In-Beam Activation Analysis at MLZ

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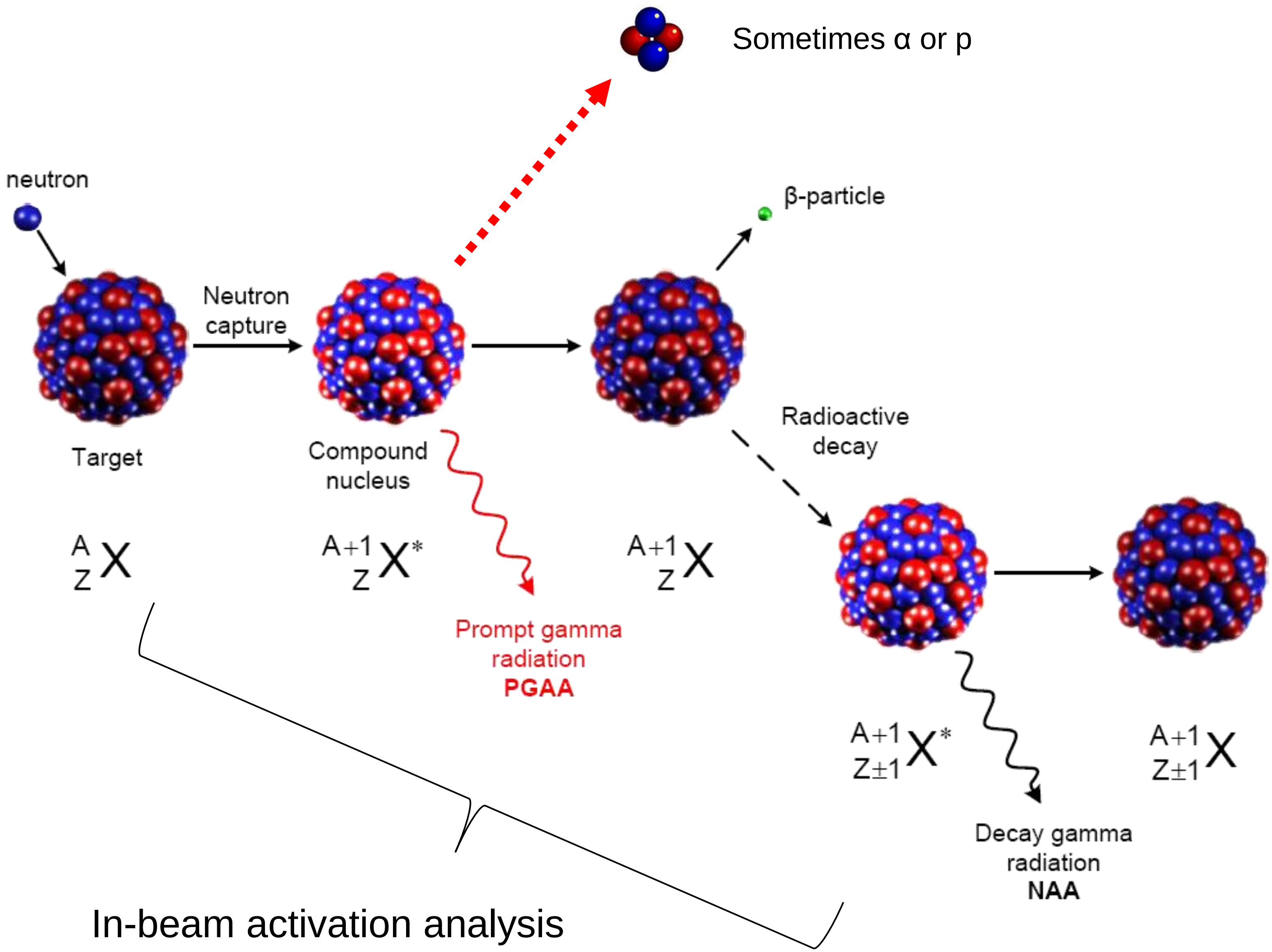
Important to know what is in our sample

-- Are you telling me that you can taste what kind of sugar I've used?
-- Of course not, but I can taste which kind you didn't use.

(Bella Martha, Germany 2002, dir: Sandra Nettelbeck, starring Martina Gedeck, ...)

PGAA and NAA: based on radiative neutron capture



Cross section differences

PGAA

$(\sigma_\gamma)_{max}$

NAA

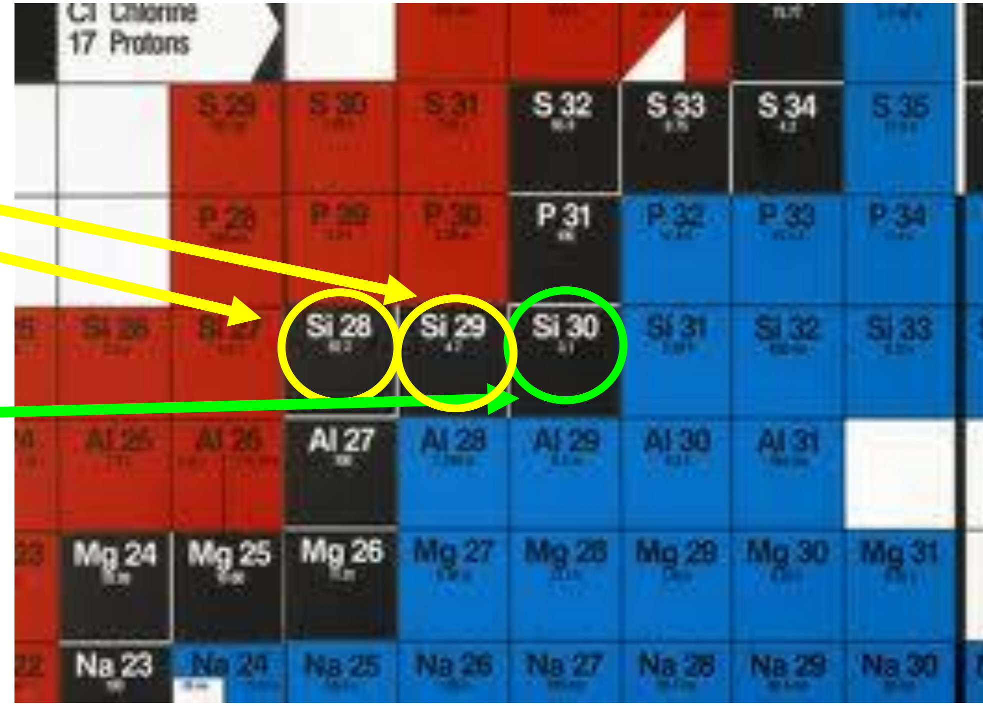
$(\sigma_\gamma)_{last}$

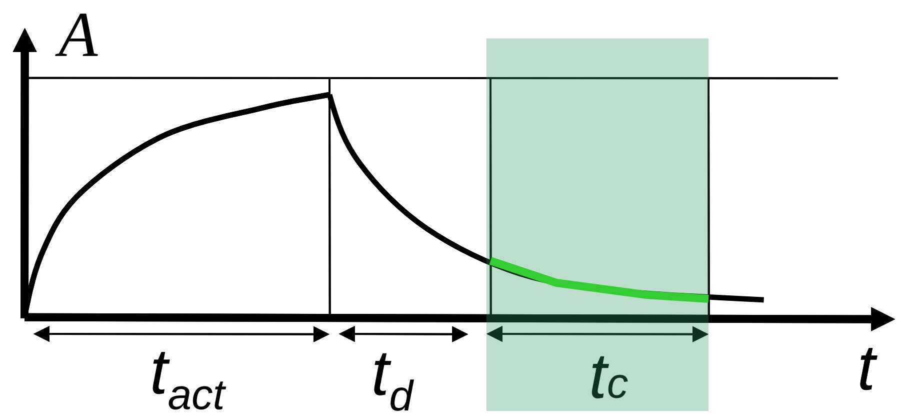
(Not always the last)

$(\sigma_\gamma)_{last} < (\sigma_\gamma)_{max}$

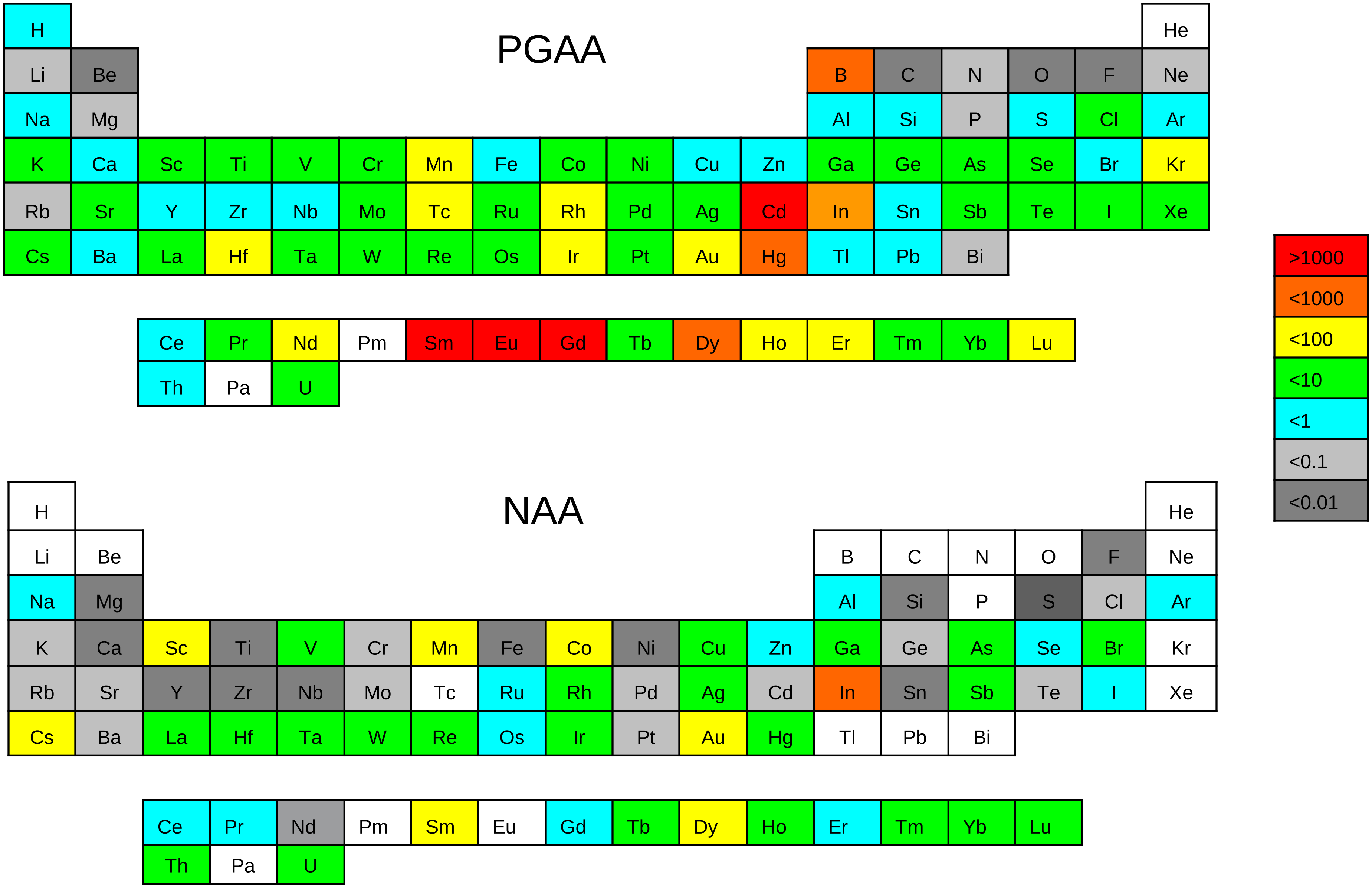
(~order of magnitude)

Activity counted after irradiation

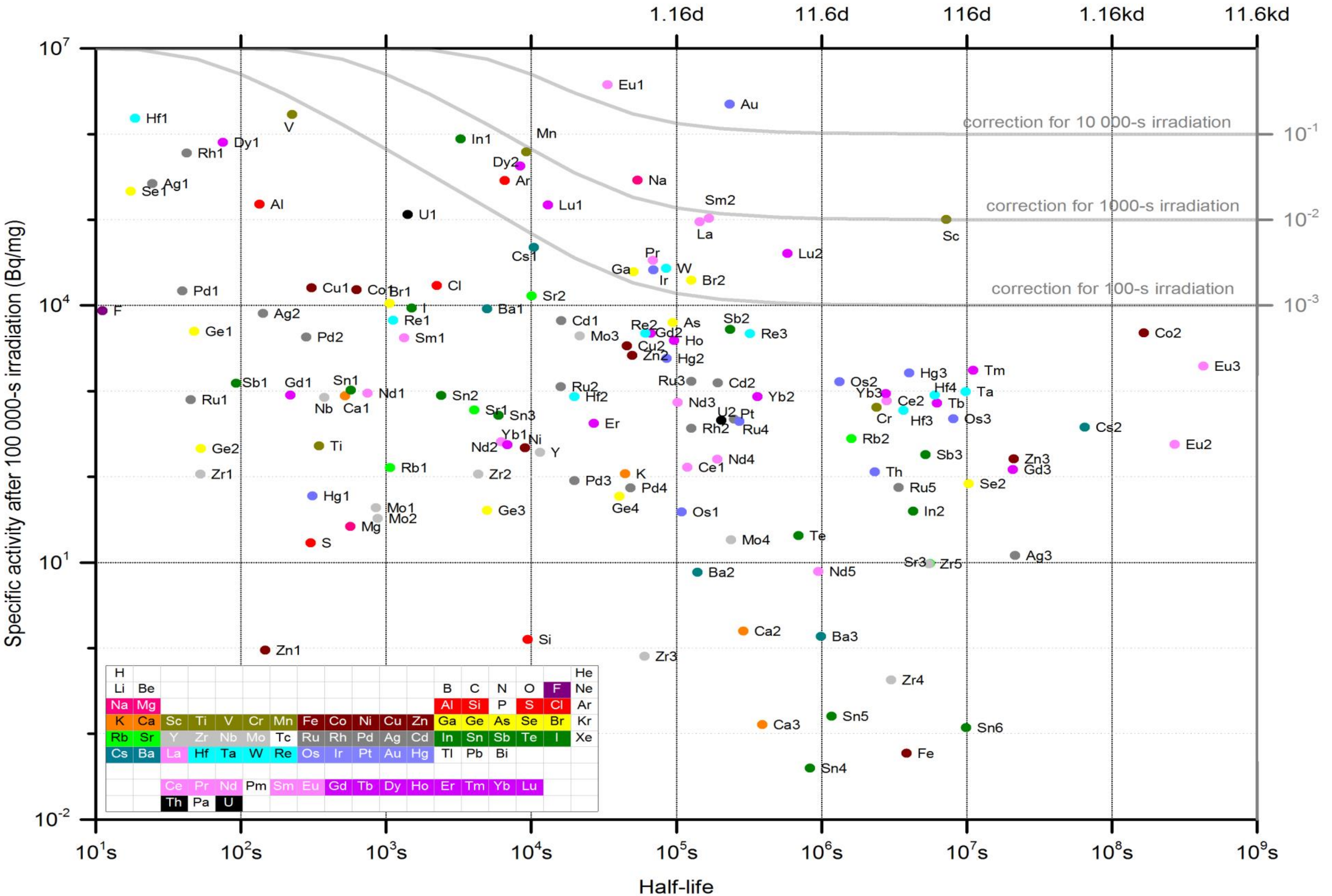




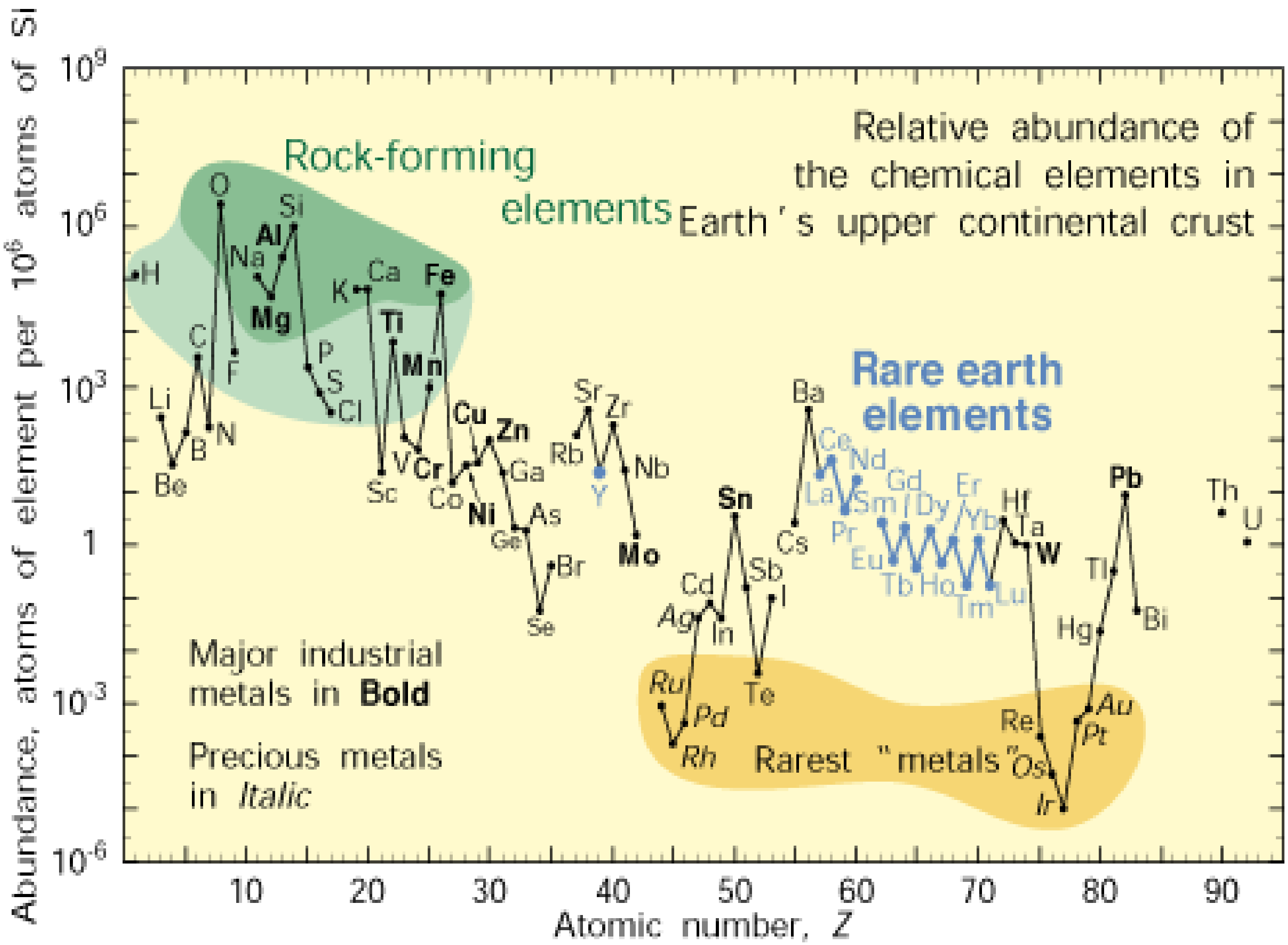
Partial cross sections for PGAA and NAA



Induced activity of different nuclides



Abundance of elements in ”natural” samples



PGAA: Light elements (H), Matrix constituents

Certain trace elements (B, Cd, REE)

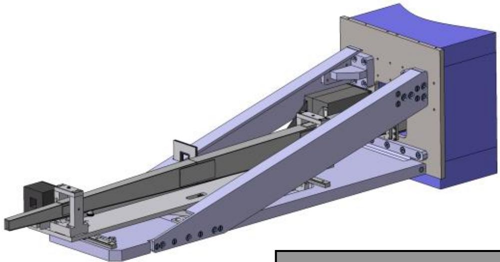
NAA: good Z>20

Trace elements

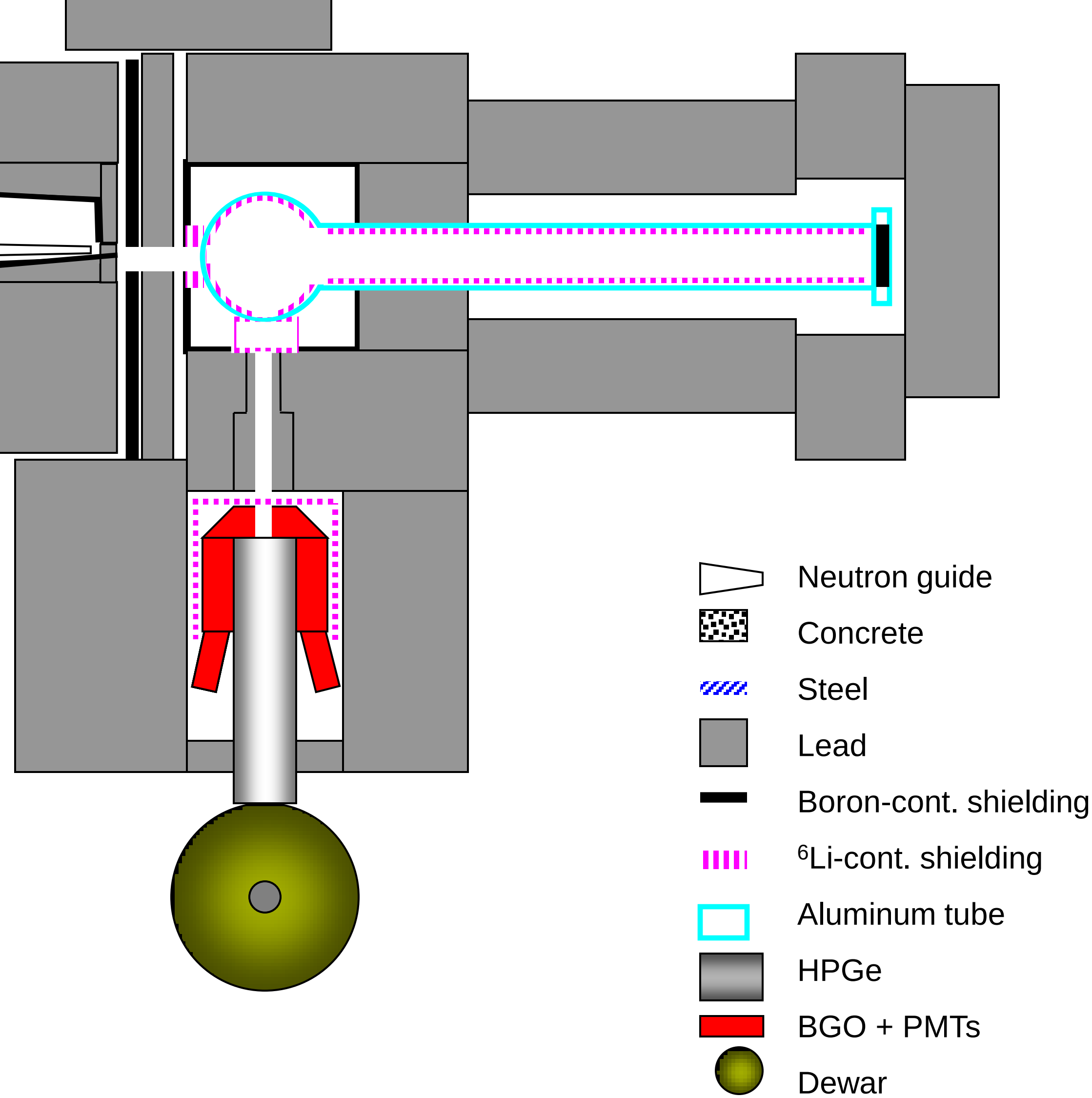
Combination of PGAA and NAA

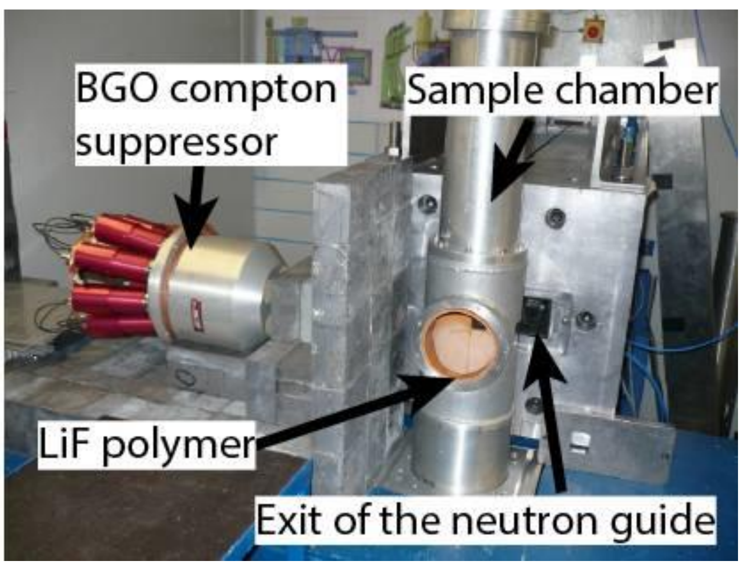
- Advantages
  - Complete analysis
  - Neutron-absorbing components are also seen
  - No flux monitoring is needed
    - If we see all (major and minor) components, from the mass ratios we can determine the concentrations
  - No matrix effect
  - Non-destructive
- Disadvantages
  - NAA needs 10^{3-5} higher flux
  - One cannot (always) perform PGAA measurement during the irradiation
  - Nuclides can mask each other
  - Samples can be active after measurements

PGAA facility



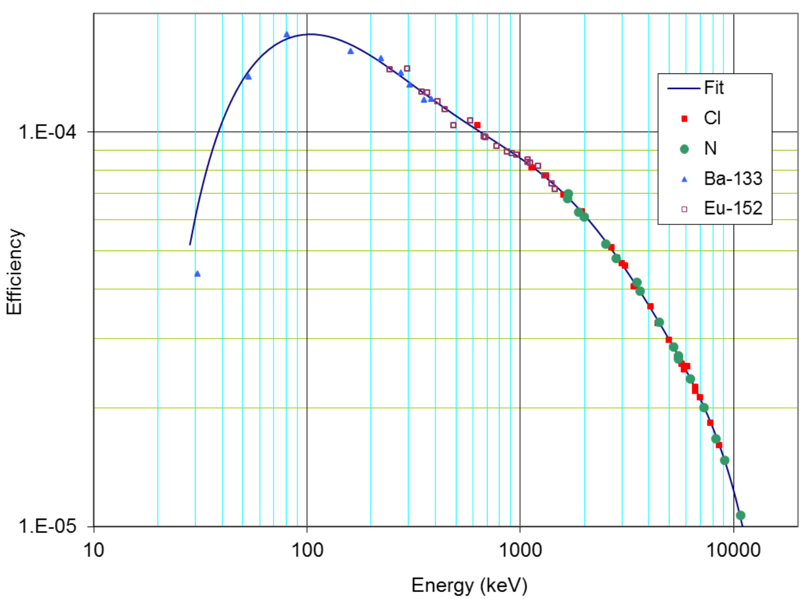
$\Phi = 3{\times}10^{10}\ \mathrm{cm^{-2}\ s^{-1}}$



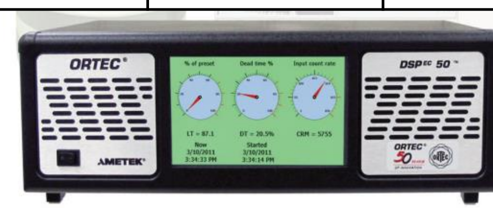


Flux, Background in the Beam, Efficiency

- Collimated beam
  - Flux: 1.35×10^{9} cm^{-2} s^{-1}
  - Beam background: 12 cps
- Focused beam
  - Flux: 2.7×10^{10} cm^{-2} s^{-1}
  - Beam background: 290 cps

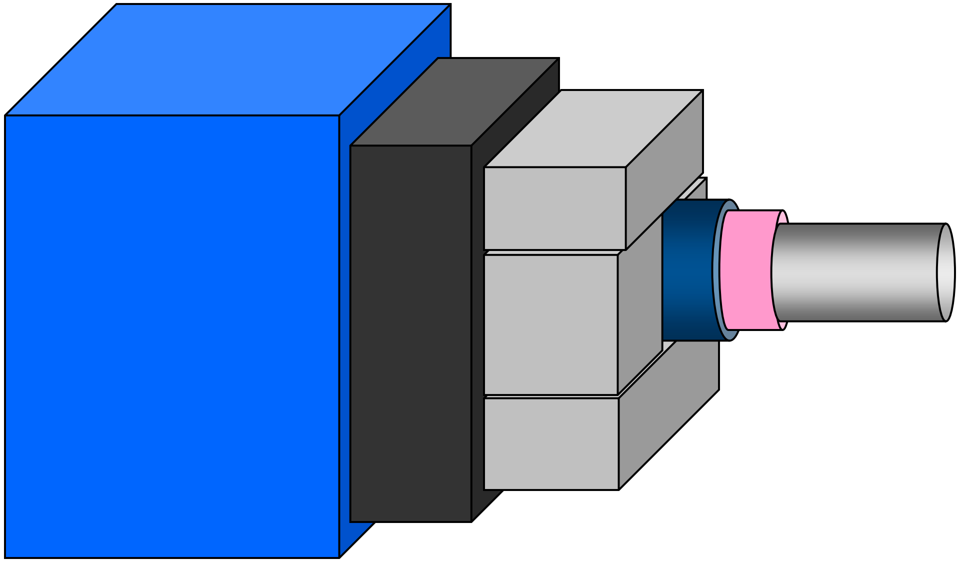


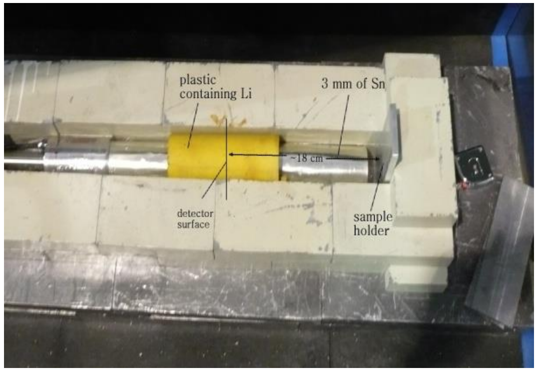
Element	collimated	focused
H	5.8×10^{-7}	3.5×10^{-7}
B	1.9×10^{-8}	1.5×10^{-8}
N	-	8.2×10^{-6}
Na*	-	9.3×10^{-6}
Al	2.2×10^{-5}	2.4×10^{-5}
Si*	-	4.0×10^{-5}
Ca*	-	3.1×10^{-6}
Ti*	-	5.6×10^{-7}
Cr	-	7.0×10^{-6}
Fe	1.0×10^{-6}	1.5×10^{-6}
Ni*	-	3.1×10^{-7}
Ge	-	1.8×10^{-6}
In	4.8×10^{-8}	5.6×10^{-8}
Pb	4.8×10^{-4}	4.6×10^{-4}

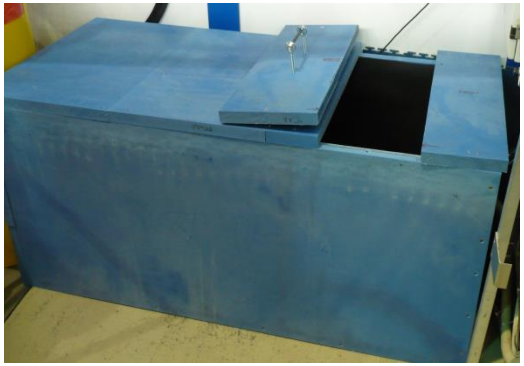


Low-background chamber next to neutron guides???

- ^{6}Li-containing plastic (2.5 mm)
- Sn sheets (4 mm) – instead of Cd
- 10 cm of lead
- 5mm boron rubber (40% B_4C)
- 5cm boron plastic (20% H_3BO_3)

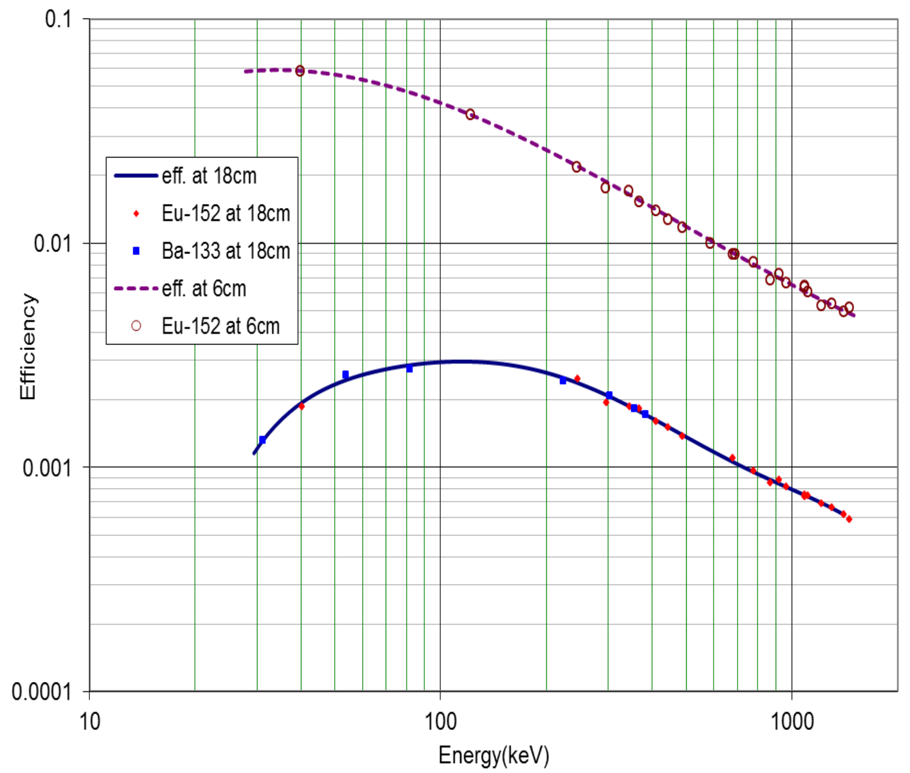






Characteristics of low-background chamber

- 2.5 cps during reactor operation
- 1.5 cps room background
- Background components from
  - 40K (1460 keV)
  - U, Th series
  - some neutron activation (Ge (n,γ) peaks, or with Cd)
  - 41Ar (1294 keV) – from reactor operation
  - 124Sb (601 keV, 1693 keV) – activated lead brick
  - Cosmic muons
- No Compton suppression



Solutions we tried

1. PGAA: short irradiation in $2x10^8$ cm^{-2} s^{-1} for 5min
   – Fluence <1% of the full-flux irradiation (mostly negligible correction)
   – To see the major components
2. NAA: longer irradiation in $4x10^{10}$ cm^{-2} s^{-1} for 60 min
   • No good spectrum will be seen
3. Decay counting with PGAA detector (to see the short-lived components)
4. Decay counting(s) in low-background chamber
   • Immediately, after 1day, after 1 week...

Verification and Applications

Reference materials

- NIST SRMs:
  - Coal fly ash (1633b)
  - Peach leaves (1547)
  - Estuarine sediment (1646a)
    - The irradiation was not really optimized for the task
    - The visible elements agreed well

  - Bovine liver (1577c)
    - 800ppb Al was confirmed as a possible new certified element
- Synthetic MultiELement Standard (SMELS)
  - Type I short-lived (minutes-hours): Cl, Cs, Cu, I, La, Mn, V
  - Type II medium-lived (hours-days): As, Br, Ce, Mo, Pr, Sb, Th, Yb, Zn
  - Type III long-lived (days-weeks-months): Co, Cr, Cs, Fe, In, Sb, Sc, Se, Sr, Th, Tm, Yb, Zn

SMELS 1 – 2

El	E	mass of the element	unc [%]	mass norm. to Au	unc%	ref value [ppm]	unc %	Z score
Au	411.8	7.22E-07	21.4	8.54E+01	21.8	82.7	2.1	0.1
Cl	1642.7	3.53E-05	10.0	4.17E+03	10.8	4330.0	3.9	-0.3
Cl	2167.4	3.33E-05	7.0	3.93E+03	8.0	4330.0	3.9	-1.1
Cs	127.5	6.55E-06	12.0	7.75E+02	12.6	897.0	4.1	-1.2
Cu	1039.2	2.96E-05	9.5	3.50E+03	10.3	3930.0	3.1	-1.1
I	442.9	1.40E-06	17.9	1.66E+02	18.3	152.0	3.3	0.4
La	1596.2	0.00E+00	1.5	0.00E+00	4.2	265.0	3.8	
La	487	0.00E+00	1.3	0.00E+00	4.2	265.0	3.8	
La	328.7	0.00E+00	1.4	0.00E+00	4.2	265.0	3.8	
Mn	846.7	9.93E-07	5.4	1.17E+02	6.7	113.9	2.9	0.4
Mn	1810.7	1.05E-06	13.0	1.24E+02	13.6	113.9	2.9	0.6
Mn	2113.1		1.0	0.00E+00	4.1	113.9	2.9	
V	1434.1	3.53E-07	10.9	4.17E+01	11.6	39.0	4.1	0.5
As	559.1	1.21E-06	2.0	9.12E+01	3.0	92.3	3.9	-0.3
As	657	1.35E-06	4.0	1.02E+02	4.5	92.3	3.9	1.6
As	1216.1	1.25E-06	8.2	9.43E+01	8.5	92.3	3.9	0.2
Au	411.8	5.21E-08	1.3	3.93E+00	2.5	3.9	1.8	0.0
Br	554.3	2.09E-06	1.7	1.58E+02	2.8	157.0	3.2	0.1
Br	619.1	2.06E-06	2.5	1.56E+02	3.4	157.0	3.2	-0.2
Br	776.5	2.06E-06	1.6	1.56E+02	2.7	157.0	3.2	-0.2
Ce	293.26	2.03E-04	1.2	1.53E+04	2.5	15600.0	5.1	-0.3
Ce	231.55	1.88E-04	3.8	1.42E+04	4.4	15600.0	5.1	-1.4
Ce	664.57	2.12E-04	2.9	1.60E+04	3.7	15600.0	5.1	0.4
Mo	140.5	6.54E-05	1.6	4.93E+03	2.7	5170.0	4.8	-0.8
Mo	739.5	7.63E-05	7.0	5.76E+03	7.3	5170.0	4.8	1.2
Pr	1575.8	1.59E-05	1.4	1.20E+03	2.6	1193.0	3.1	0.1
Sb	564.1	2.31E-06	1.9	1.74E+02	2.9	172.0	4.7	0.3
Sb	692.7	2.35E-06	8.7	1.78E+02	9.0	172.0	4.7	0.3
Sb	1691	2.62E-06	4.4	1.97E+02	4.9	172.0	4.7	2.0
Th	300.1	4.93E-05	1.7	3.72E+03	2.8	3670.0	4.9	0.2
Th	311.9	5.03E-05	1.2	3.79E+03	2.5	3670.0	4.9	0.6
Th	340.5	4.83E-05	2.3	3.64E+03	3.1	3670.0	4.9	-0.1
Yb	282.5	2.61E-06	1.6	1.97E+02	2.7	187.0	5.3	0.9
Yb	396.3	2.64E-06	1.5	1.99E+02	2.6	187.0	5.3	1.0
Zn	1115.5	8.70E-05	5.4	6.56E+03	5.8	6570.0	3.0	0.0

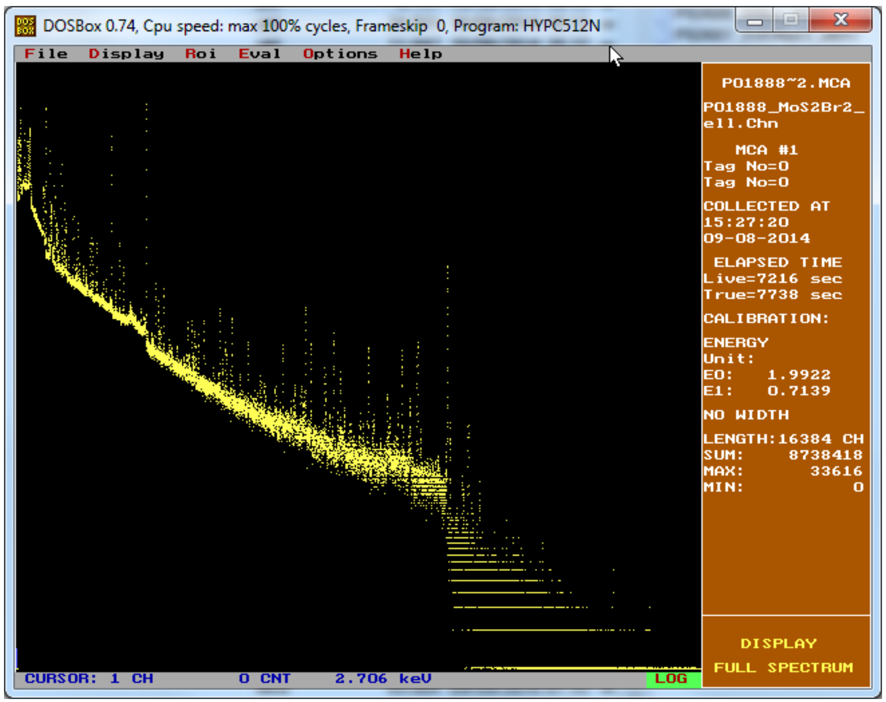
- Short-lived
  - Irradiation 20 min
  - Counting 40 min
- Medium-lived
  - Irradiation 18 h
  - Counting 2 days
- Good agreement

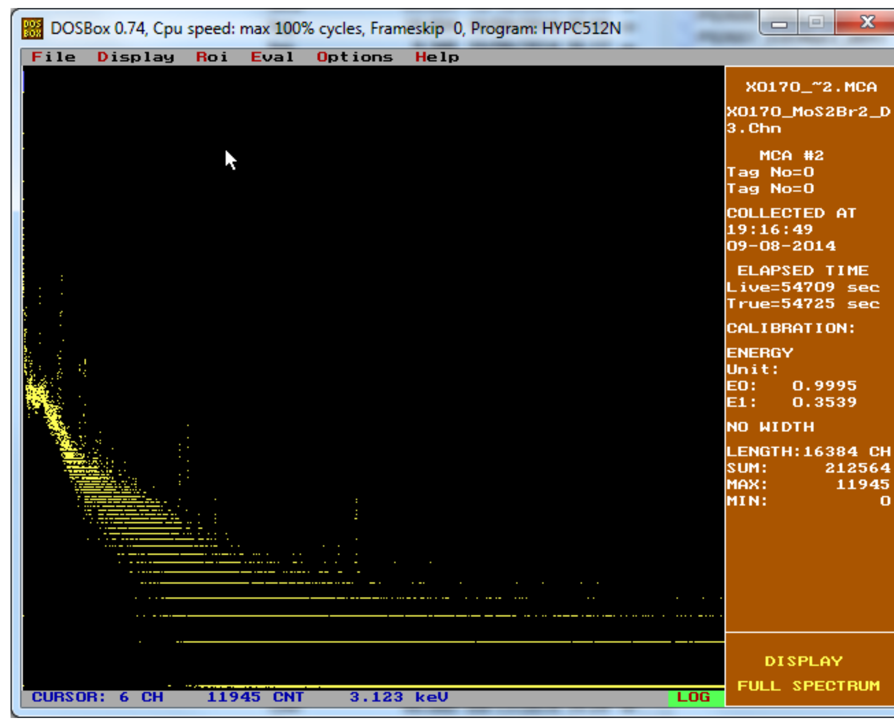
SMELS 3 in beam and in reactor

- In reactor
  - Irradiation 20 min in 3×10^{13} cm^{-2} s^{-1}
  - Counting 15 h
  - Systematic discrepancies for Co, Cs, Sb, ... High resonance integrals (Q_0 = 2, 12, 20)
  - The irradiation channel needs to be characterized (f, α)
- In beam
  - In good agreement

Pollutants of MoS_2

- From prompt spectrum:
  - B 0.067--0.87--240 ppm   Cl 0.4 ppm   Mo 59 ppm
- From decay spectra:
  - La 300 ppm   Br 500 ppm   Ir 0.29%   In 1.7 ppm





Pollutants in MgO

Z	El	M	m meas	unc %	w% el/ox	unc %
12	Mg	24.31	0.252	5.	56	2.2
		0				
1	H	1.008	3.82E-3	0.4	0.843	2.8
5	B	10.81	2.59E-6	0.3	5.69 ppm	2.8
17	Cl	35.45	3.72E-6	13.	8 ppm	14.
19	K	39.1	2.60E-5	5.	57 ppm	6.
20	Ca	40.08	9.45E-4	30.	0.2	30.
25	Mn	54.94	4.90E-7	11.	1.1 ppm	11.
47	Ag	107.9	1.10E-4	4.0	240 ppm	5.
49	In	114.8	1.40E-7	3.0	0.31 ppm	4.
57	La	138.9	9.23E-5	7.	200 ppm	8.
64	Gd	157.3	1.25E-8	8.	0.028ppm	8.
		0				
		0				
		0				
		0				

- Smallest masses
  10^{-7}—10^{-8} g
- Detection limits
- Down to 10^{-10} g

Irradiation radioactive nuclides

Induced activity > natural radioactivity

- MBq activities
- Half-life >100a
- Mass > 0.1mg
- Cross-sec. >100b

Radioactive samples irradiated at FRM2:

- 30µg (!!) ^{241}Am
- ~100 µg ^{237}Np, ^{242}Pu

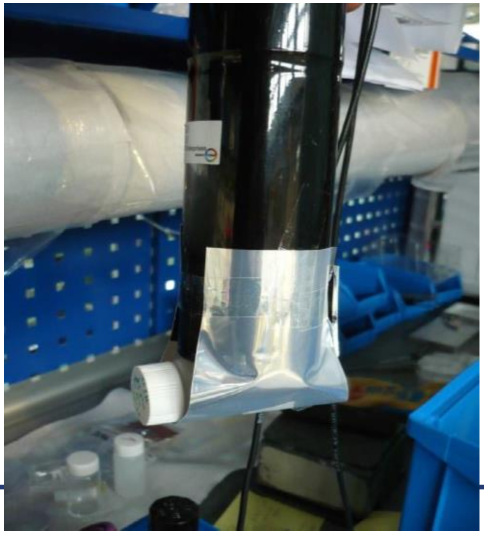
Determination of cross section

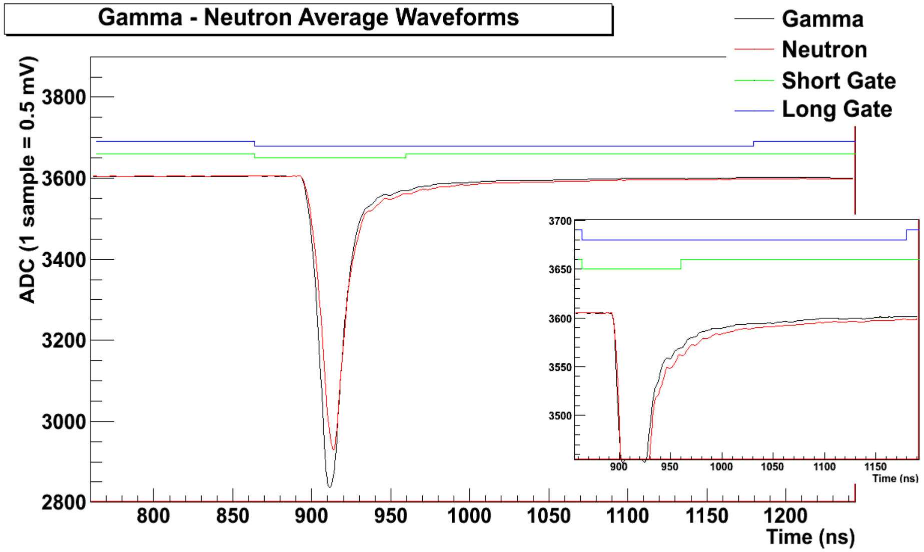
Used in the actinide project

In-beam liquid scintillation

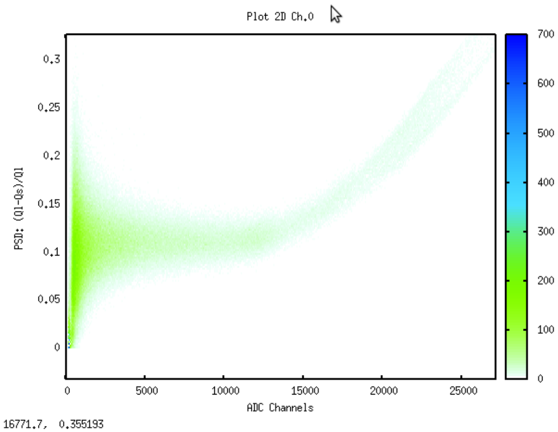
- For detecting n-induced charged particles
- $^{10}B(n,\alpha)^{7}Li$, $^{6}Li(n,t)^{4}He$, $^{14}N(n,p)^{14}C$

- It works well off beam
- In beam too high gamma background
- Needs pulse shape discrimination

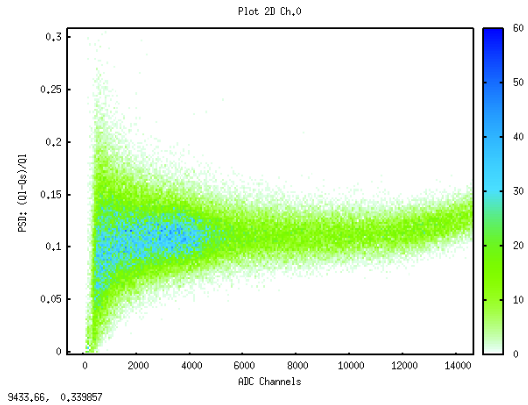




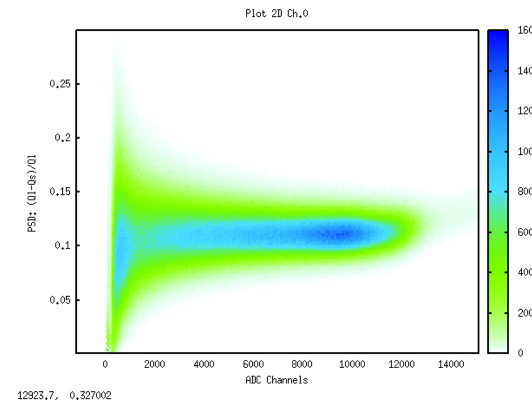
Pulse-shape-Energy 2D histograms



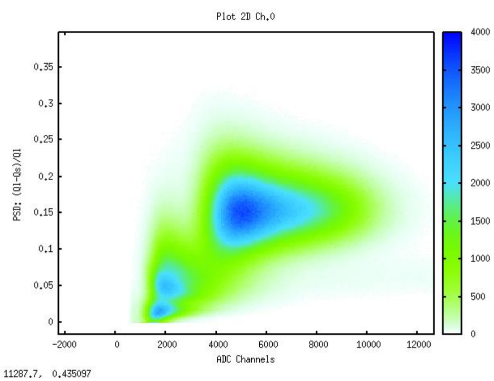
Long background



0.6MeV β^- (^{124}Sb)

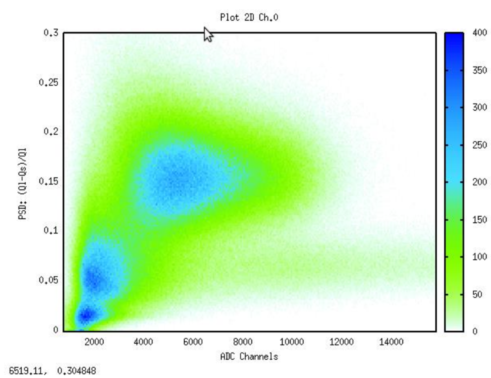


1.3 MeV γ (^{60}Co)



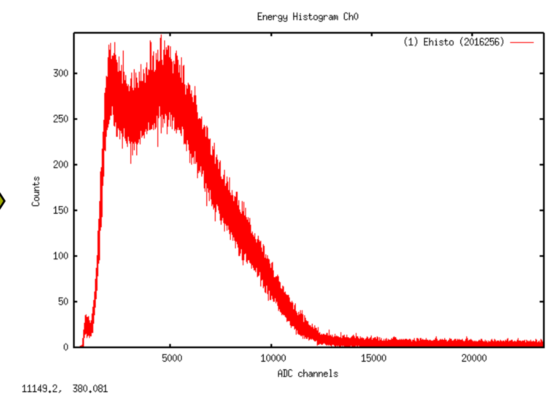
5 MeV α (^{210}Po)

0.3 MeV β^- (^{87}Rb)



5 MeV α (^{210}Po)

0.6MeV β^- (^{124}Sb)



Energy spectrum