
  - **ENDF/B-VI.8 (2002, US)**
  - **JEFF-3.0 (2002, NEA+EU)**
  - **JENDL-3.3 (2002, Japan)**

Measurement of (n,g) cross-sections

$$s_g(E) = \frac{\text{Number of capture reactions}}{\text{Number of target nucleus per unit area x Number of neutrons of energy E}}$$

$$s_g(E) = \frac{N_g}{n_T\ [\text{at/barn}] \cdot N_n(E)}$$



Needs:

- **sample of known mass and dimensions**
- **count the number of incident neutrons of energy E**
- **count the number of capture reactions**

… but there are a number of experimental complications

Neutron Beams

- **Need to span a huge energy range: 1meV – 100MeV**
- **Since neutrons cannot be accelerated, they have to be produced by nuclear reactions at certain energy and eventually decelerated by nuclear collisions (moderated)**
- **Energy determination:**
  - **kinematics of two-body reaction**
  - **mechanical selection of velocities ("chopper")**
  - **Time Of Flight measurement**

$$E_n = \frac{1}{2} m_n \frac{L^2}{t^2}$$

- **Sources:**
  - **Radioactive**
  - **Nuclear detonations**
  - **Reactor**
  - **Light-ion accelerator**
  - **Electron LINACS**
  - **Spallation**

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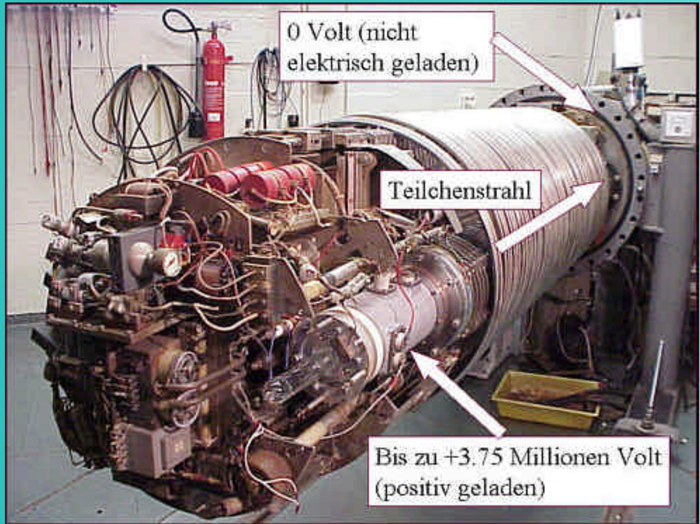
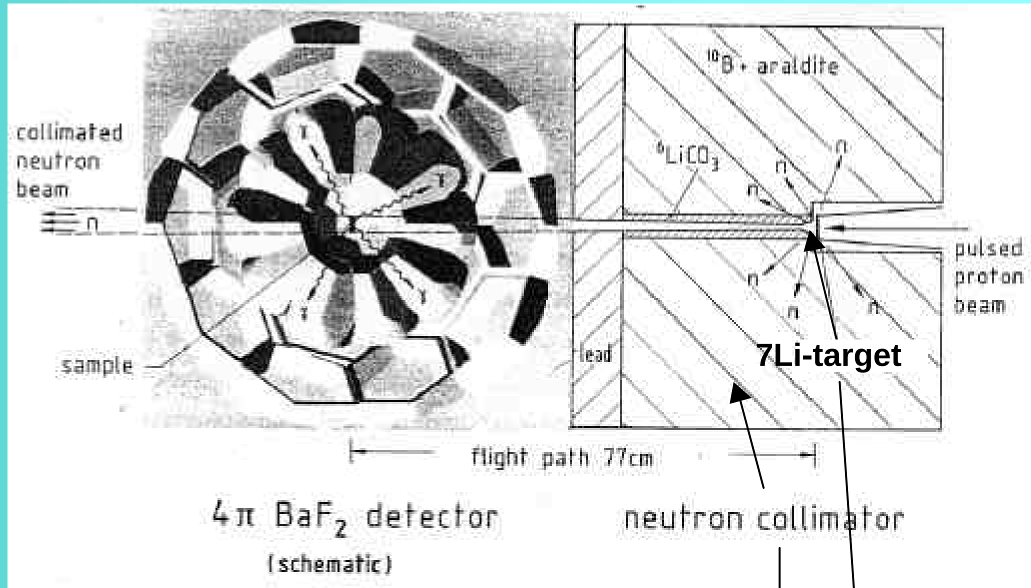
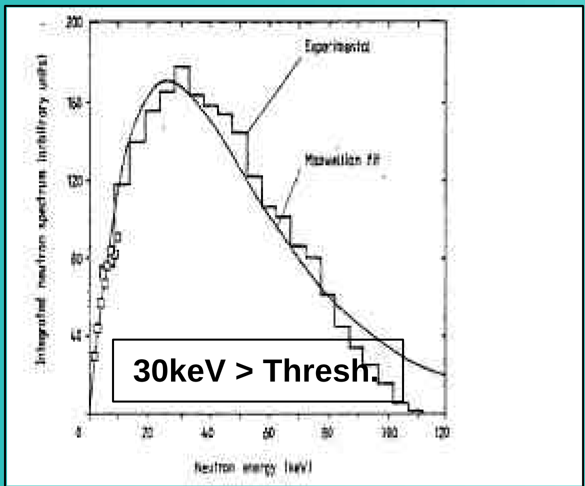






- **$^7Li(p,n)^7Be$, Q= -1.644MeV**
- **$E_p \sim 2$MeV → $E_n \sim 5$-200keV**
- **Rate: 250kHz, Δt = 0.7ns**

Geel Electron LINear Accelerator

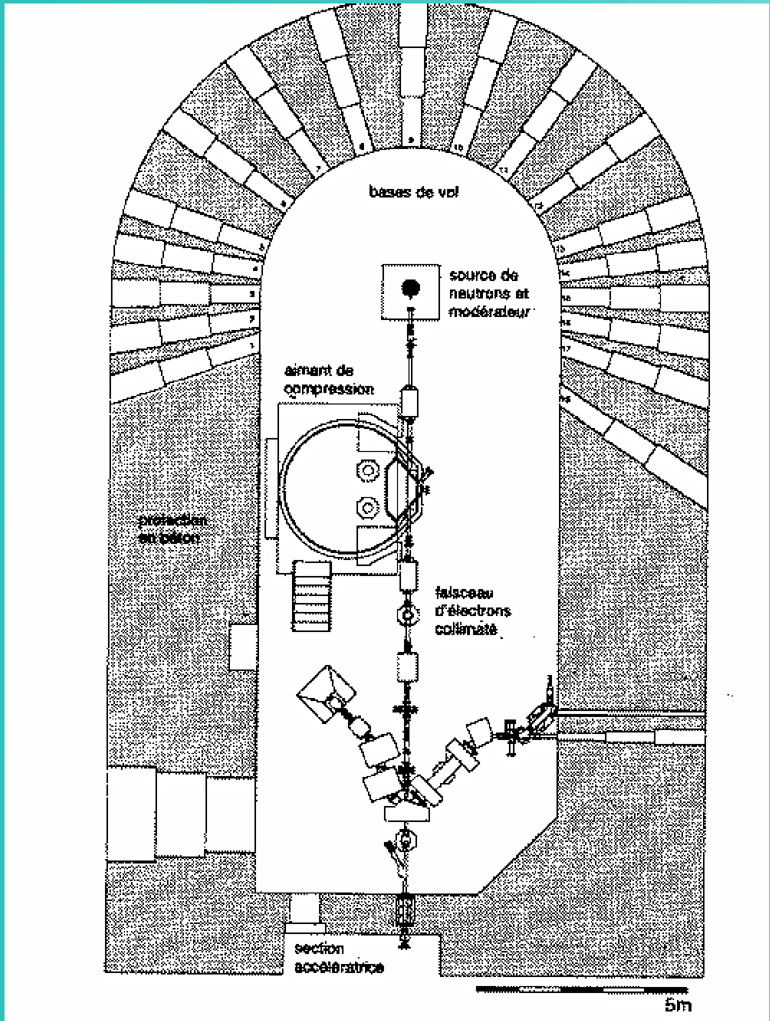


Figure 2.3 : Schéma général de la partie accélératrice et de la salle des cibles

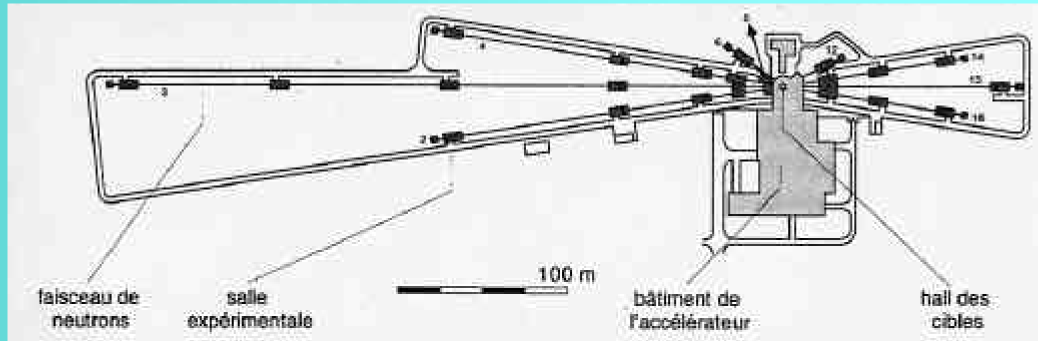


Figure 2.8 : Localisation des bases de vol et des salles expérimentales

- (γ,n), (γ,f) on U (bremsstrahlung)
- E_e ~ 100MeV, I_e ~ 10-100μA
- Rate: 100-800Hz, Δt ~ 0.6-15ns

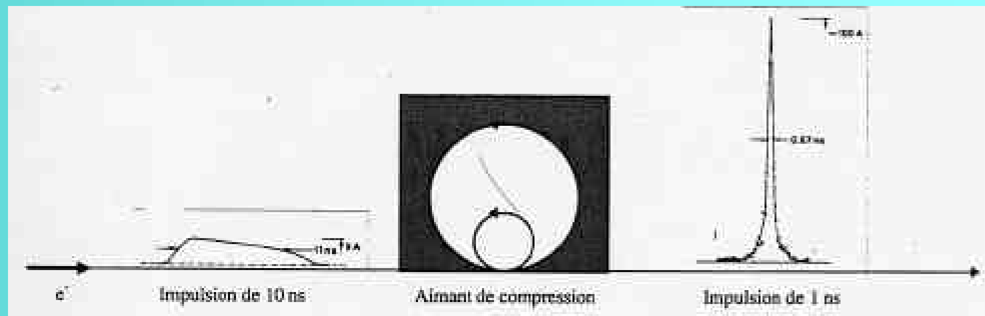
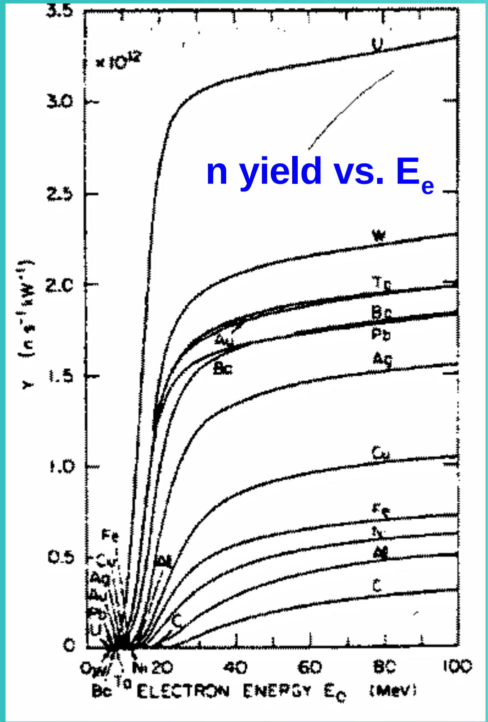
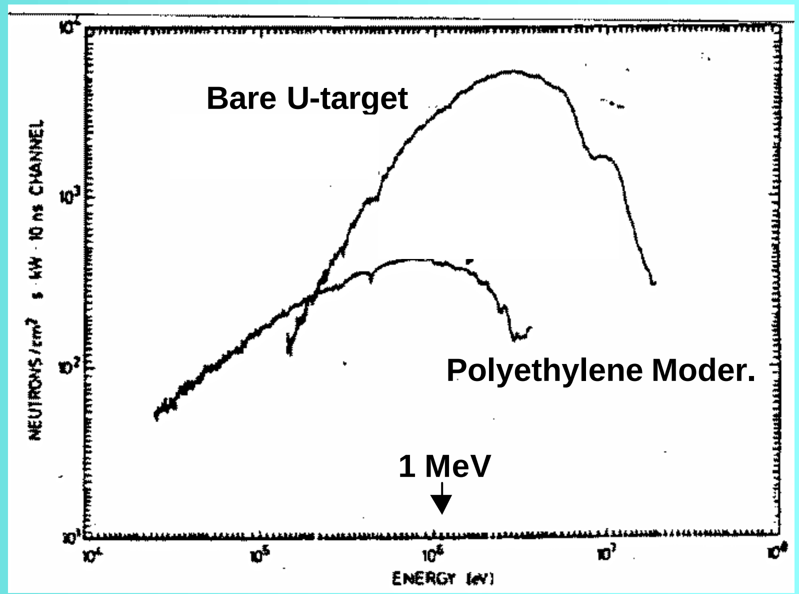


Figure 2.4 : Principe de compression magnétique des impulsions

… GELINA





NEUTRON SPECTRA

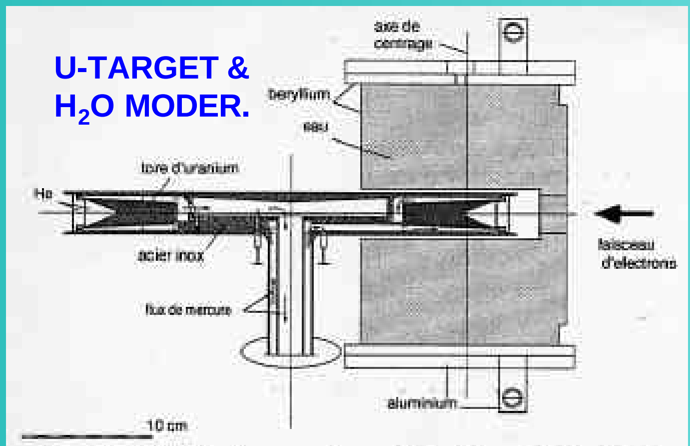
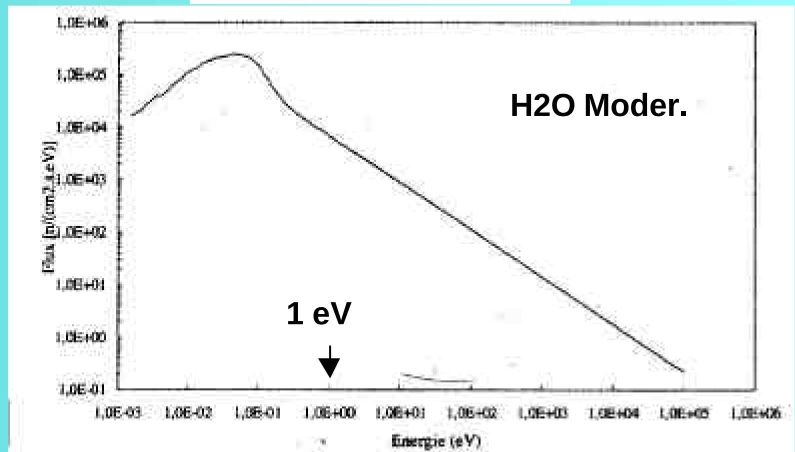
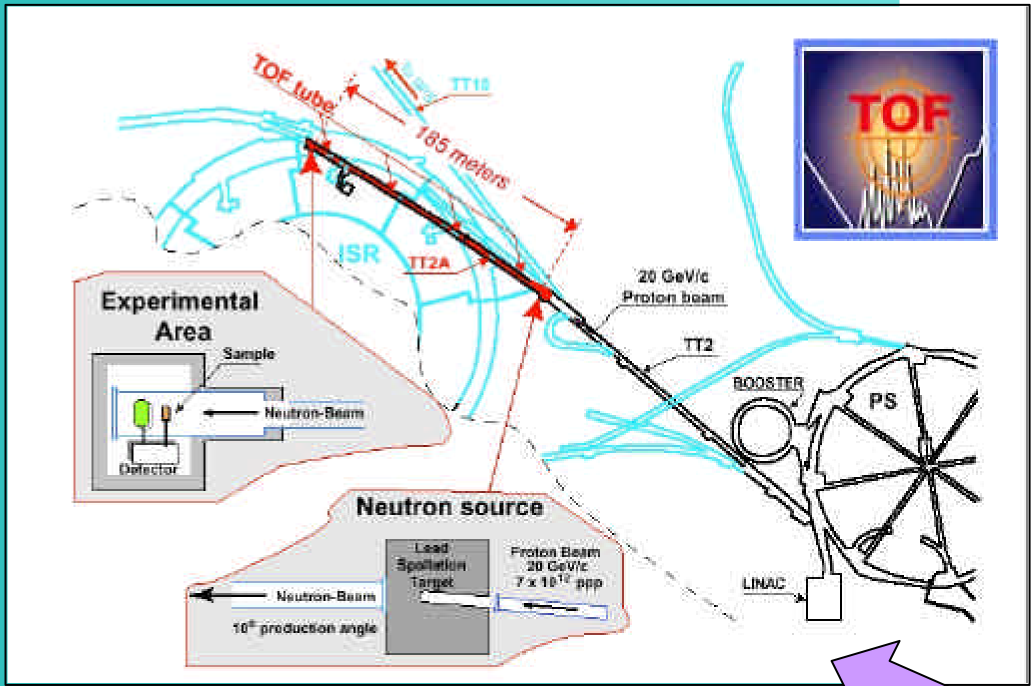
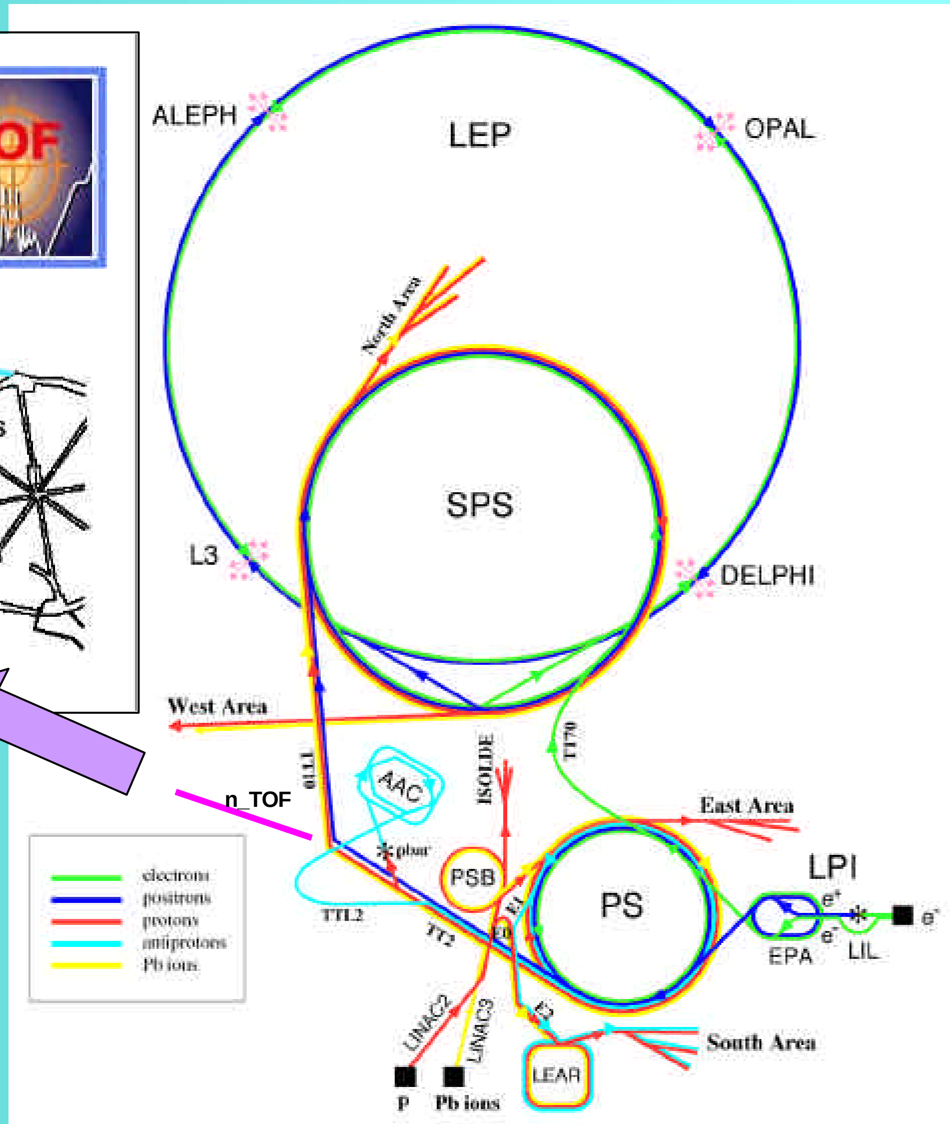


Figure 2.5 : Cible d'uranium et modérateur de l'accélérateur linéaire de Geel

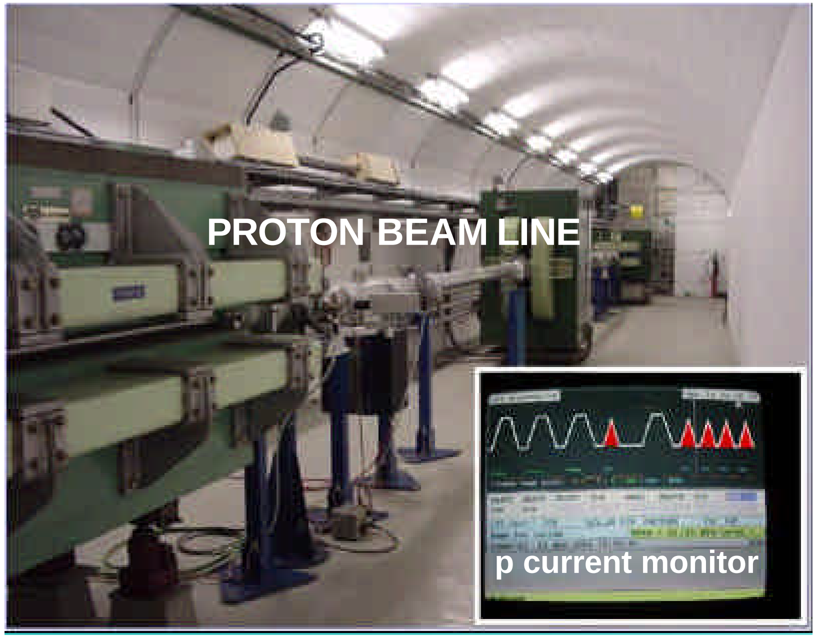


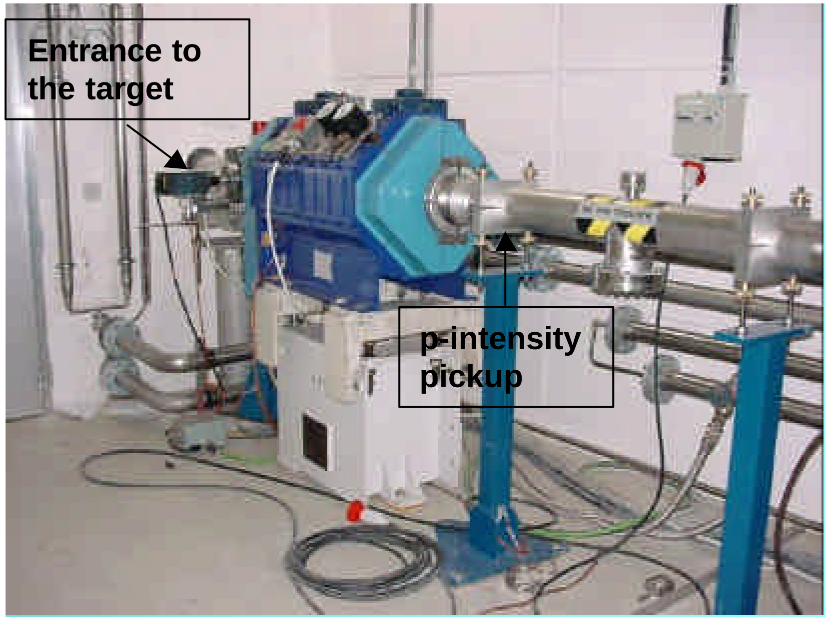
CERN neutron Time Of Flight

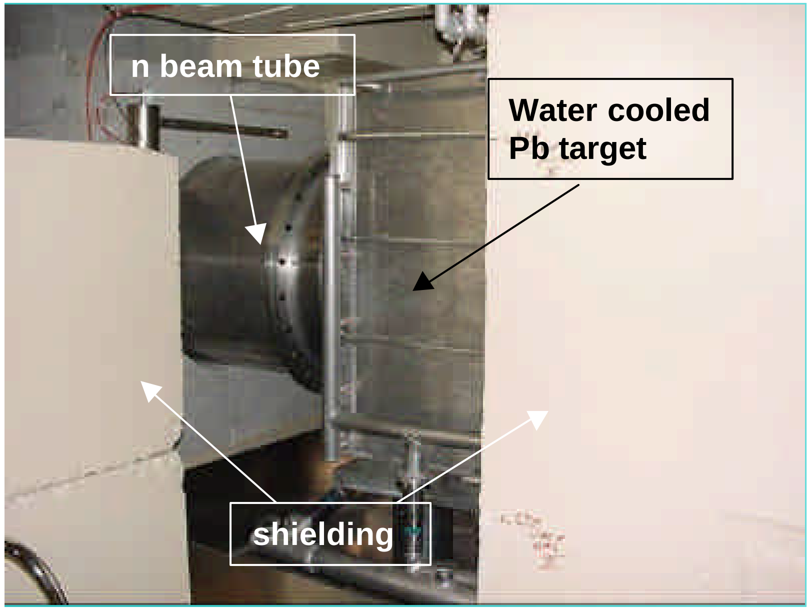


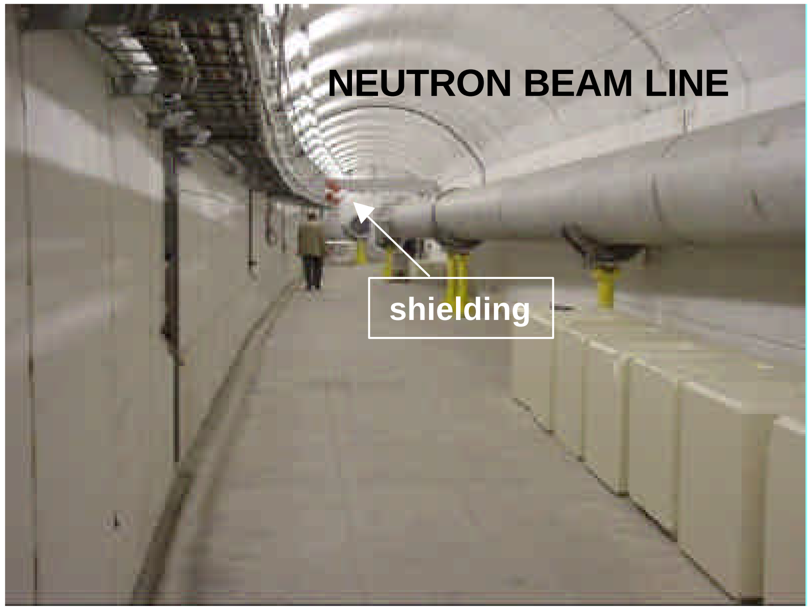


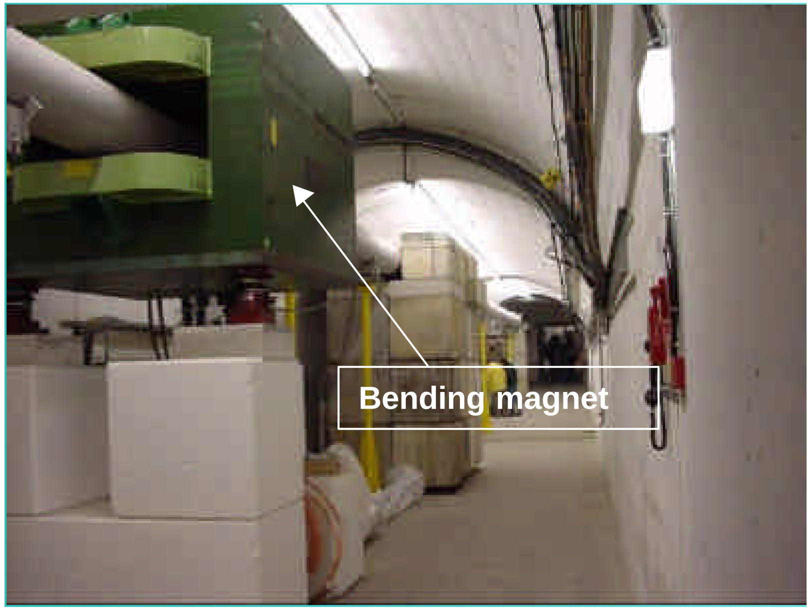
- **p-spallation on Pb target**
- **E_p = 20GeV, I_p = 7x10^{12} ppp**
- **Rate: 2.4s^{-1} , Dt = 14ns**


PROTON BEAM LINE
p current monitor

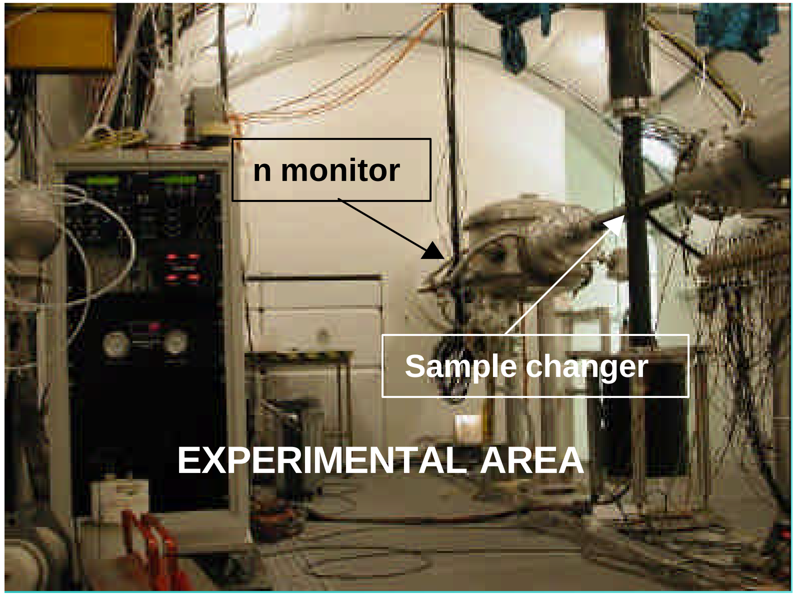

Entrance to
the target
p-intensity
pickup

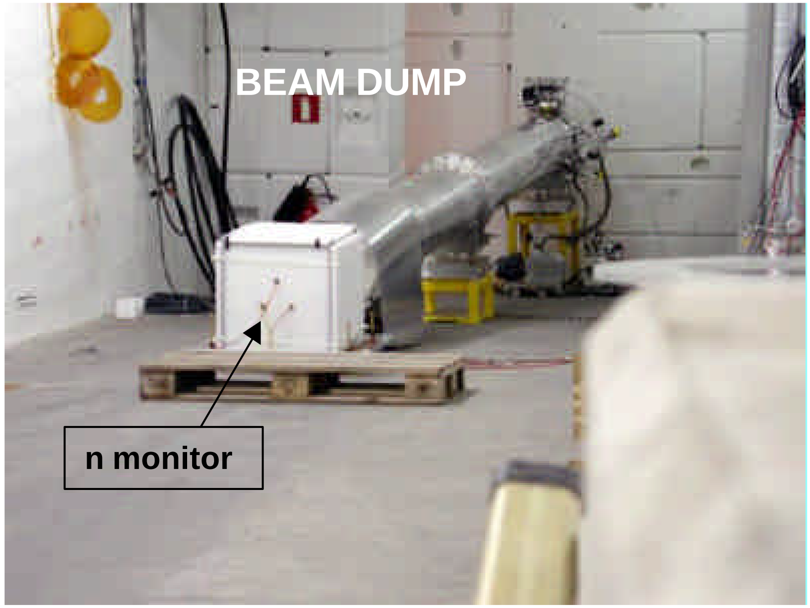

n beam tube
Water cooled
Pb target
shielding


NEUTRON BEAM LINE
shielding


Bending magnet


2nd collimator
shielding

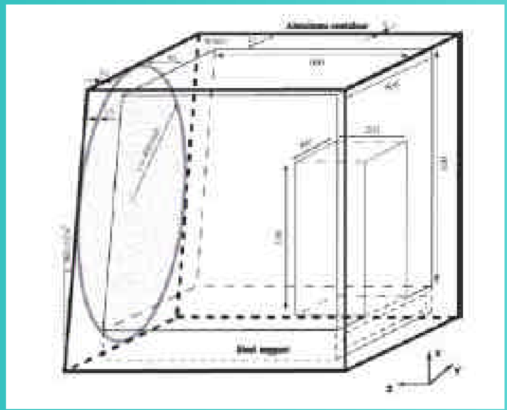

n monitor
Sample changer
EXPERIMENTAL AREA


BEAM DUMP
n monitor

- **The characteristics of the spallation-moderation process and the collimators in use determine the neutron beam parameters: intensity-energy distribution, energy resolution and spatial distribution.**

TARGET:

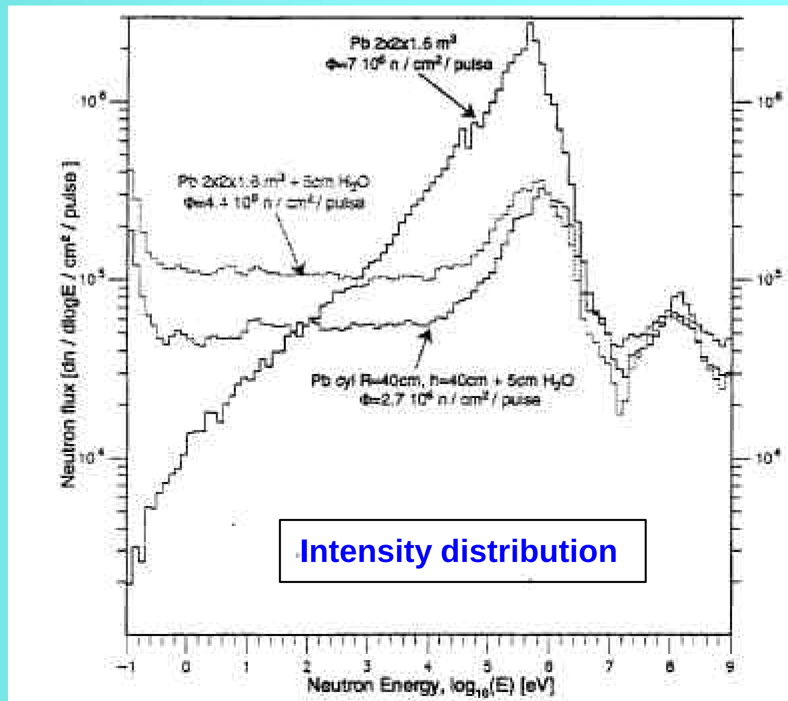
80x80x60 cm^3 Pb + 5cm H_2O



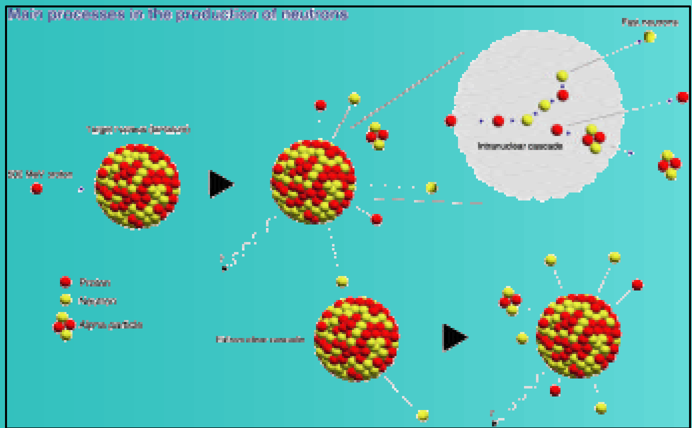
MODERATION:

$$\xi = \langle \ln(E_i/E_f) \rangle \approx 1 + \frac{(A-1)^2}{2A} \ln \frac{A-1}{A+1}$$

($\xi(H)=1$, $\xi(Pb)=0.01$)

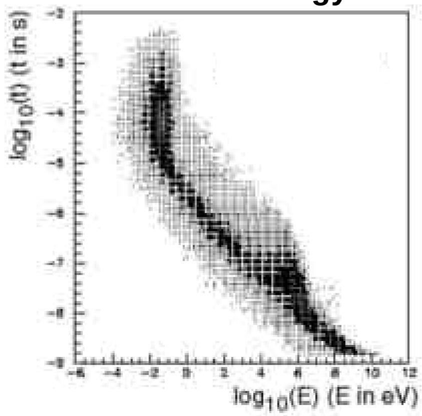


SPALLATION PROCESS: ~600 n/p

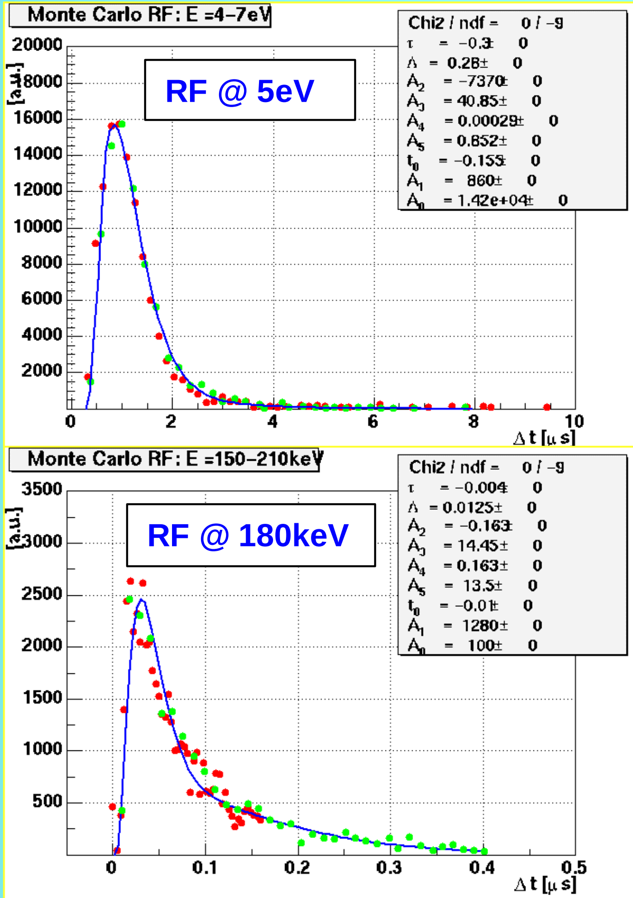


The statistical nature of the moderation process produces variations on the time that a neutron of a given energy exits the target assembly

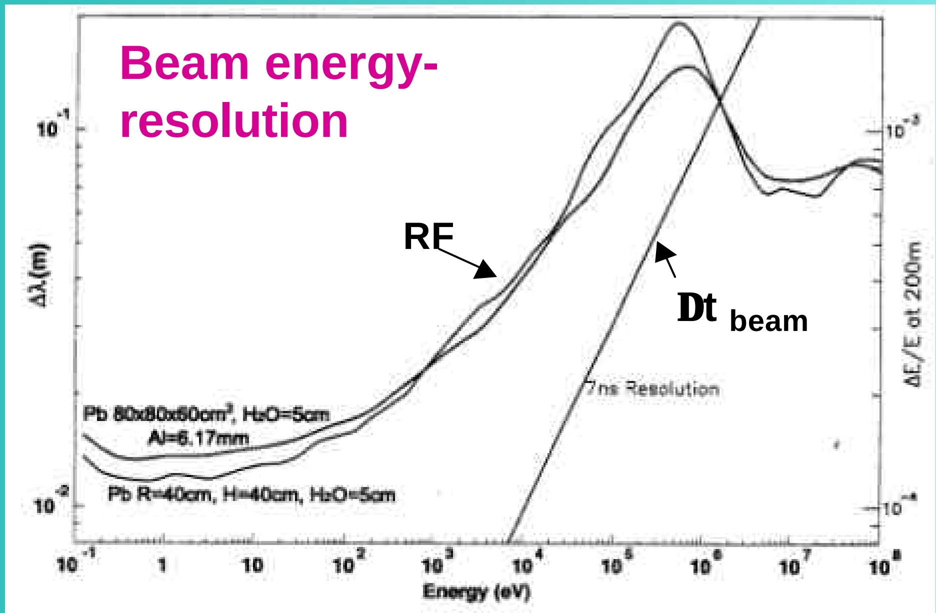
time vs. energy



Resolution Function

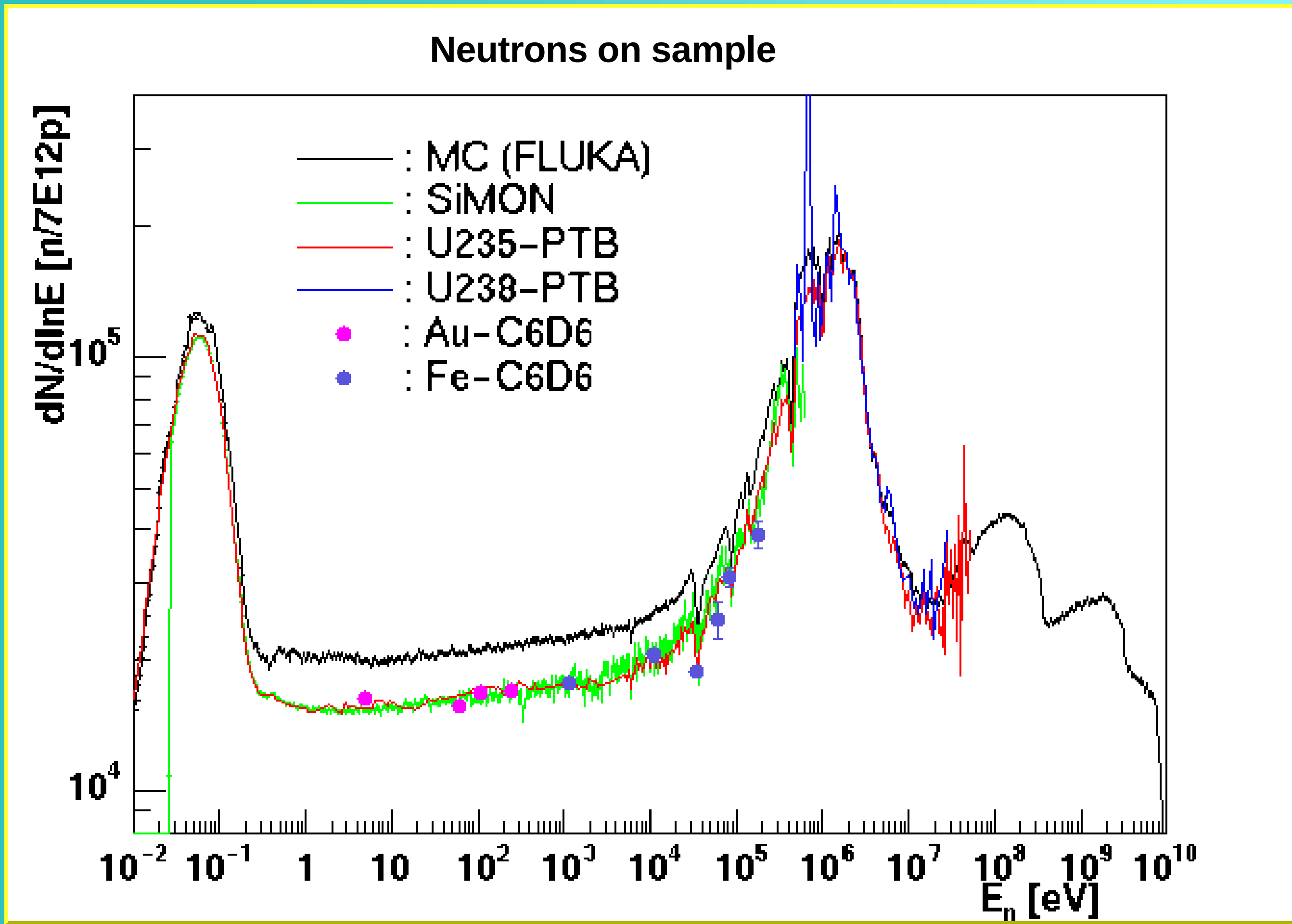


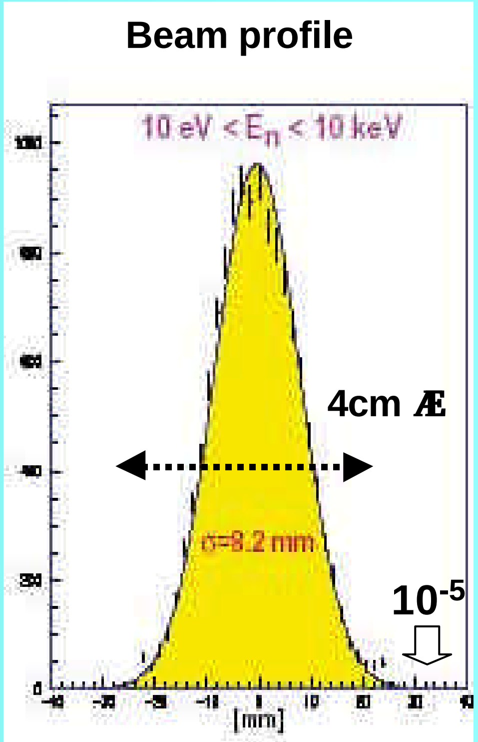
$$\frac{\Delta E_n^2}{E_n^2} = 2\frac{\Delta L^2}{L^2} + 2\frac{\Delta t^2}{t^2}$$



… also important: time spread of beam

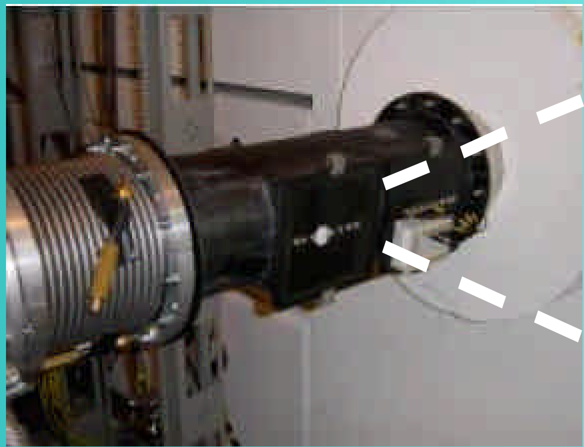
The collimation system determines the final number of neutrons arriving to the sample an its spatial distribution

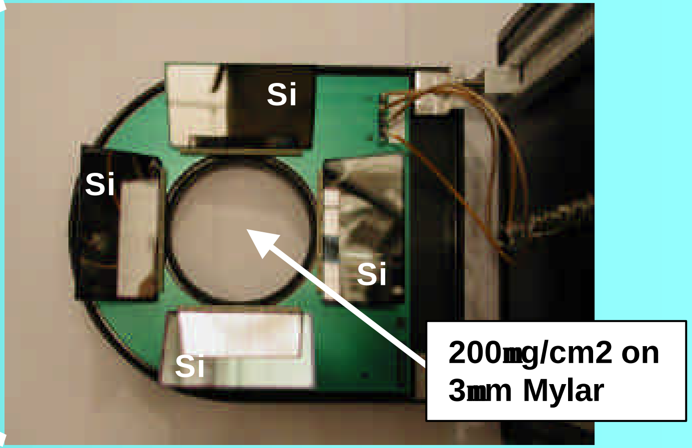


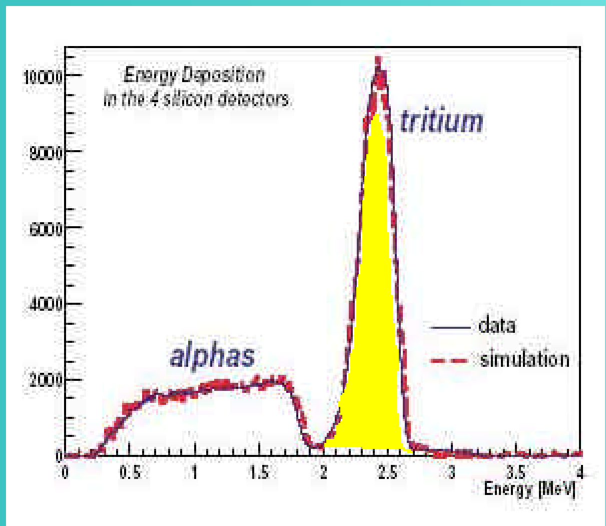


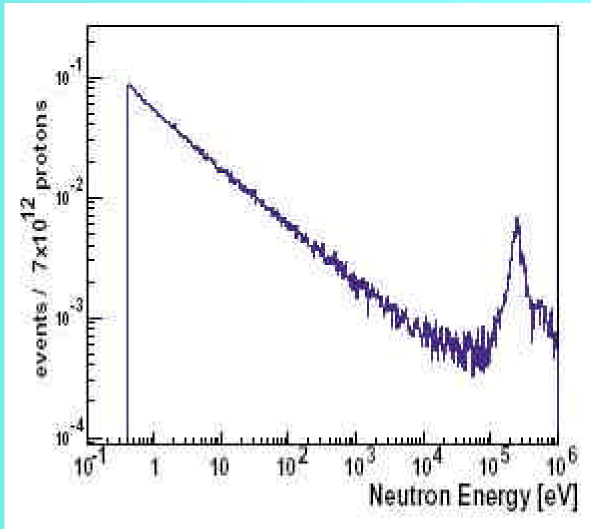
Neutron Intensity Monitoring:

- Reaction: $n + {}^{6}Li \rightarrow t + \alpha$
- Si detectors







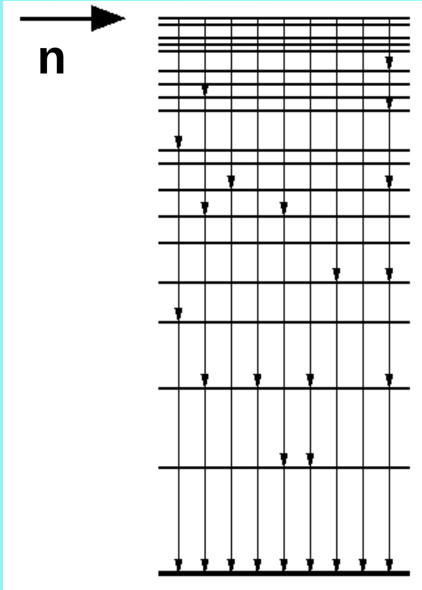


Techniques for radiative capture detection:

- **Detection of the capture nucleus**
  - **Activation measurements**

> • Irradiation: A(n,g)A+1
>
> • A+1 radioactive with suitable $T_{1/2}$
>
> • Measurement of characteristic g-ray of known I_g with Ge detector

- **Detection of g-ray cascade**
  - **Total Absorption Spectrometers**
  - **Total Energy Detectors**
    - **Moxon-Rae Detectors**
    - **Pulse Height Weighting Technique**



Define:

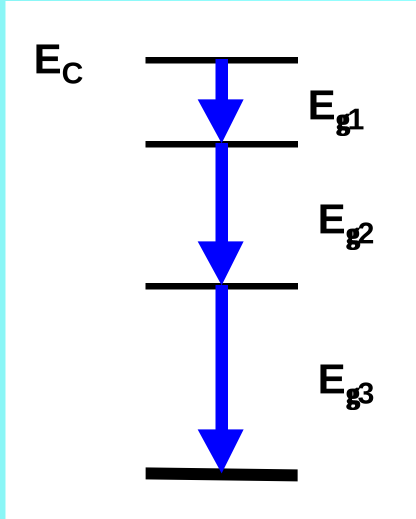
$\varepsilon_{\gamma i}$: total efficiency for γ-ray of energy $E_{\gamma i}$

$\varepsilon^{p}_{\gamma i}$: peak efficiency for γ-ray of energy $E_{\gamma i}$

Then:

total efficiency for cascade: $\varepsilon_C = 1 - \prod_{i=1}^{m_\gamma} (1 - \varepsilon_{\gamma i})$

peak efficiency for cascade: $\varepsilon^{p}_C = \prod_{i=1}^{m_\gamma} \varepsilon^{p}_{\gamma i}$



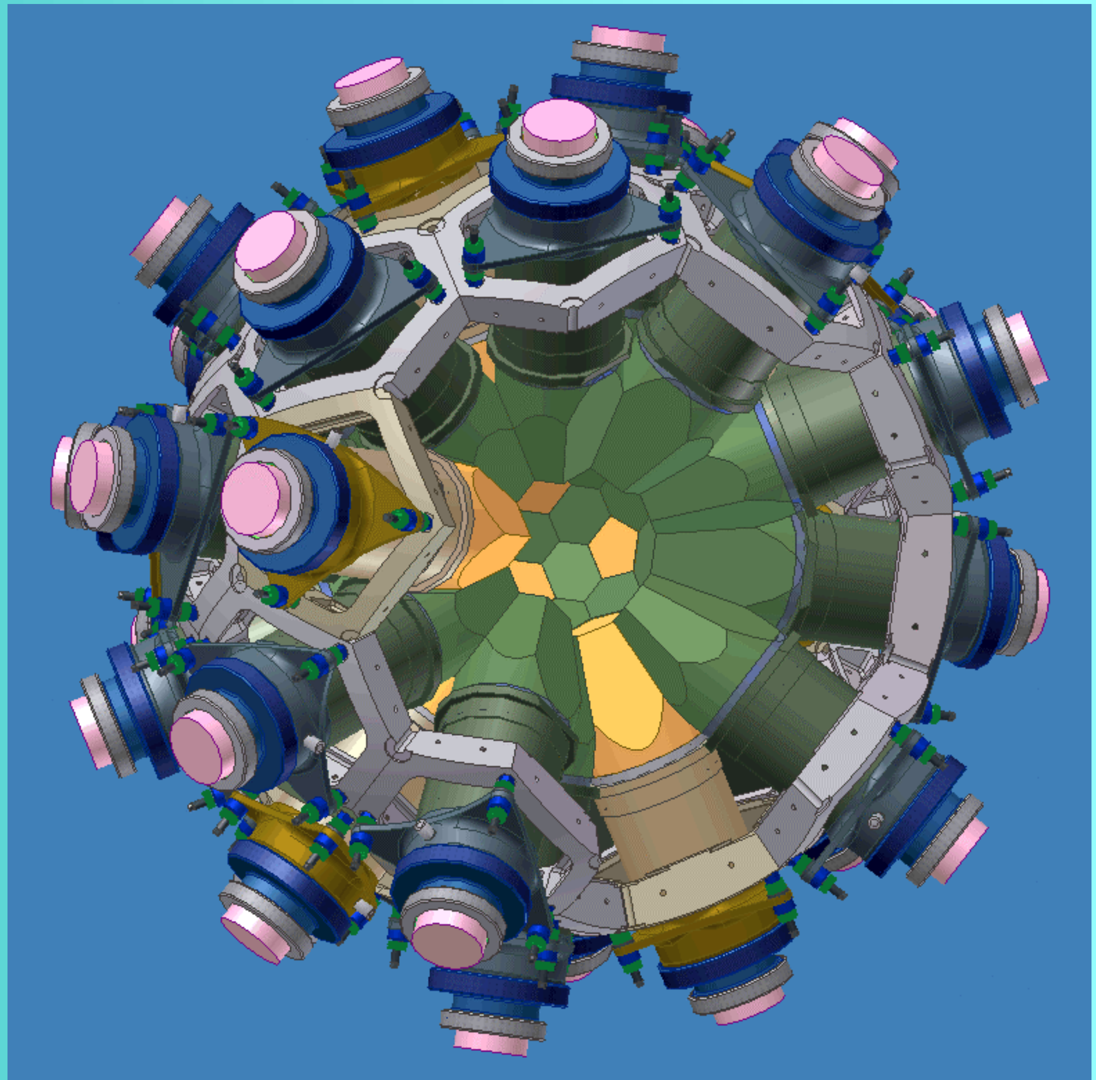
If $\varepsilon^{p}_{\gamma i} = 1, \forall i \Rightarrow \varepsilon^{p}_C = \varepsilon_C = 1$ ⇨ **T**otal **A**bsorption **S**pectrometer

If $\varepsilon_{\gamma i} \ll 1$ & $\varepsilon_{\gamma i} = kE_{\gamma i}, \forall i \Rightarrow \varepsilon_C \cong \sum_{i=1}^{m_\gamma} \varepsilon_{\gamma i} = k \sum_{i=1}^{m_\gamma} E_{\gamma i} = k E_C$

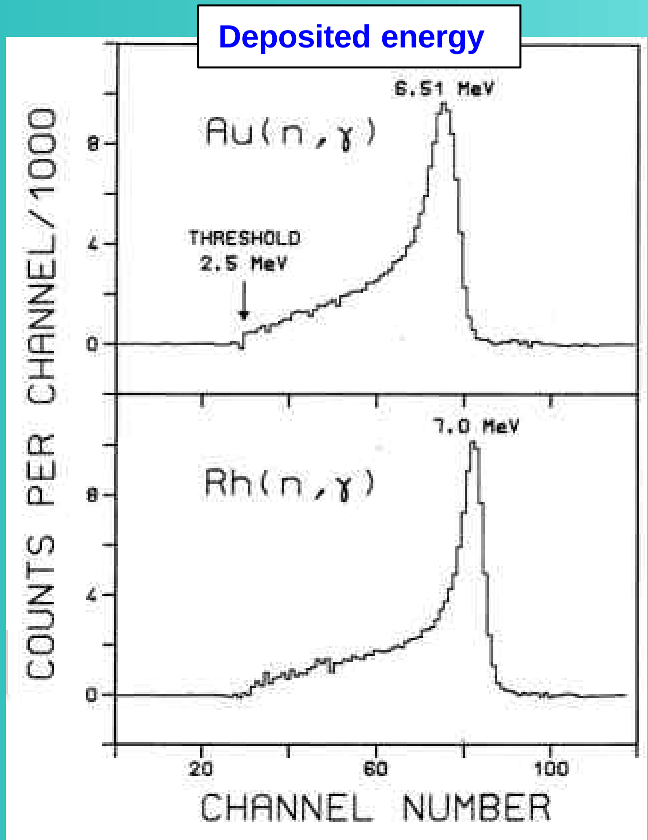
⇨ **T**otal **E**nergy **D**etector

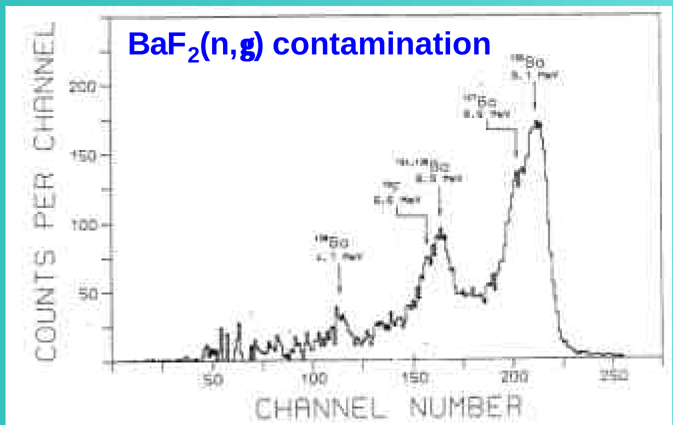
n_TOF Total Absorption Calorimeter

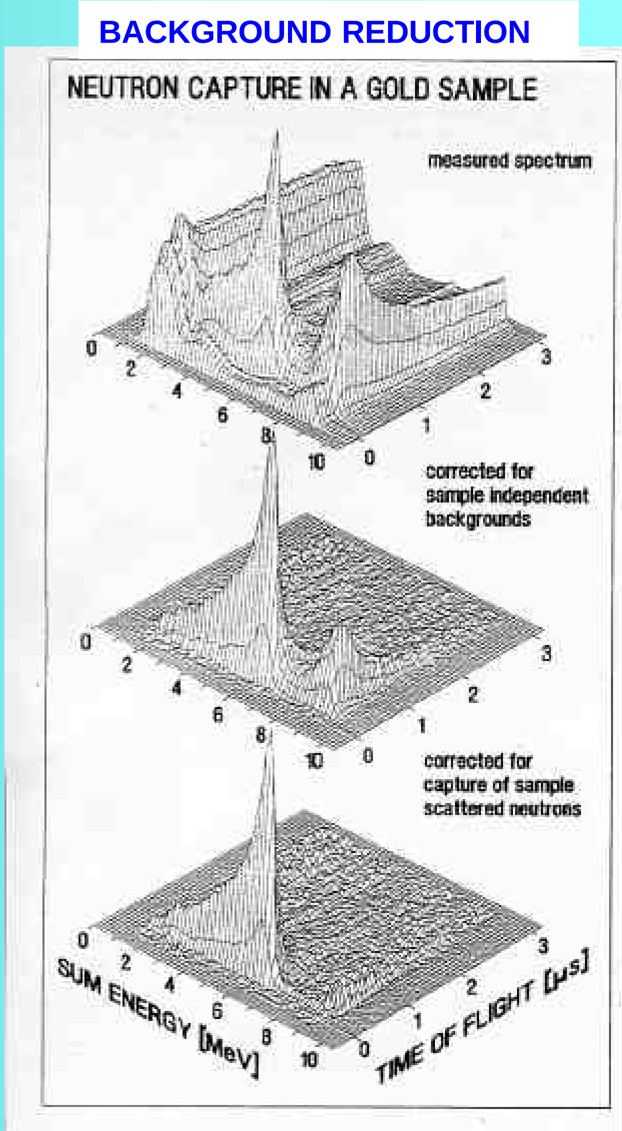
- **40 BaF_2 crystals**
- **DW/4p = 95%**
- **DE @ 6%**



(from Karlsruhe 4π BaF_2 detector)

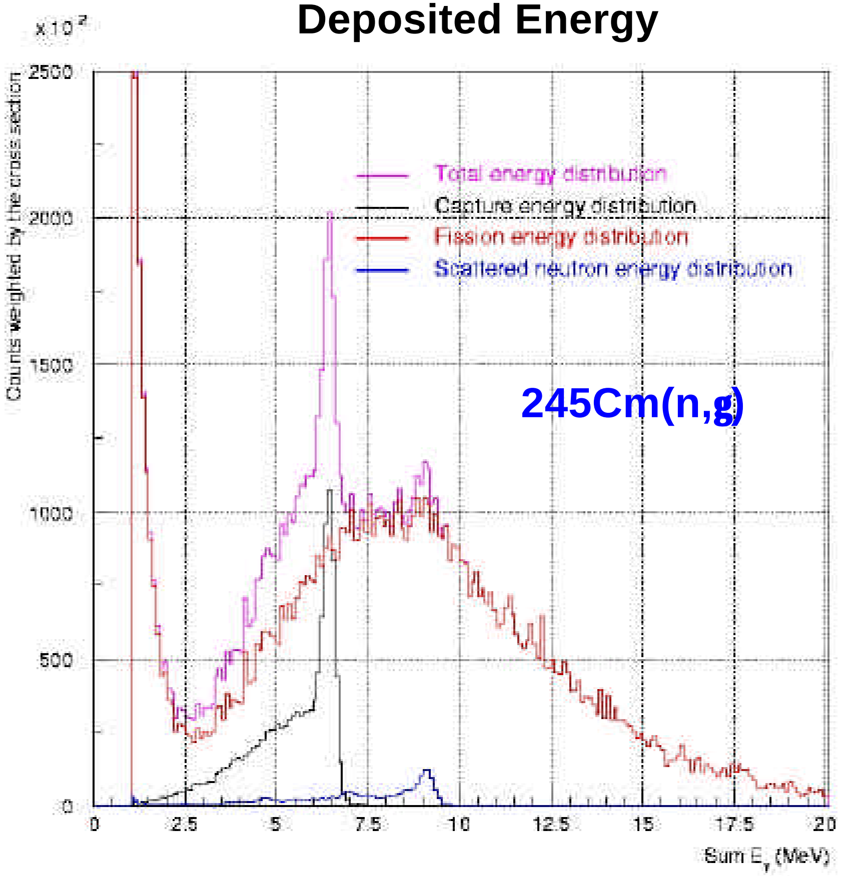




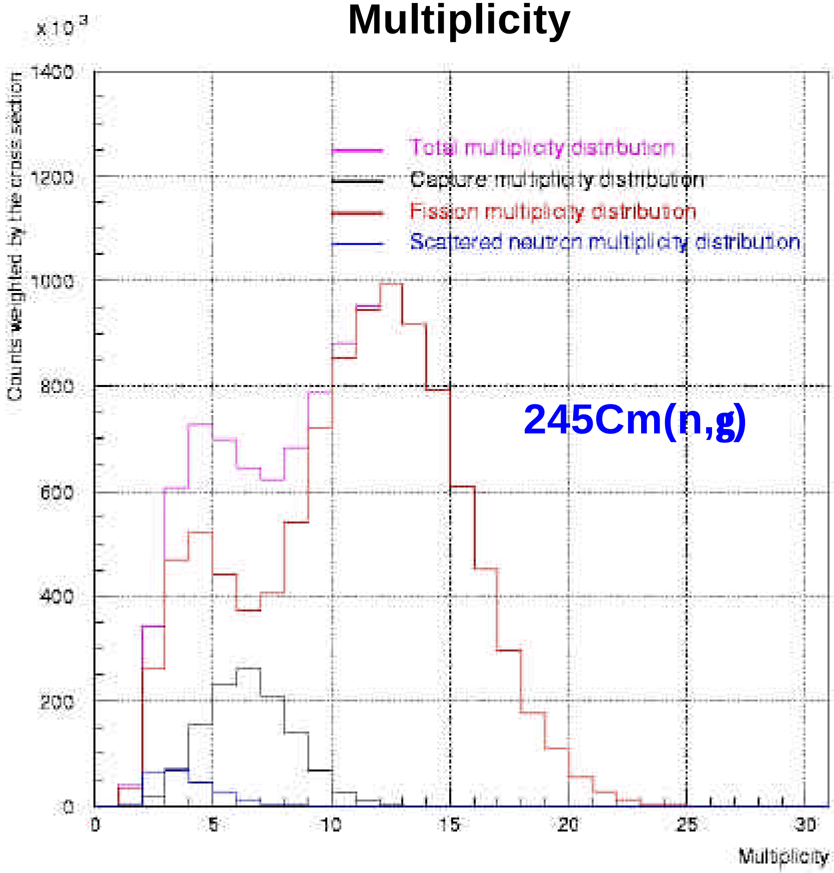


• **The good energy resolution and detector granularity makes feasble the measurement of fisioning nuclei**

Deposited Energy



Multiplicity



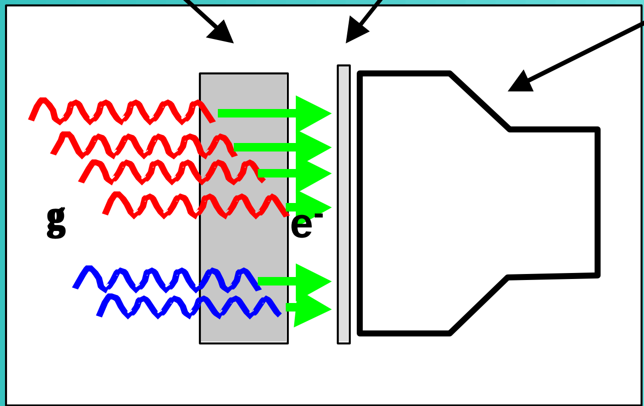
(Monte Carlo simulation D. Cano - CIEMAT)

Total Energy Detectors

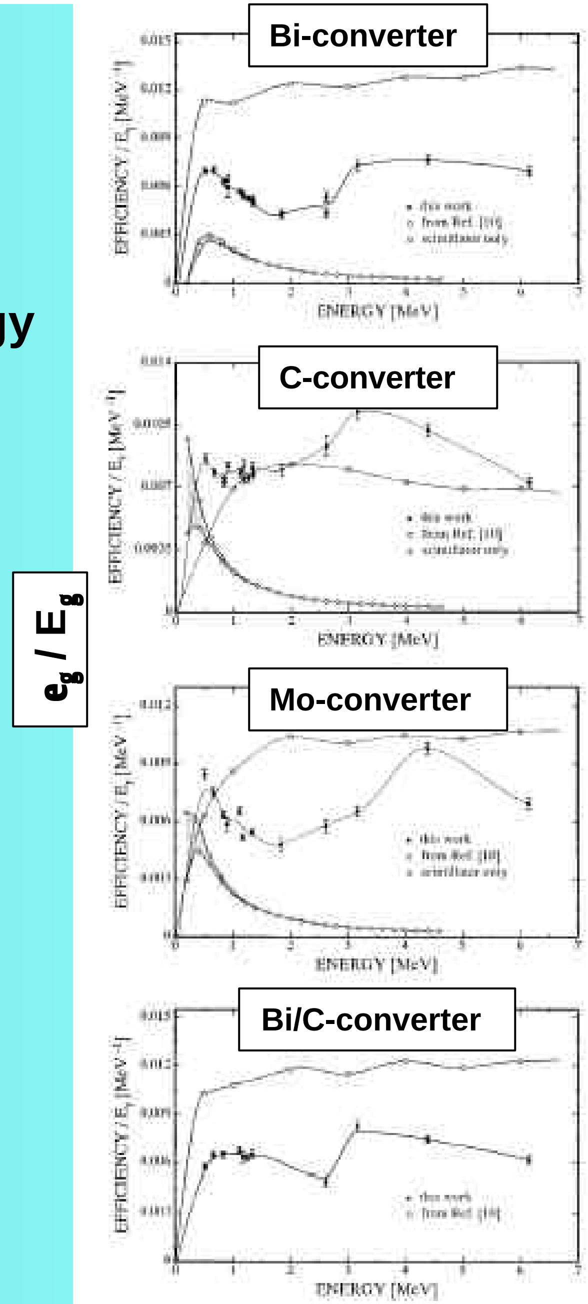
- **Moxon-Rae type detectors:**

The proportionality between efficiency and γ-ray energy is obtained by construction:

(γ,e⁻) converter + thin scintillator + photomultiplier



Maximum depth of escaping electrons increases with E_γ …



E_γ

But … proportionality only approximate (need corrections)

⇨ **Not much in use nowadays**

Total Energy Detectors

- **Pulse Height Weighting Technique:**

The proportionality between efficiency and γ-ray energy is obtained by software manipulation of the detector response (Maier-Leibniz):

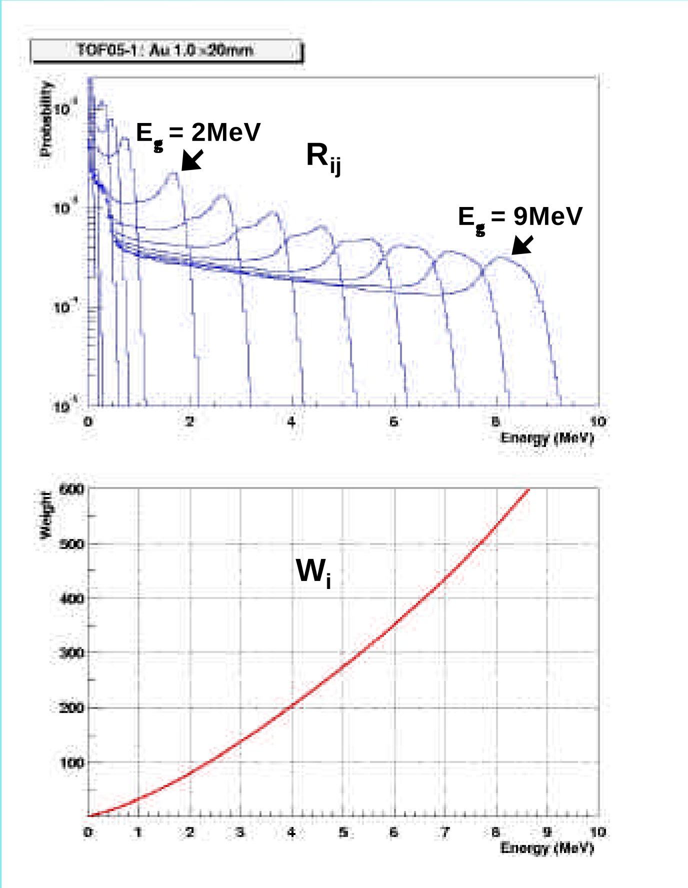
If R_{ij} represents the response distribution for a γ-ray of energy $E_{\gamma j}$:

$$\sum_{i=1}^{i_{max}} R_{ij} = \varepsilon_{\gamma j}$$

it is possible to find a set of weighting factors W_i (dependent on energy deposited i) which fulfil the proportionality condition (setting k=1):

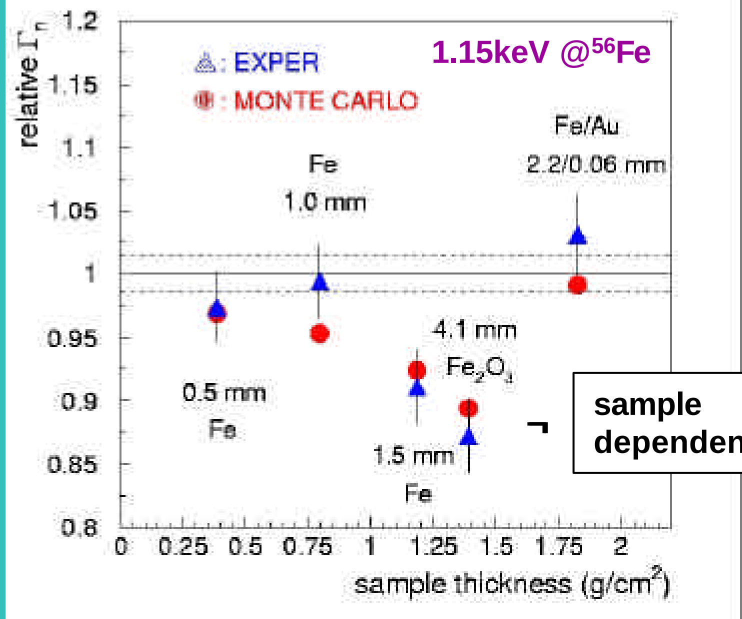
$$\sum_{i=1}^{i_{max}} W_i R_{ij} = E_{\gamma j}$$

for every $E_{\gamma j}$

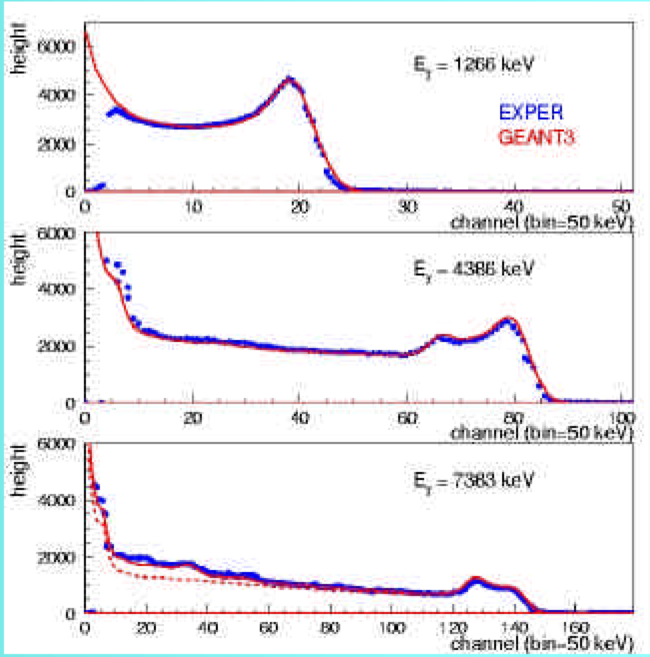


How accurate can be the weighting function?

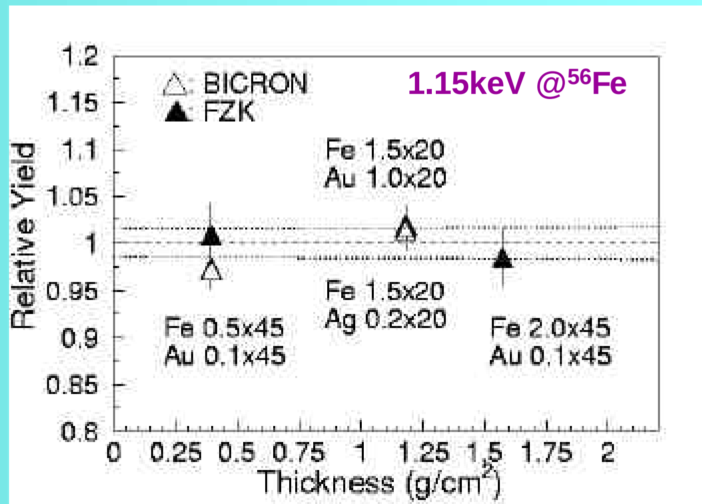
Fix "experimental" WF



→ **sample dependent**

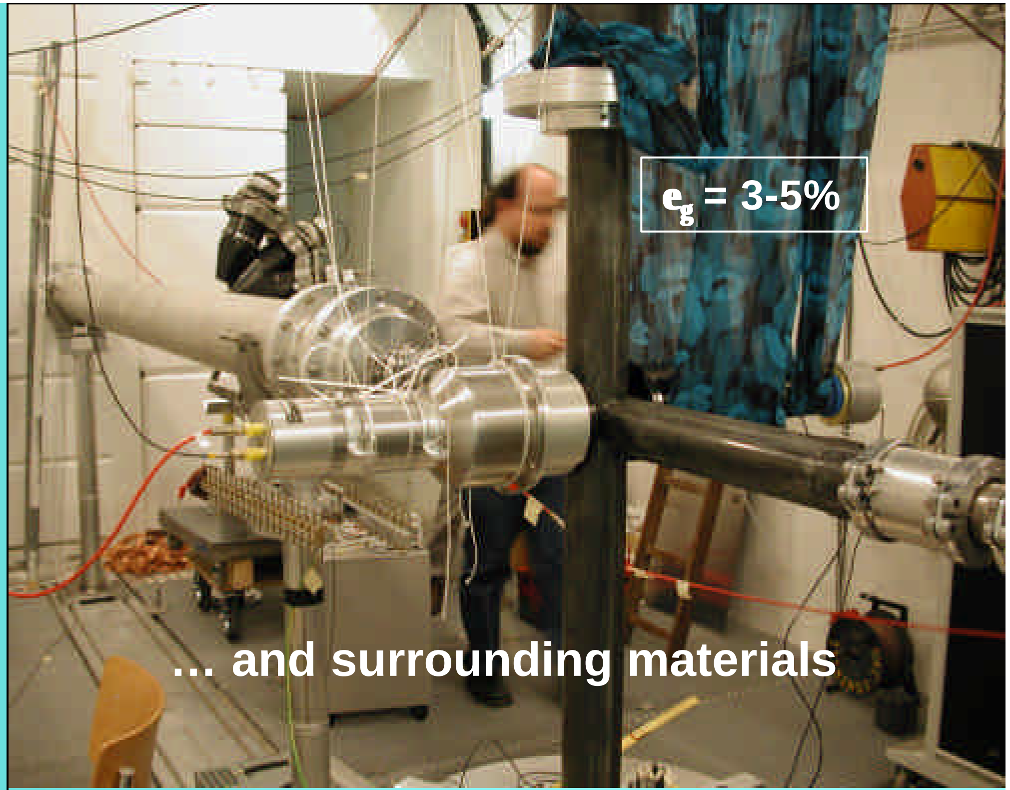


⇨ **Using GEANT Monte Carlo generated responses with full description of setup (including sample): $\sigma_{sg} \le 2\%$**



Detectors: C_6D_6 liquid scintillators

⇨ **Advantage: low neutron sensitivity**



Also detector dead material is important …

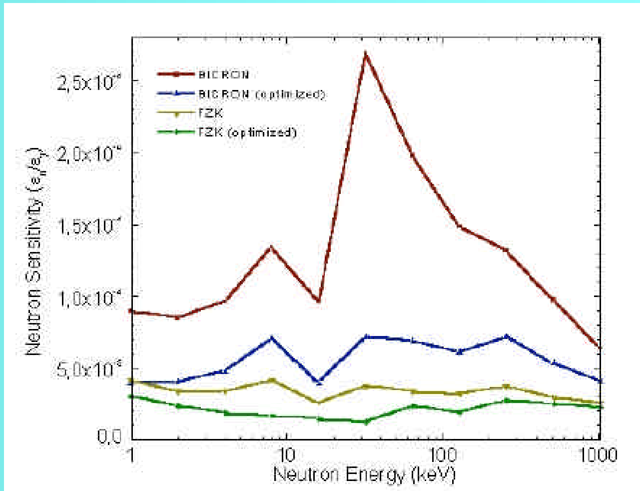
Optimized BICRON

⇩

HOME MADE:

- C-fibre cell
- No cell window
- No PMT housing

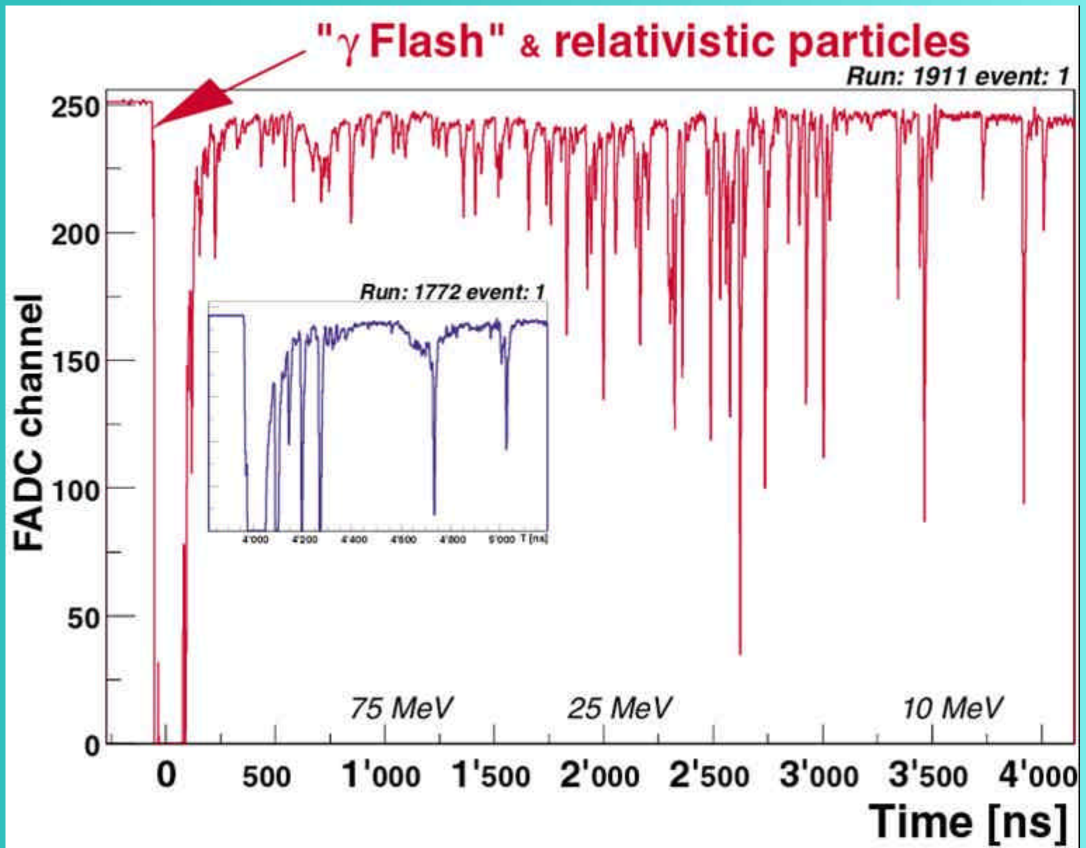
⇨

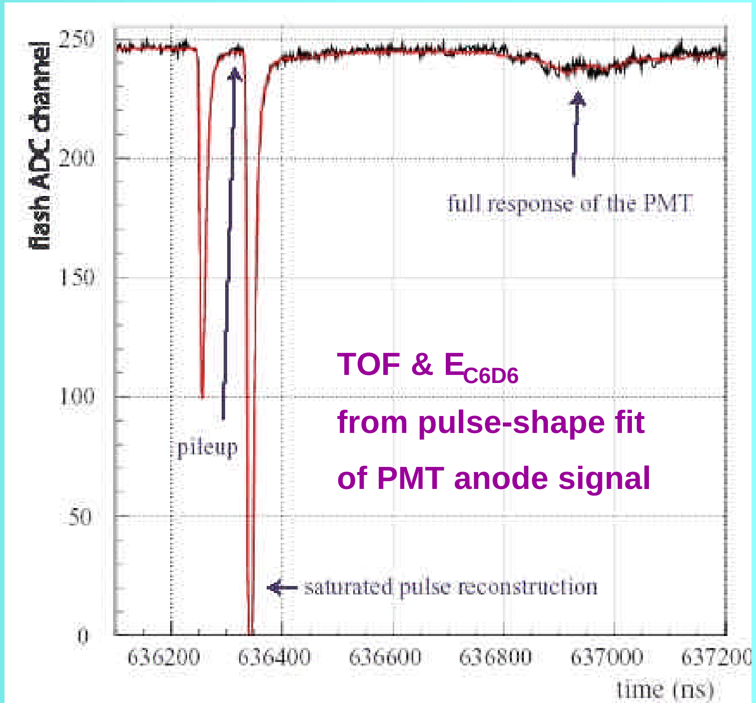


Acquiring the data: full train of detector pulses

- **Digitizer: 8bit-500MS/s FADC + 8MB memory**
- **On-line “zero” suppression**

- **Δt = 2ns**
- **E_n down to 0.6eV**
- **0.5 MB/pulse/detector**





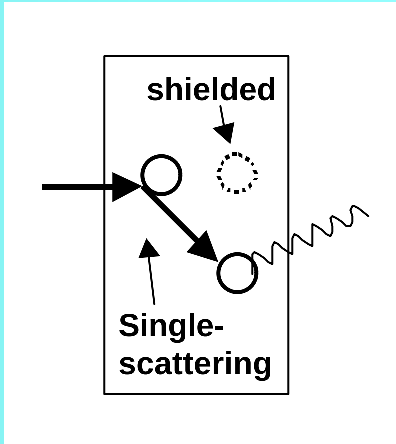
Analysing the data

$$s_g(E) = \frac{N_g}{n_T \cdot N_n(E)}$$

+ Yield: $Y_g(E) = N_g/N_n(E)$

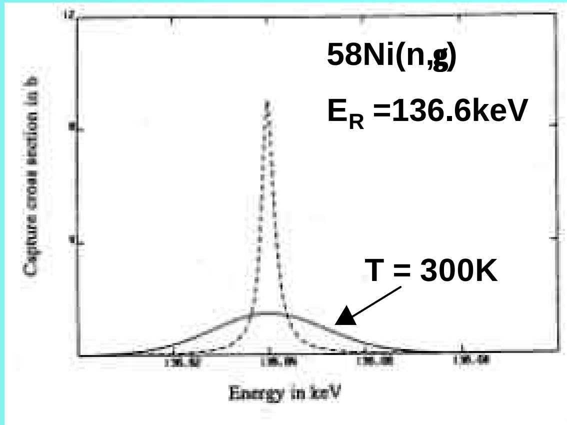
Sample effects:

- **Self-shielding**: $Y_g(E) = (1 - e^{-n_T \cdot s(E)}) \dfrac{s_g(E)}{s(E)}$



- **Multiple scattering correction:**
  elastic collision(-s) + capture

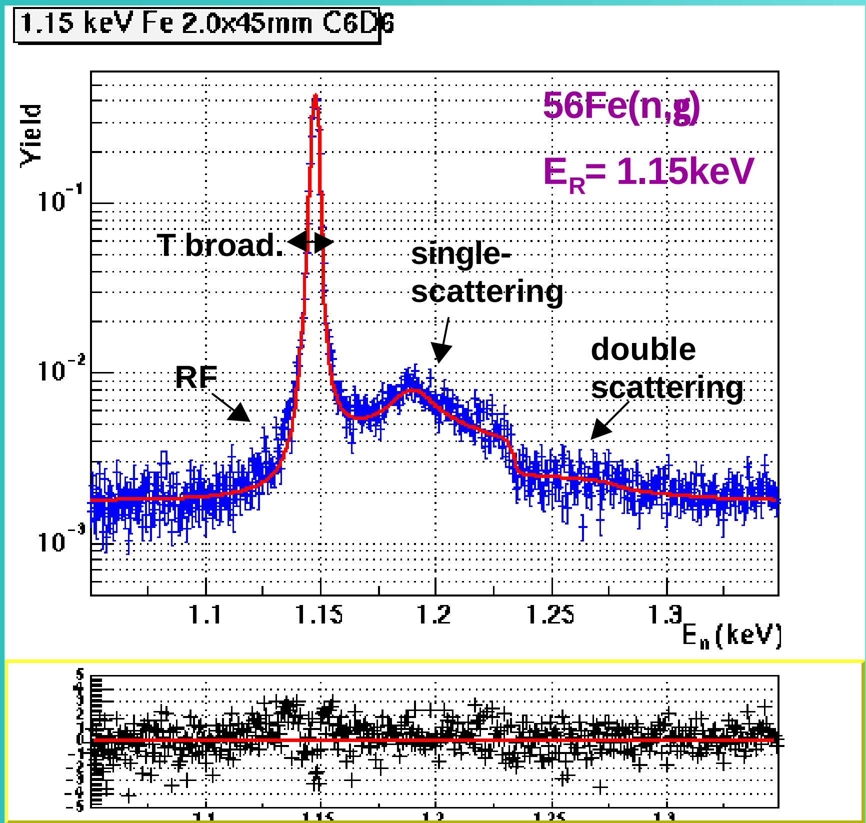
- **Thermal** (-Doppler) **broadening**

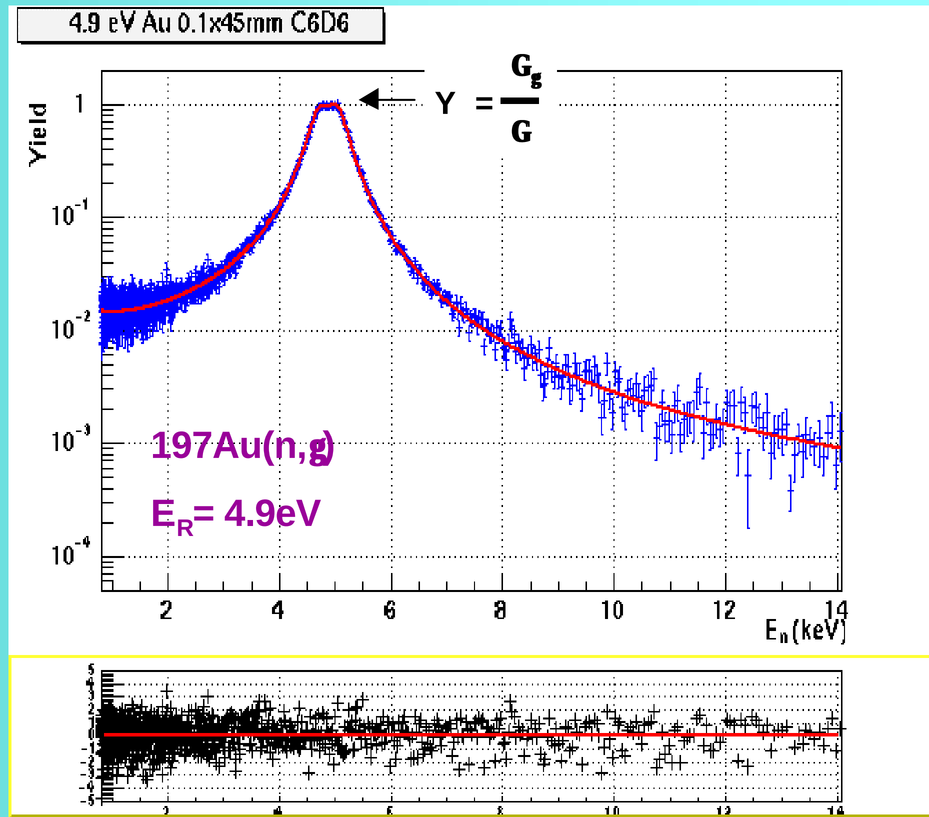


Beam effects:

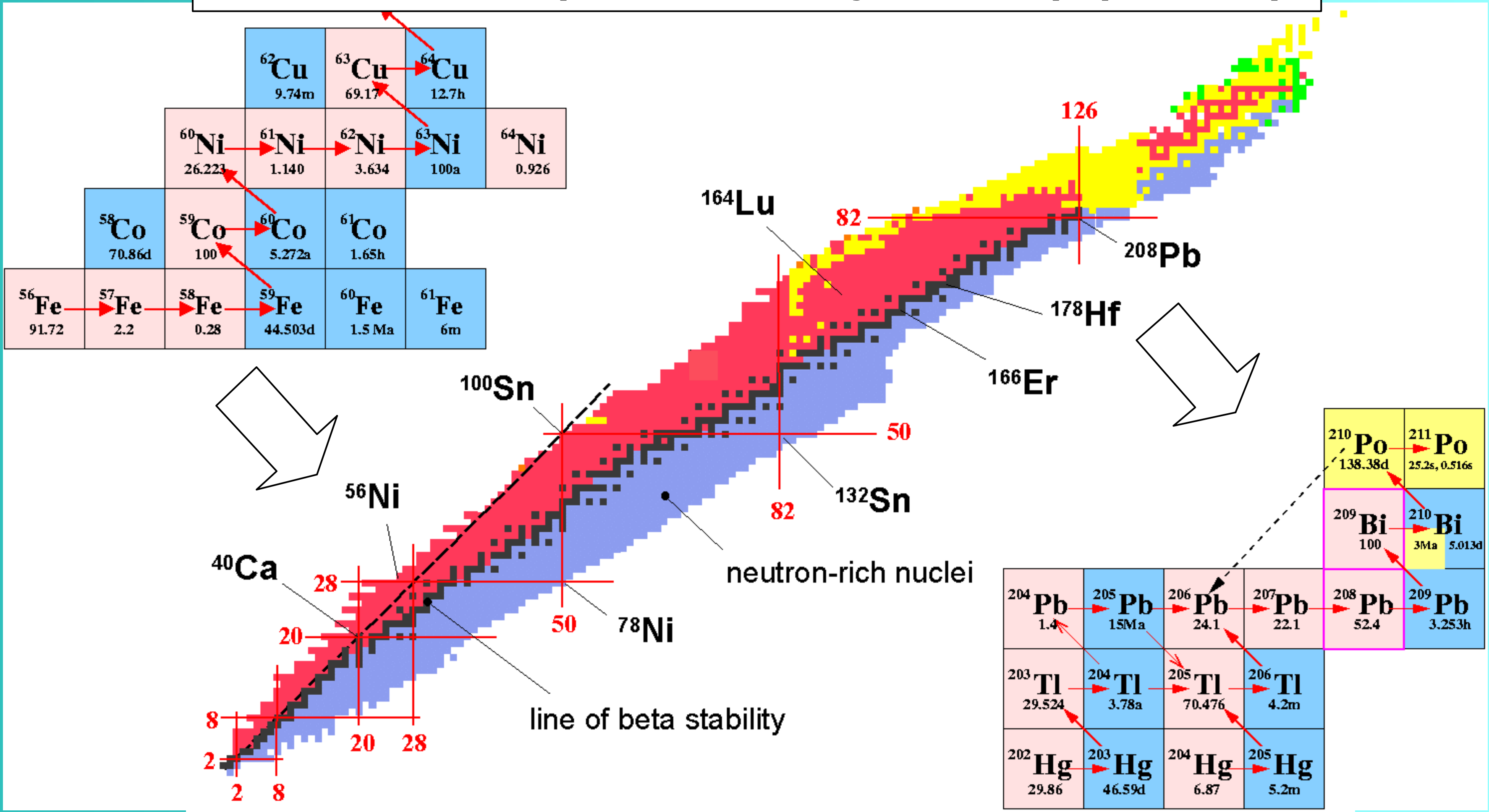
- **Resolution Function**

Use a R-Matrix code as SAMMY to fit the data and extract the parameters:
E_R, G_g , G_n, …



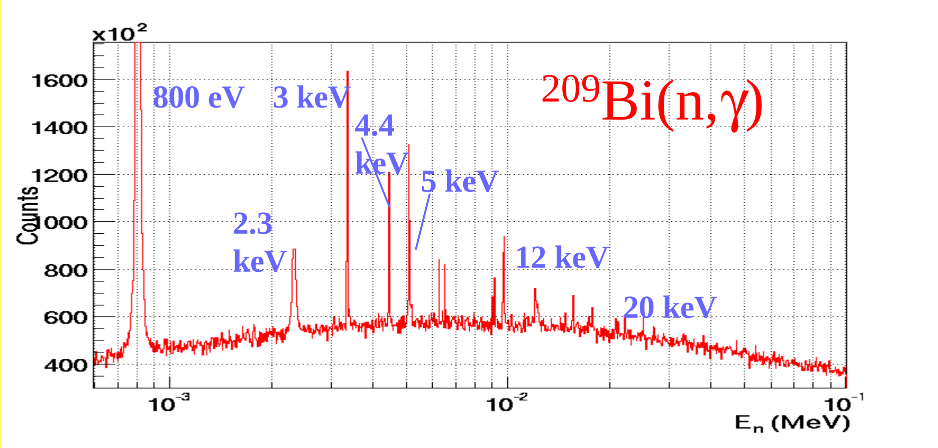


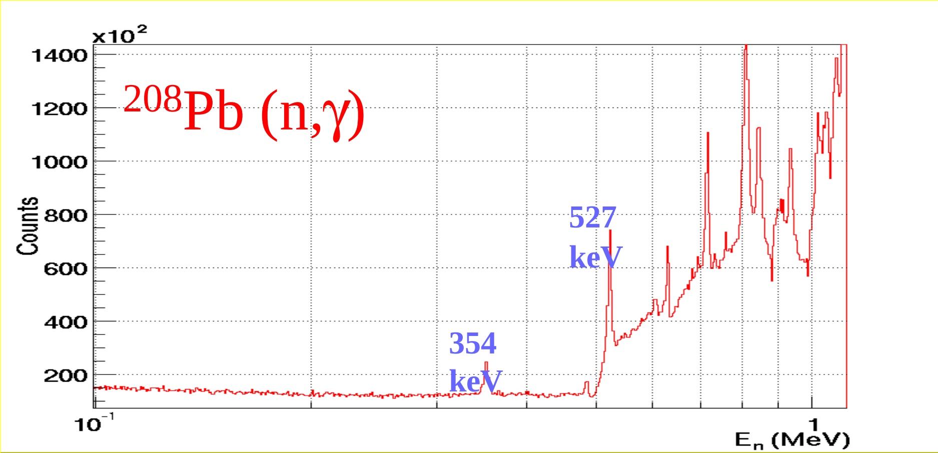
Slow neutron capture nucleosynthesis (s-process)

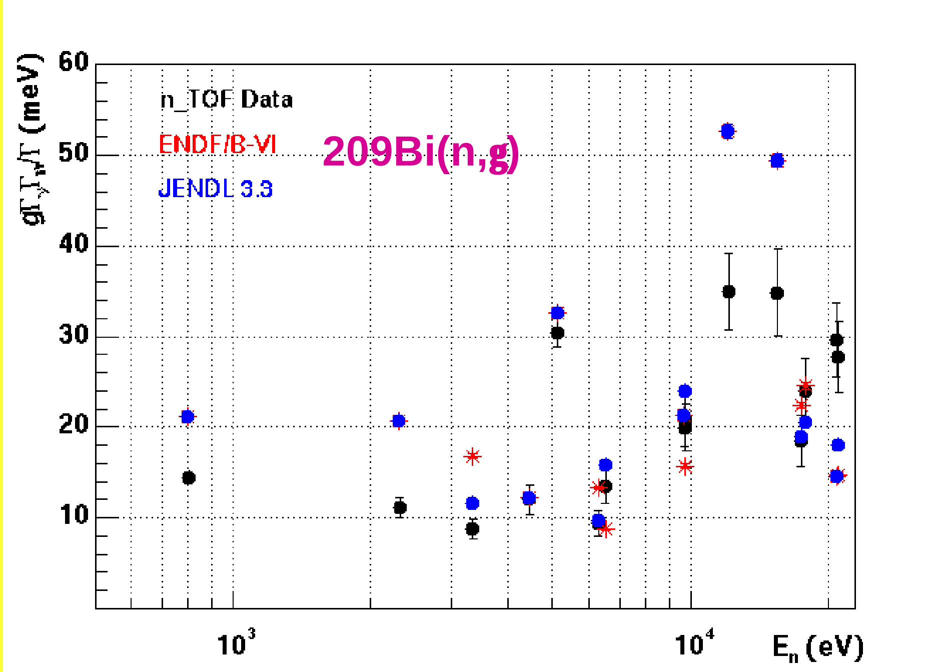


- **Pb-Bi isotopes: termination-point of s-process & ADS target-coolant**

Preliminary data
on Pb-Bi capture







$\ell = 0$
802 eV
2310 eV
5112 eV
12098 eV
15510 eV
17820 eV

$\ell = 1$
3348 eV
4456 eV
6286 eV
6524 eV
9708 eV
9760 eV
17440 eV
17820 eV
20860 eV
21050 eV

n_TOF s_g lower for ℓ=0

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