

Budapest Neutron Centre Centre for Energy Research



Prompt gamma neutron activation analysis

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Budapest Neutron Centre

<https://www.ek.hun-ren.hu/en/budapest-neutron-centre-bnc-2/>



Nuclear analytical chemistry ...



- ... is a branch of analytical science, where the natural or induced radioactivity, or nuclear reactions are used to obtain information from the composition of the sample



- Most interactions are independent of the chemical environment (i.e. interactions with the atomic nucleus or with the core electrons), elemental analysis can be done



What is activation analysis?

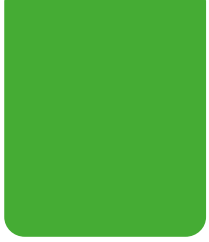
1932 – **James Chadwick** discovers the neutrons (beryllium bombarded with alpha particles from polonium) - 1935 Nobel Prize in Physics

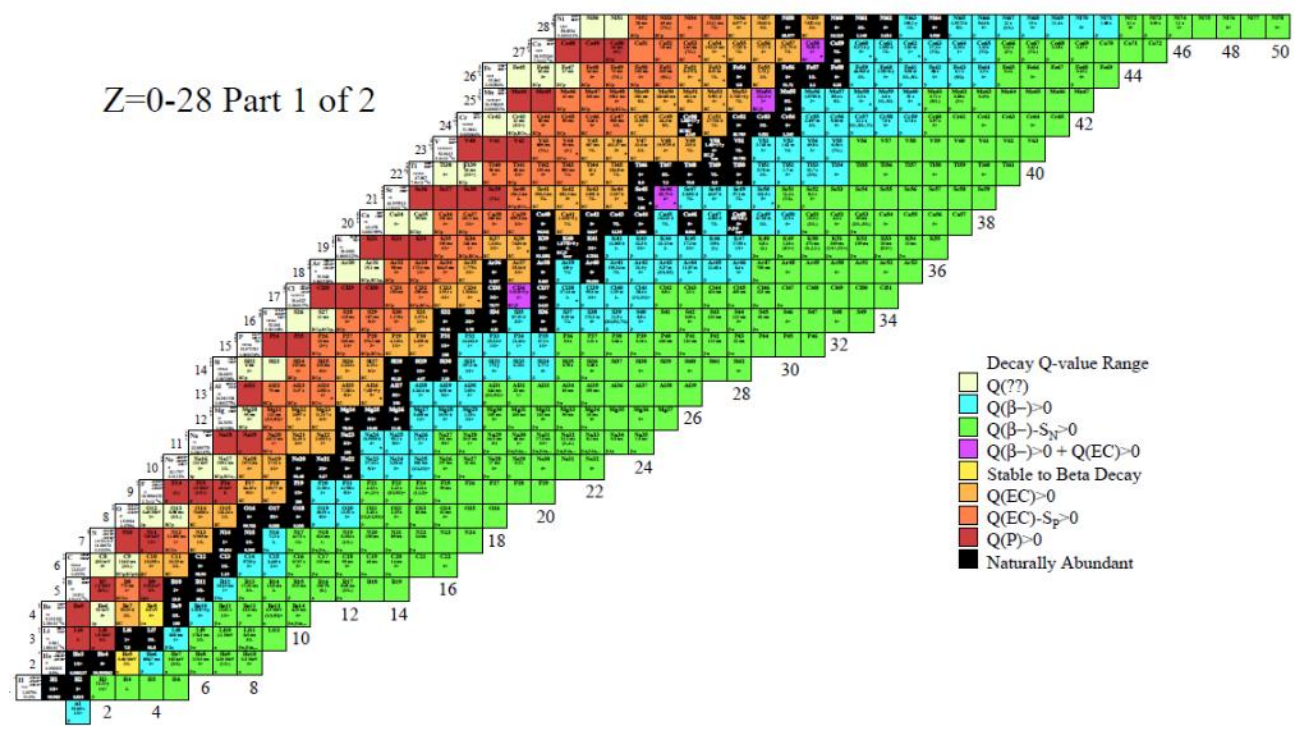
- Neutron Activation Analysis (NAA)
 - The application of neutron-induced radioactivity for element analysis
 - **György Hevesy** 1936 (discovery of Hf, radioactive tracing, Nobel prize 1943)
- Prompt Gamma Activation Analysis (PGAA)
 - The application of radiative neutron capture ((n, γ) reaction) for chemical analysis
 - First observed in 1934: $^1\text{H}(n,\gamma)^2\text{H}$ (**Douglas Edward Lea**)
 - Idea of PGAA technique and first demonstration PGAA setup 1963 & 1968: **Heinz Maier-Leibnitz** (FRM Research Reactor, Garching)

Irradiation: with neutrons

Target: atomic nuclei

Emitted: delayed or prompt gamma photons





Isotopes – nuclides with same Z

Isobars – nuclides with same A

Isotones – nuclides with same neutron number

Mass number (number of protons + neutrons)

A

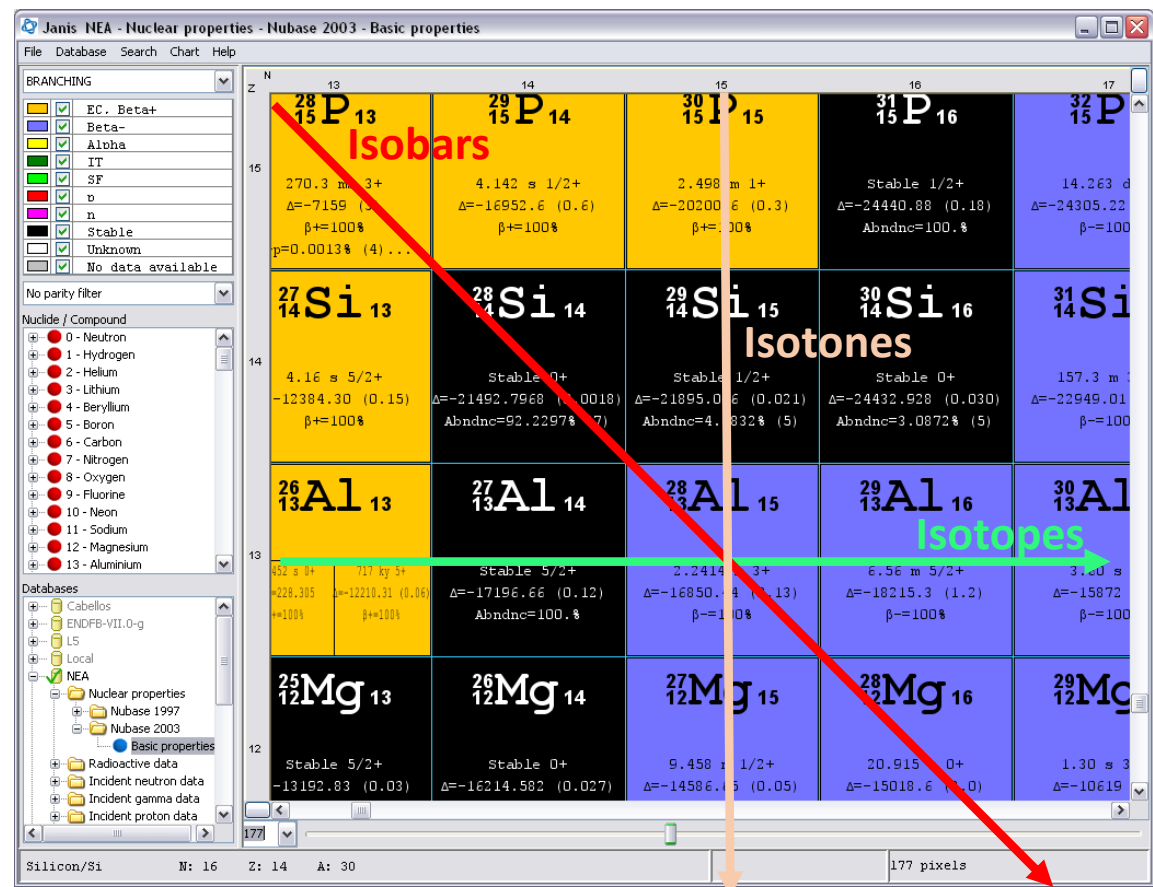
X

Z

X

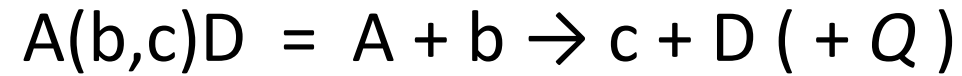
Element symbol

Atomic number (number of protons)





nomenclature:



A – target nucleus

D – final nucleus

b – projectile

c – emitted particle

Q – reaction energy (+ exothermal, - endotherm)



The **neutron** is a neutral particle with mass of $1.67492716 \times 10^{-27}$ kg, 1.00866491560(55) dalton, 939.565360(81) MeV

The free neutron is unstable, **decays with a half-life of** $T_{1/2} = 885.7 \pm 0.8$ s to a proton, an electron and an anti-electronneutrino.

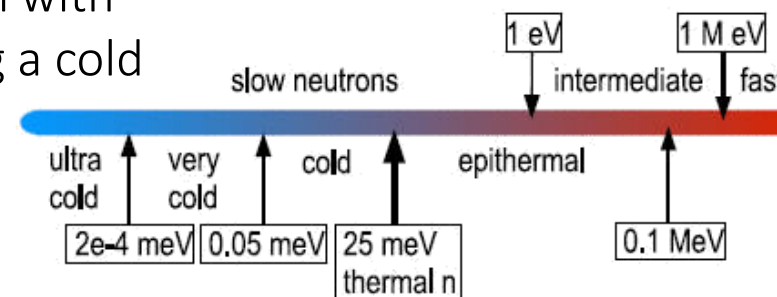
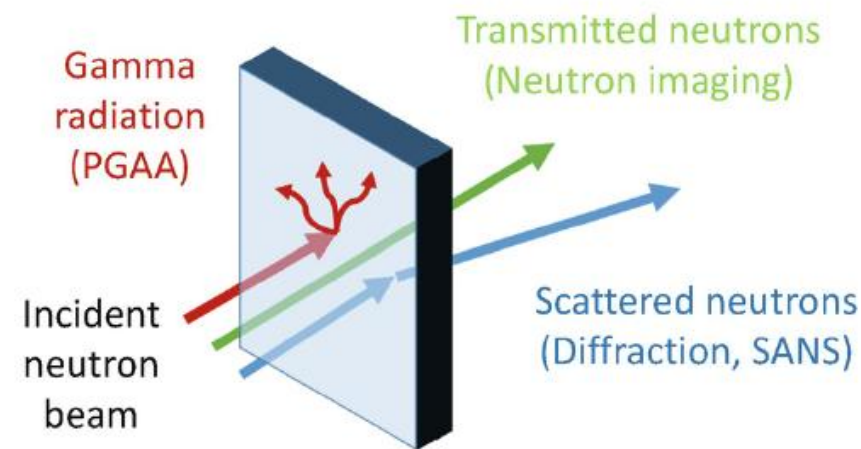
Free neutrons can be produced only by nuclear reactions.

The **neutrons are classified** according to the kinetic energy:

- ☐ Thermal : $kT = 25,26$ meV*, $v = 2198$ m s⁻¹, Maxwell-distribution
- ☐ Lower-than-thermal, i.e. approx. 5 meV: cold,
- ☐ > 0.1 eV epithermal,
- ☐ MeV: fast.

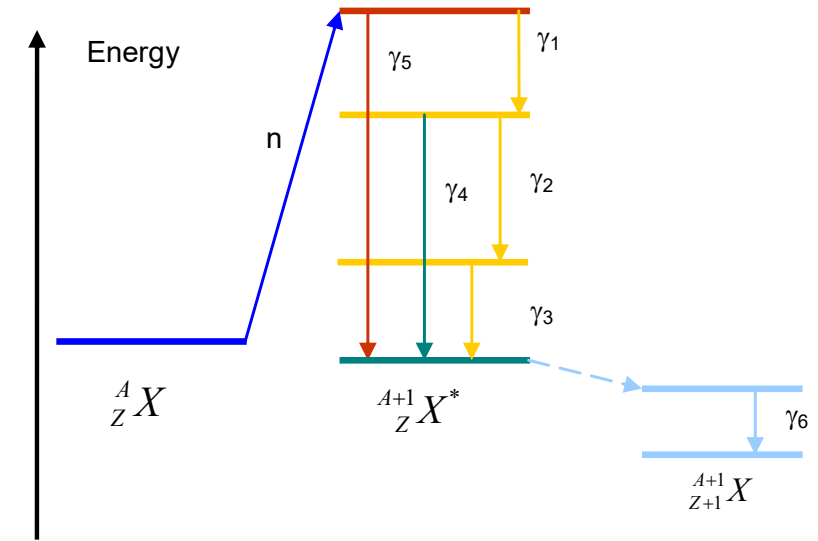
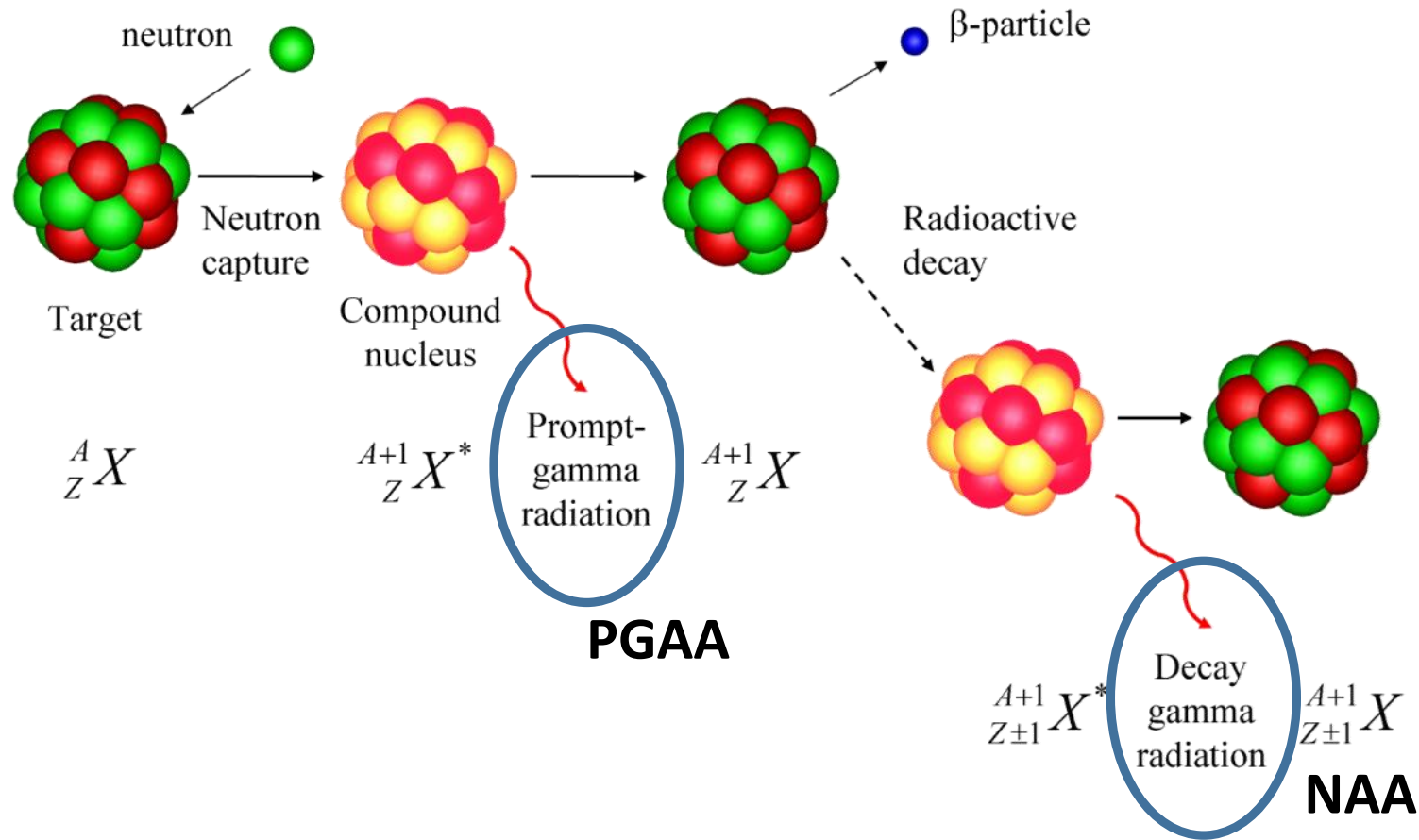
- ☐ **Moderation**: the neutron comes to a thermal equilibrium with the ambient media as it passes through a material. Using a cold material, the velocity of neutrons can be decreased.

* $1 \text{ eV} = 1.602176462(63) \times 10^{-19} \text{ J}$





The excitation energy of the compound nucleus
~ binding energy of the neutron (7–9 MeV)
(kinetic energy of the neutron is negligible).



The compound nucleus can decay through various de-excitation channels, followed by the emission of γ -photons or charged particles.

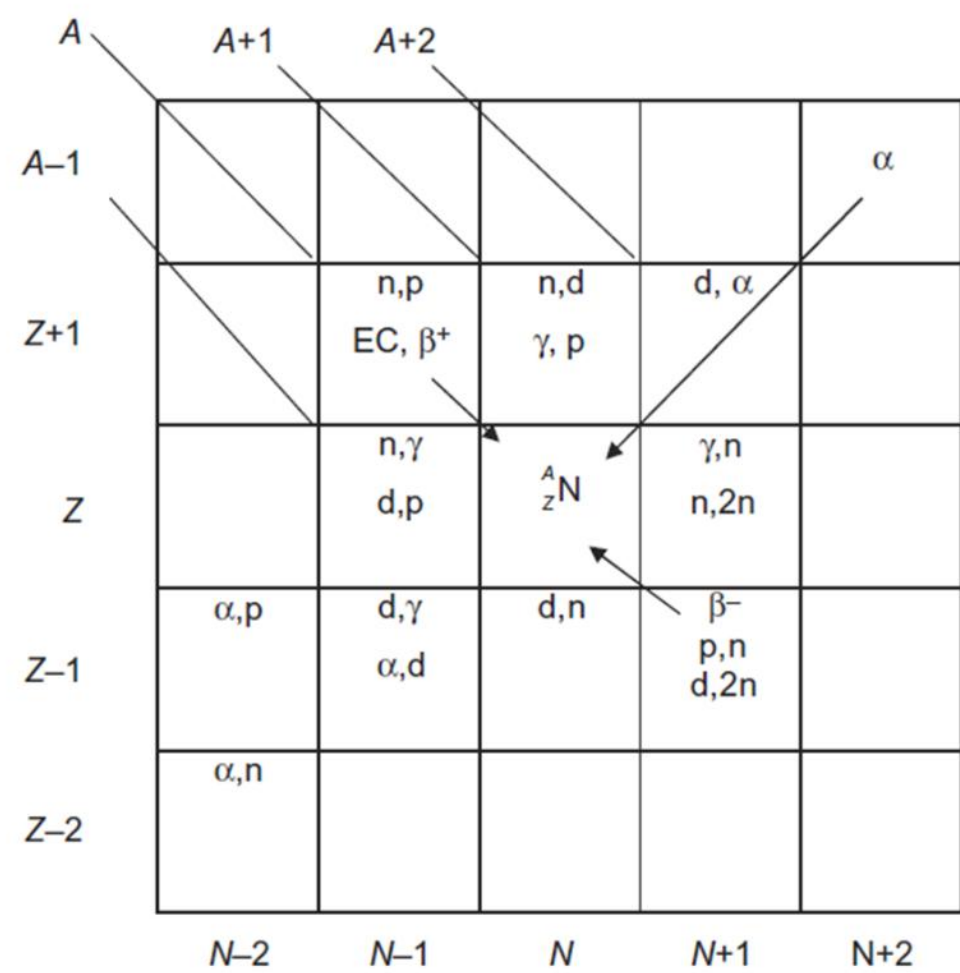


Figure 6.10 Summary of different options to produce a nuclide with Z atomic number and A mass number. It includes the formation of the nuclide by radioactive decays as well.

Ga63 32.4 s 3/2-, 5/2- EC	Ga64 2.627 m 0+ EC	Ga65 15.2 m 3/2- EC	Ga66 9.49 h 0+ EC	Ga67 3.2612 d 3/2- EC	Ga68 67.629 m 1+ EC	Ga69 3/2- 60.108
Zn62 9.186 h 0+ EC	Zn63 38.47 m 3/2- EC	Zn64 0+ 48.6	Zn65 244.26 d 5/2- EC	Zn66 0+ 27.9	Zn67 5/2- 4.1	Zn68 0+ 18.8
Cu61 3.333 h 3/2- EC	Cu62 9.74 m 1+ EC	Cu63 3/2- 69.17	Cu64 12.700 h 1+ EC, β^-	Cu65 3/2- 30.83	Cu66 5.088 m 1+ β^-	Cu67 61.83 h 3/2- β^-
Ni60 0+ 26.223	Ni61 3/2- 1.140	Ni62 0+ 3.634	Ni63 100.1 y 1/2- β^-	Ni64 0+ 0.926	Ni65 2.5172 h 5/2- β^-	Ni66 0+ 54.6 h
Co59 7/2- 100	Co60 5.2714 y 5+ *	Co61 1.650 h 7/2- β^-	Co62 1.50 m 2+ β^-	Co63 27.4 s (7/2)- β^-	Co64 0.30 s 1+ β^-	Co65 1.20 s (7/2)- β^-



Components of the activation analysis instrument

- Neutron source
 - spallation neutron source
 - nuclear reactor
 - neutron generator
 - isotopic neutron source (^{252}Cf , Pu-Be, ...)
- Moderator (H containing material)
- Shielding against ...
 - Neutrons (PGAA)
 - Gammas
- Pneumatic transfer (= rabbit) system (only in NAA)
- Detectors
 - high-purity germanium (HPGe)
 - scintillator (NaI, ...)
 - Low-background counting chamber (NAA)



- 10 MW tank type
- Water-cooled
- Water-moderated
- thermal flux
 $10^{14} \text{ cm}^{-2} \text{ s}^{-1}$



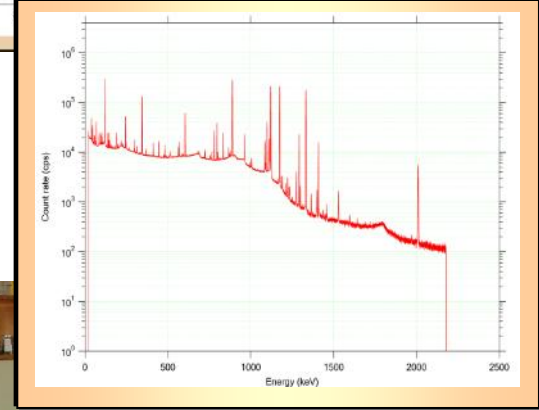
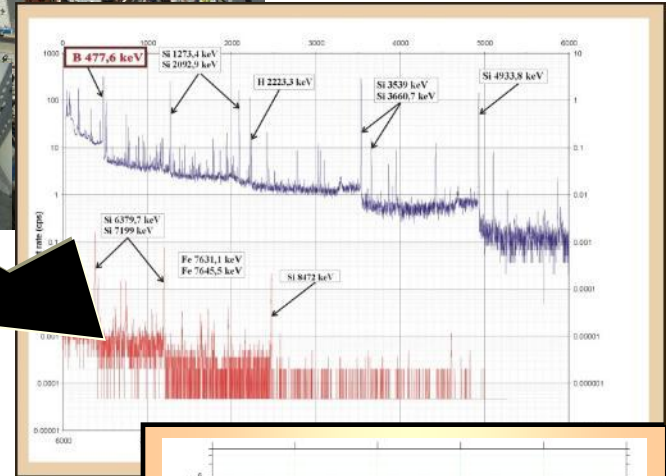
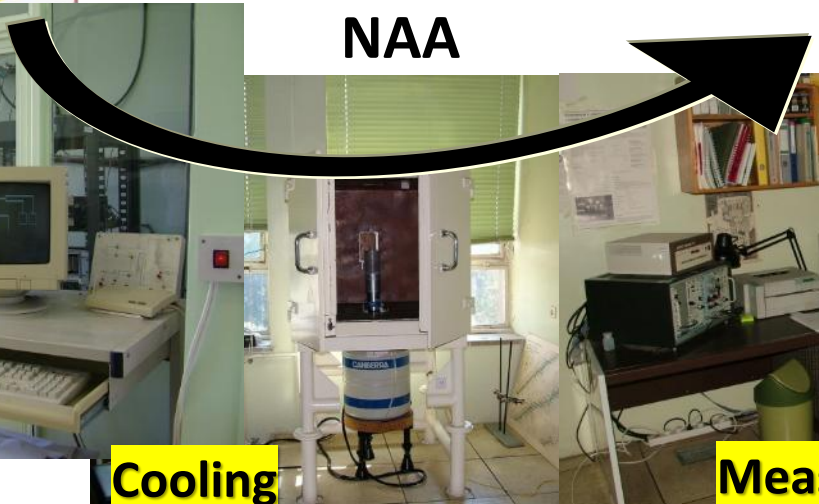
**TRADITIONAL NEUTRON INSTRUMENTS:**

RAD: DYNAMIC NEUTRON/GAMMA & STATIC RADIOGRAPHY
 BAGIRA4: BUDAPEST ADVANCED GAS-COOLED IRRADIATION RIG WITH ALUMINIUM STRUCTURE
 BIO: PORT USED FOR BIOLOGICAL IRRADIATION
 MTEST: MATERIAL TESTING DIFFRACTOMETER
 TAST: TRIPLE AXIS SPECTROMETER ON THERMAL BEAM
 PSD: POWDER DIFFRACTOMETER
 NAA: NEUTRON ACTIVATION ANALYSIS (VERTICAL IRRADIATION CHANNEL)
 TOF-ND: TIME-OF-FLIGHT DIFFRACTOMETER

COLD NEUTRON INSTRUMENTS:

GINA: POLARIZED NEUTRON REFLECTOMETER
 IBMS: IN-BEAM MÖSSBAUER SPECTROMETER
 ATHOS: Triple-Axis Spectrometer
 SANS: SMALL ANGLE SCATTERING SPECTROMETER
 PGAA: PROMPT GAMMA ACTIVATION ANALYSIS
 NIPS-NORMA: NEUTRON INDUCED PROMPT GAMMA SPECTROMETER – NEUTRON OPTICS AND RADIOGRAPHY FOR MATERIAL ANALYSIS
 REF/V14: REFLECTOMETER
 DÖME: LOW BACKGROUND COUNTING CHAMBER

Overview of the Reactor Hall and the Cold Neutron Guide Hall (CNGH)



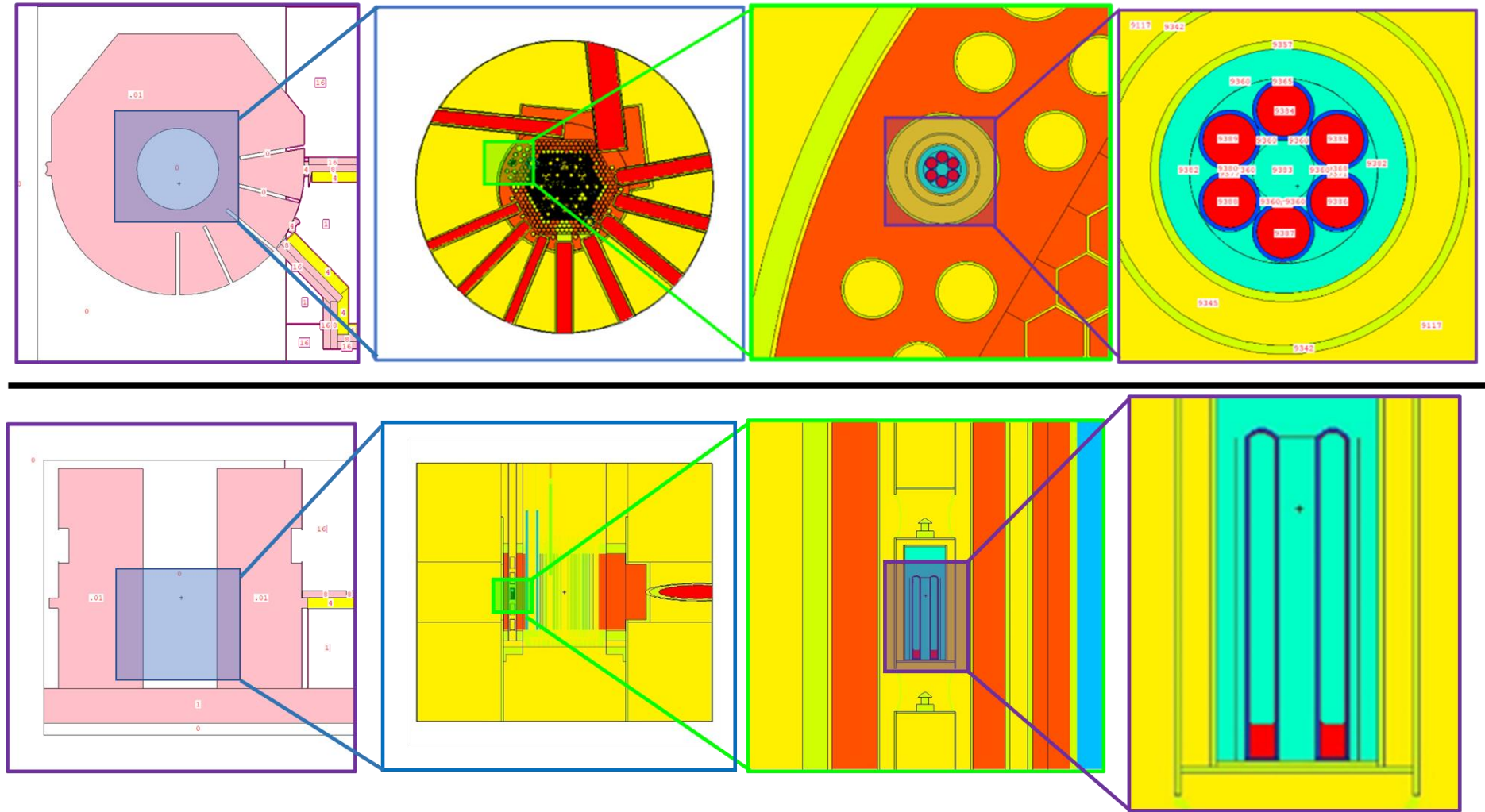
Irratiation in the reactor core

Cooling

Measurement in low-background chamber

Full-scale MCNP6 model of the 10 MW Budapest Research Reactor

by András Horváth



Reactor biological shielding

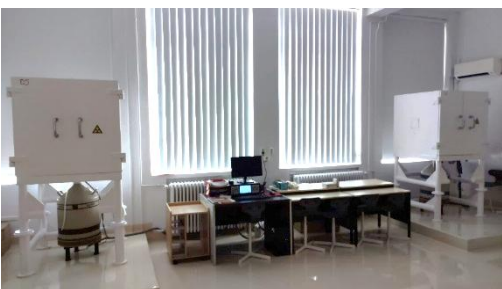
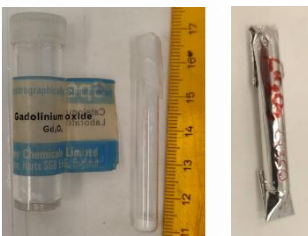
Reactor vessel

Irradiation position No.17

Samples in quartz glass vials



Multi element methods
Negligible matrix effect,
Good reproducibility,
Reliable uncertainty budget



- Sample
- Sample preparation
- Irradiation
- Detection
- Spectrum
- Result turnover time
- Detection limits
- Cooling time after irradiation



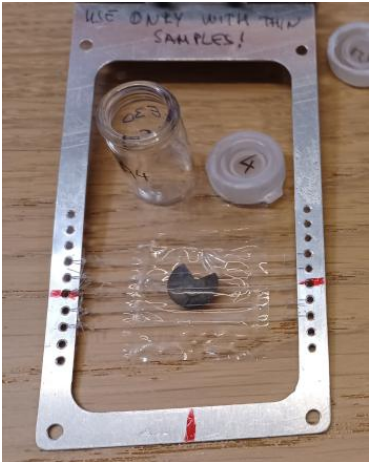
NAA

10 mg
Drying, glass
Reactor core
Separated in time and space
10-100 peaks, 3 MeV
weeks
ppm-ppb
Several weeks or months

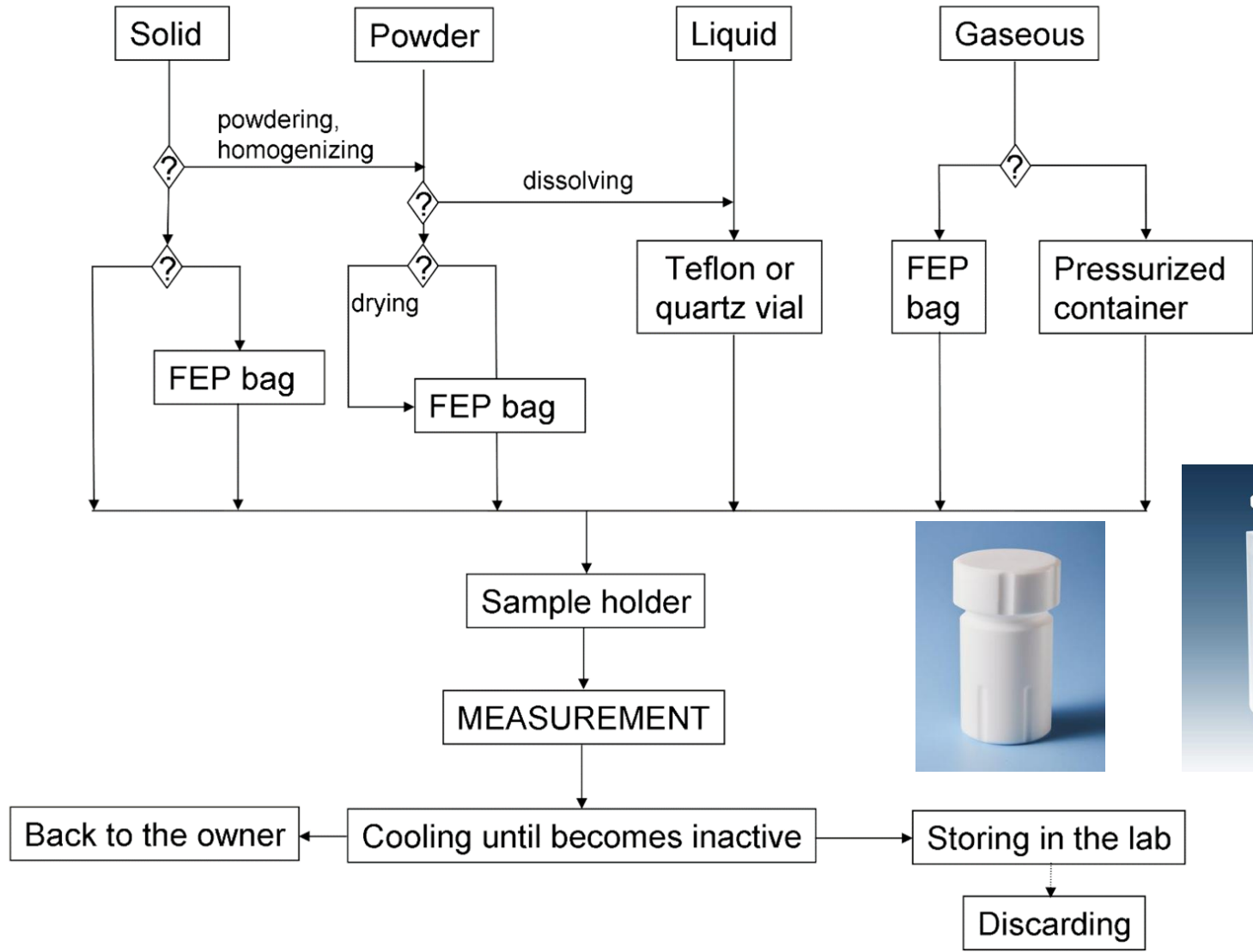
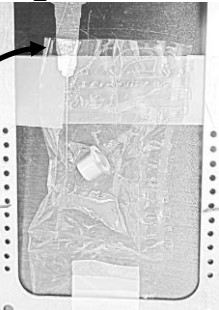


PGAA

100 mg - 1 g or more
None/ PTFE bag, vial
Guided beam of neutrons
On-line, during the irradiation
100-5000 peaks, 12 MeV
Few hours
>ppm, %
1-2 days



N₂ gas in





H 1 1.00794 0.3326 b 82.02 b		He 2 3.01603 4 4.002602 0.007 b 1.34 b	
Li 3 6.941 70.5 b 1.37 b		Be 4 9.0122 0.0076 b 7.63 b	
Na 11 22.98977 0.530 b 3.28 b		Mg 12 24.305 0.063 b 3.71 b	
K 19 39.0983 2.1 b 1.96 b		Ca 20 40.078 27.5 b 23.5 b	
Rb 37 85.4678 0.38 b 6.25 b		Sr 38 87.62 1.28 b 6.25 b	
Cs 55 132.90545 29.0 b 3.90 b		Ba 56 137.327 8.97 b 3.38 b	
(Fr) (223)		(Ra) (226)	
(Ac) (227)		104 105 106	

PGAA

Zsolt Révay, 2015

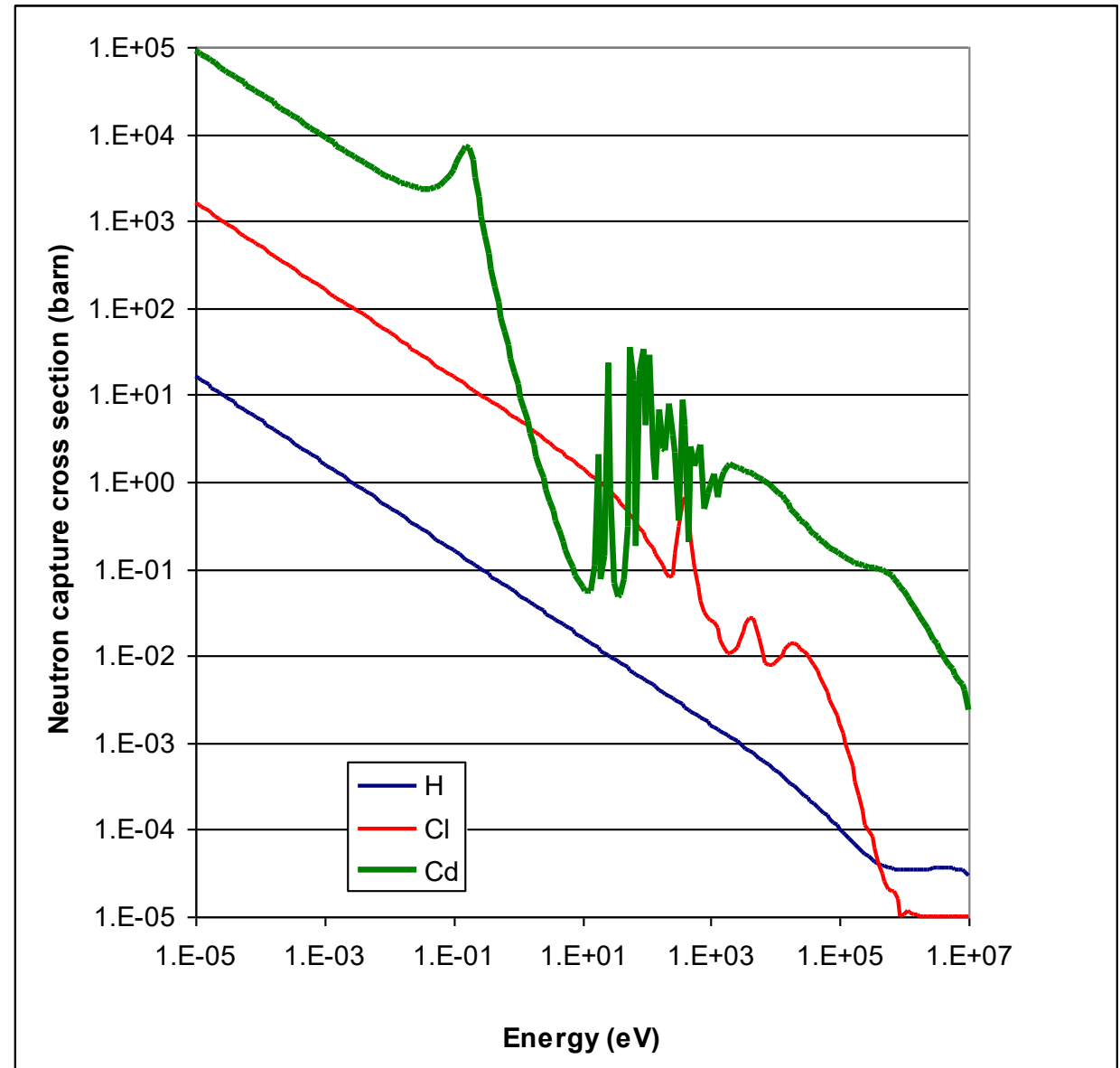
No light elements!

No light elements!																					
1 H																2 He					
3 Li	4 Be															5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg															13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr				
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe				
55 Cs	56 Ba	57 ¹ La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn				
87 Fr	88 Ra	89 ² Ac	104 Rf	105 Db																	
¹ Lanthanide		58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu						
² Actinide series		90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr						
		No n-gamma radioactive isotopes																			
		Radioactive isotopes can be produced. Limitation is short half-life or flux energy																			
		Elements routinely determined by INAA																			

NAA



- neutron capture cross section
- at low E: $1/v$ law
 - highest reaction rate for **cold neutrons**
- neutrons should be moderated or cooled





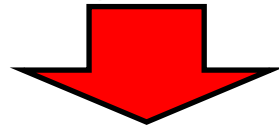
Cold neutron source at the Budapest Neutron Centre (operational since 2000)

400 cm³

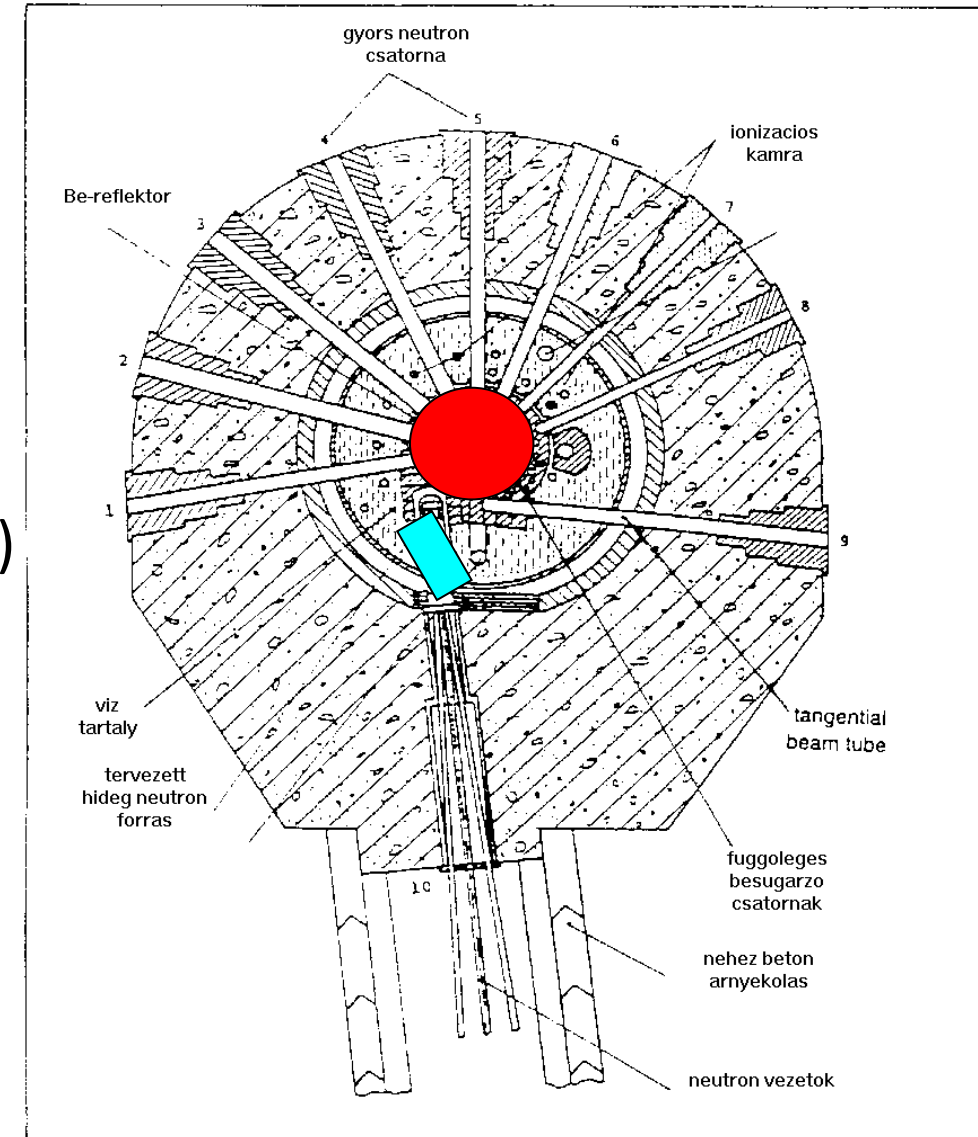
20 K liquid H₂

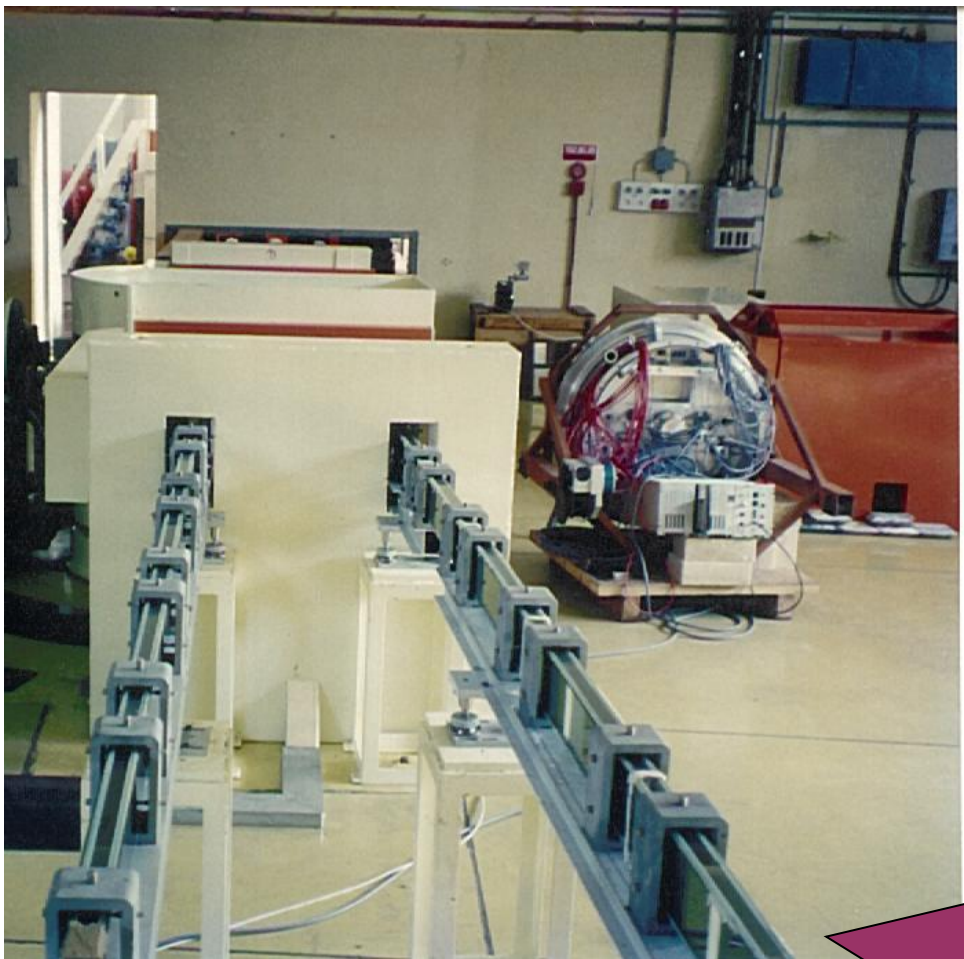
Advantages of cold neutron beams

- Higher flux (neutron guides higher throughput)
- every nuclide behaves regularly
 - (follows the $1/v$ -law)
- every nuclide has higher cross section

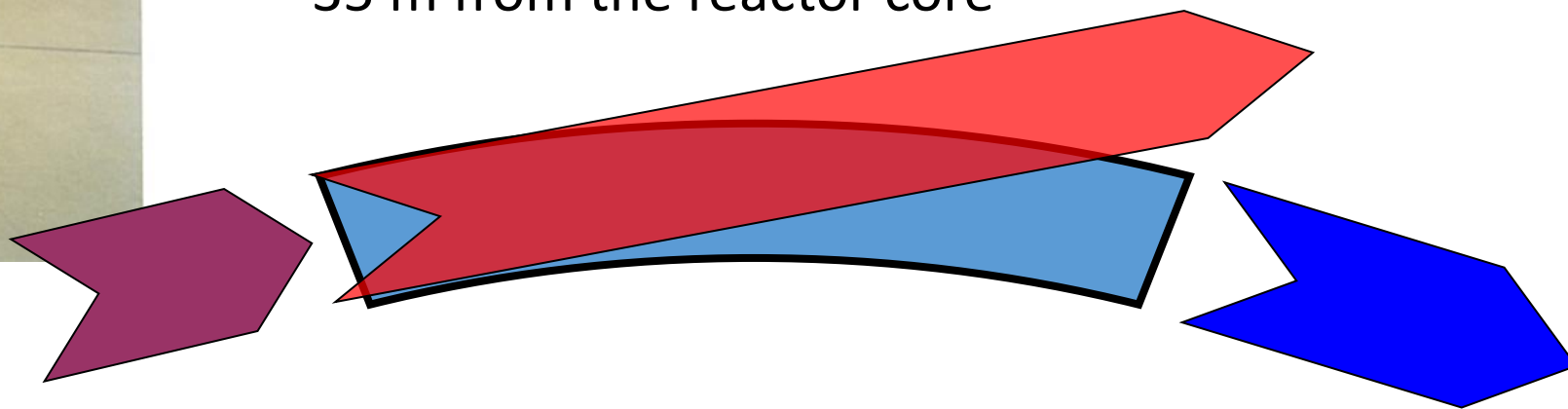


a higher reaction rate

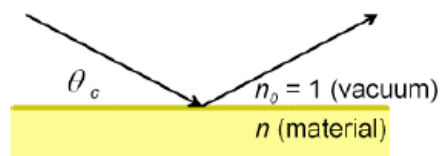




- the guides are the extension of a horizontal neutron beam
- evacuated Ni or supermirror guides
- relatively small losses
 - Cold and thermal neutrons are transmitted
 - Epithermal and fast neutrons are filtered out
- low background
 - 35 m from the reactor core

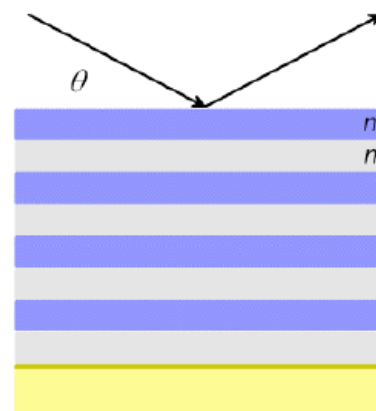


single mirror



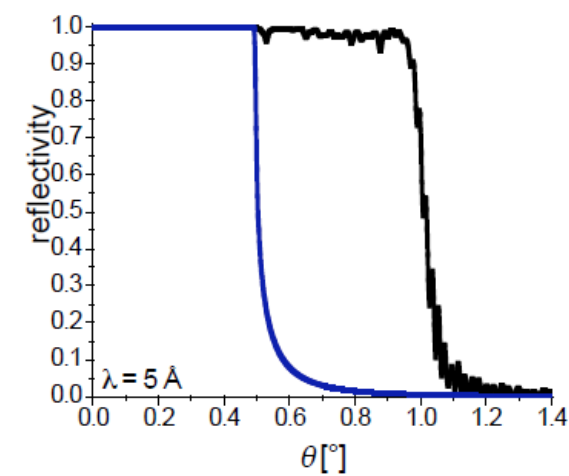
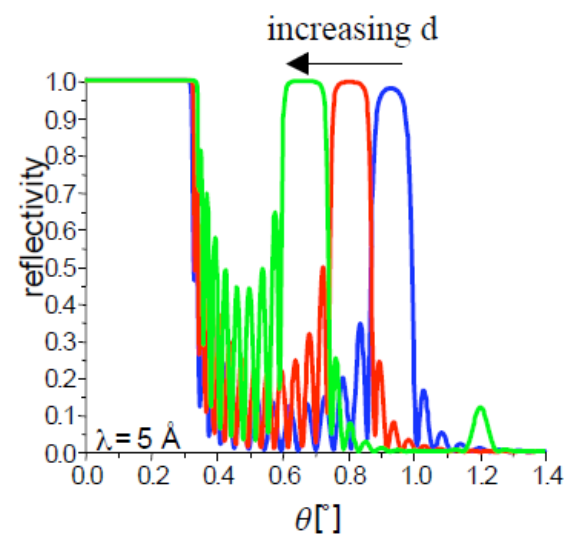
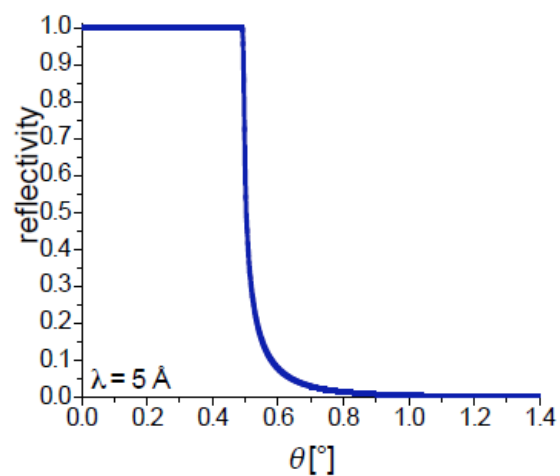
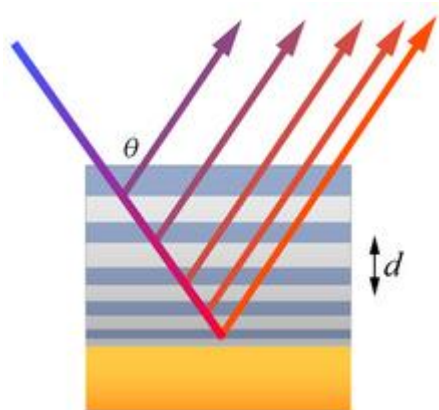
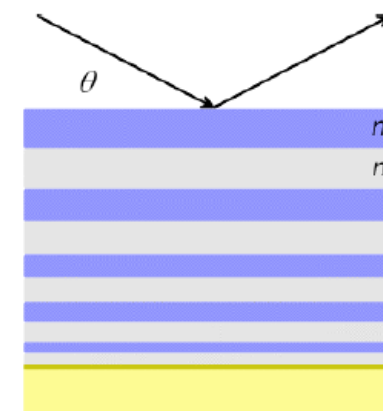
- refractive index $n < 1$
- total external reflection
e.g. Ni $\theta_c = 0.1^\circ/\text{\AA}$

multilayer

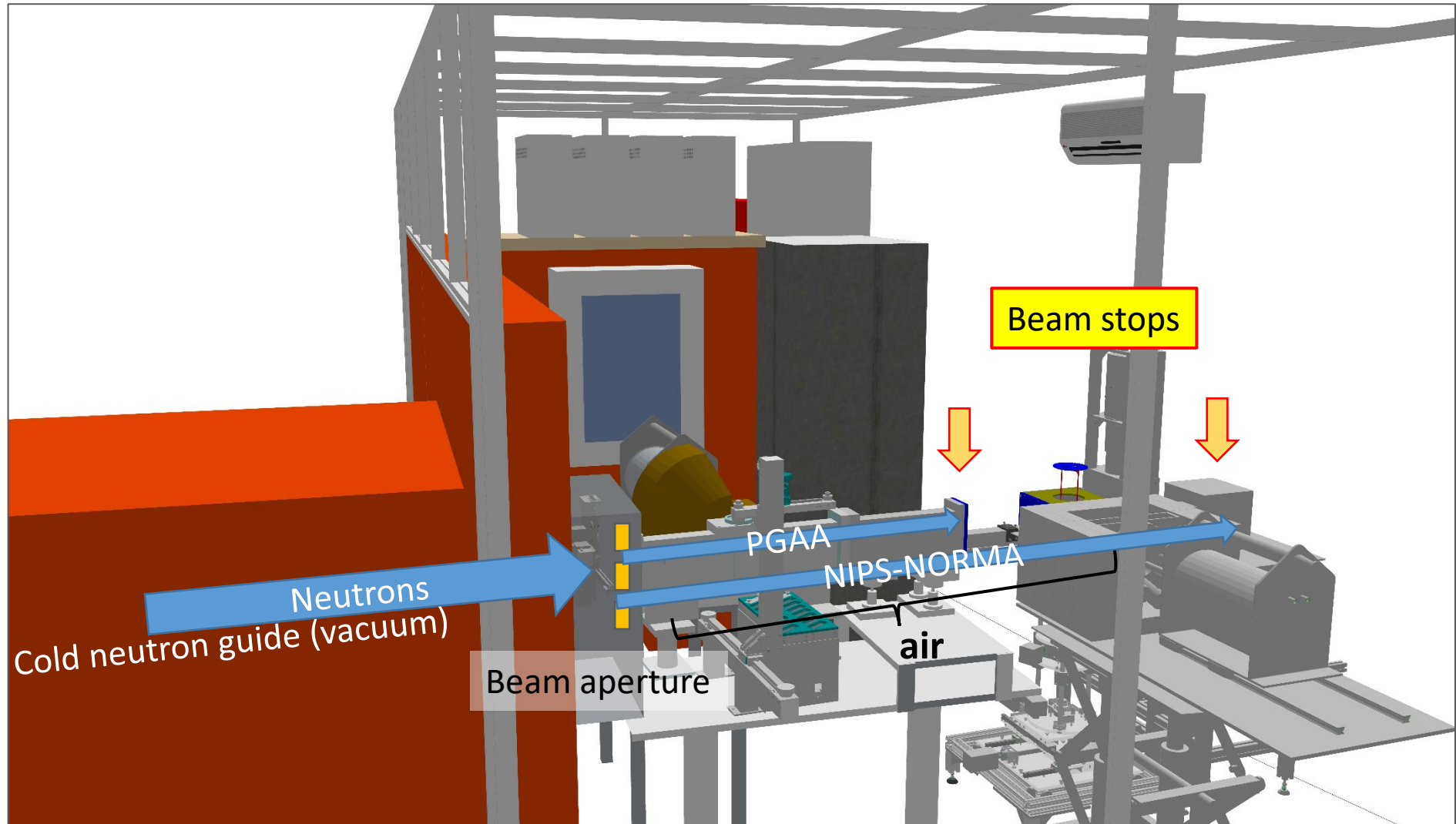


$$\lambda = 2nd \sin \theta$$

supermirror



PGAA and NIPS-NORMA facilities

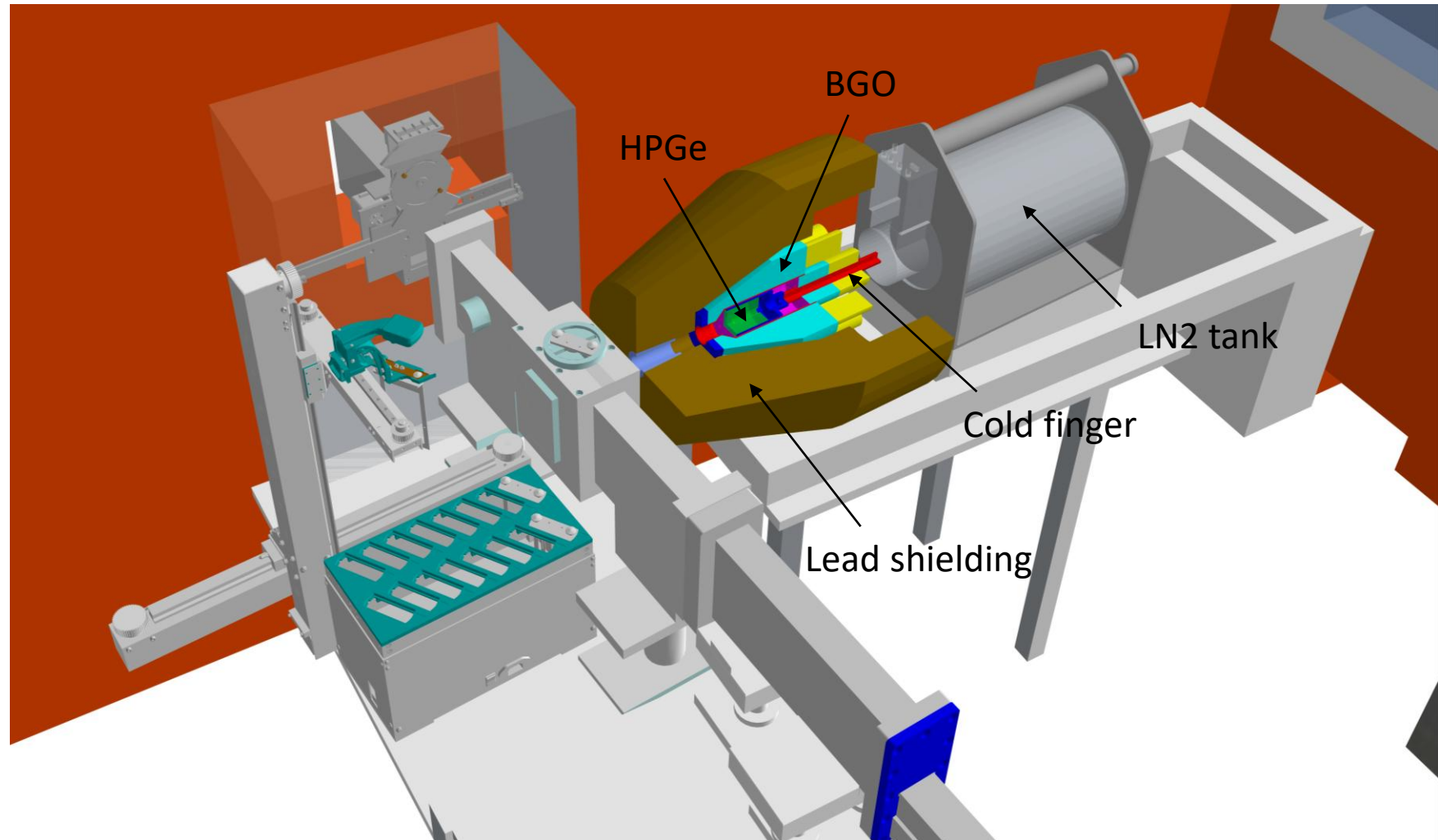


3D model by László Szentmiklósi

The two facilities can be operated independently and simultaneously

PGAA facility

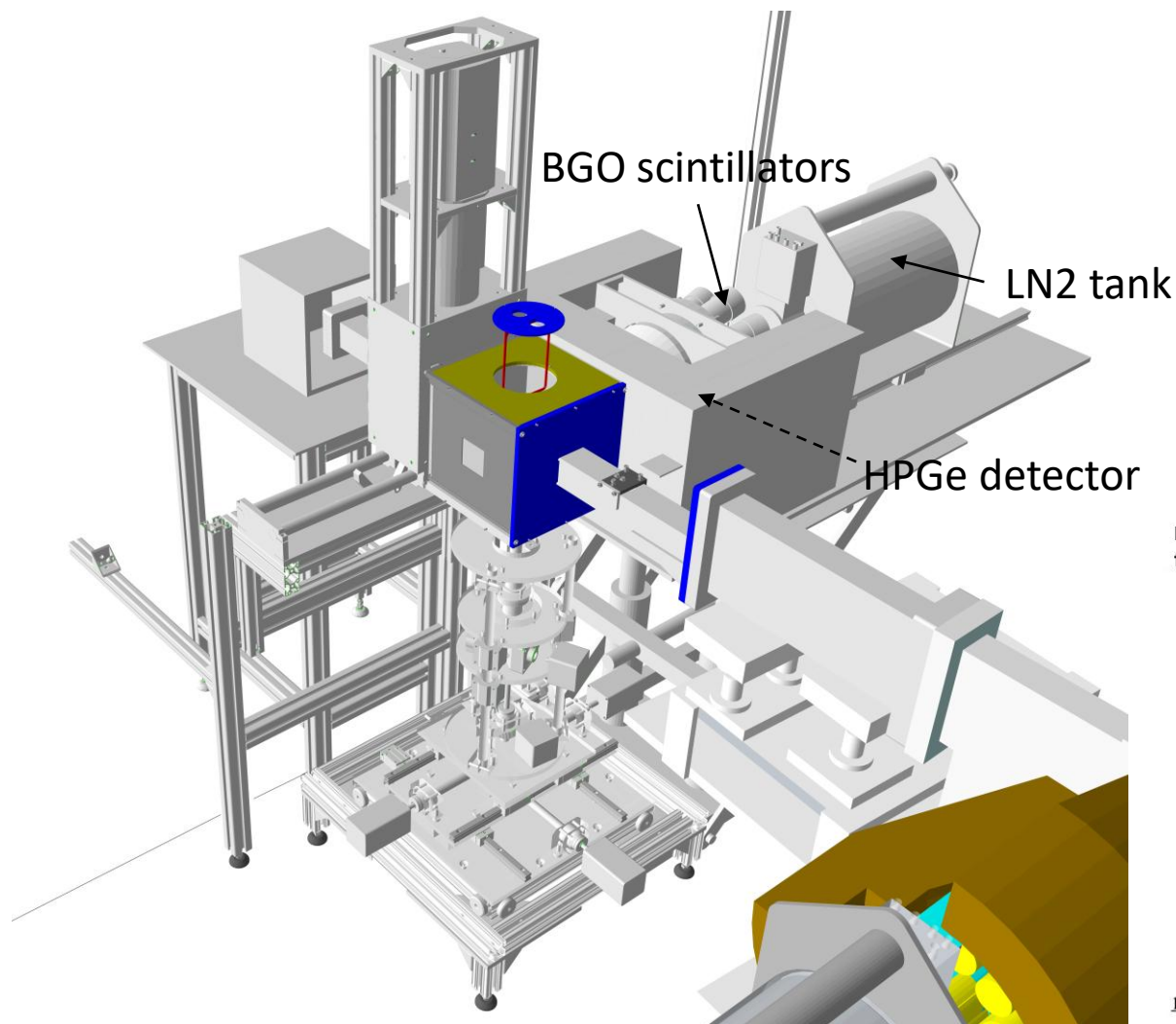
- element analysis of smaller samples (up to ~300 g; smaller than 10×10×8 cm; minimum 0.5 g)
- neutron flux is four times higher than in case of NIPS-NORMA station (PGAA is closer to the neutron guide end)



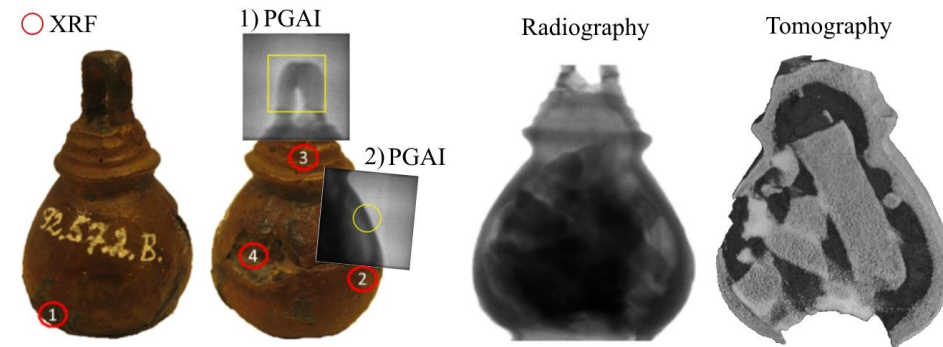
3D model by László Szentmiklósi

NIPS-NORMA facility

- element analysis combined with neutron imaging (optional)
- designed to accommodate larger samples (up to ~5 kg)

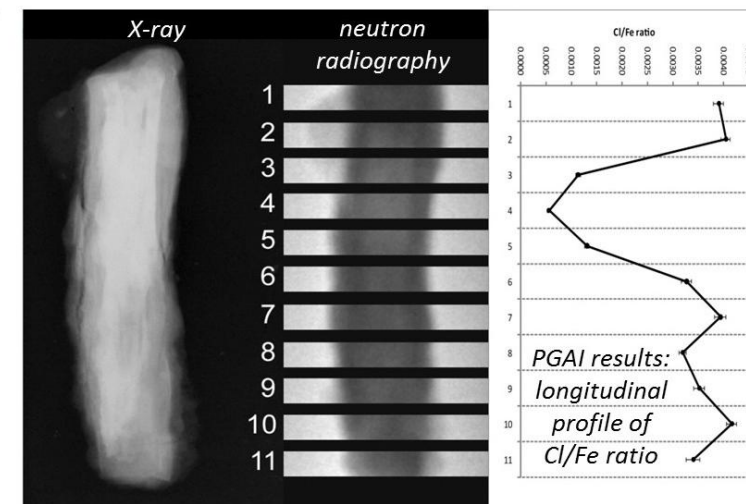


3D model by László Szentmiklósi



Maróti B. 2018 (doi: [10.15476/ELTE.2018.146](https://doi.org/10.15476/ELTE.2018.146))

Iron nail from a Roman fortress site



Watkinson et al. 2014 (doi: [10.1111/arc.12058](https://doi.org/10.1111/arc.12058))



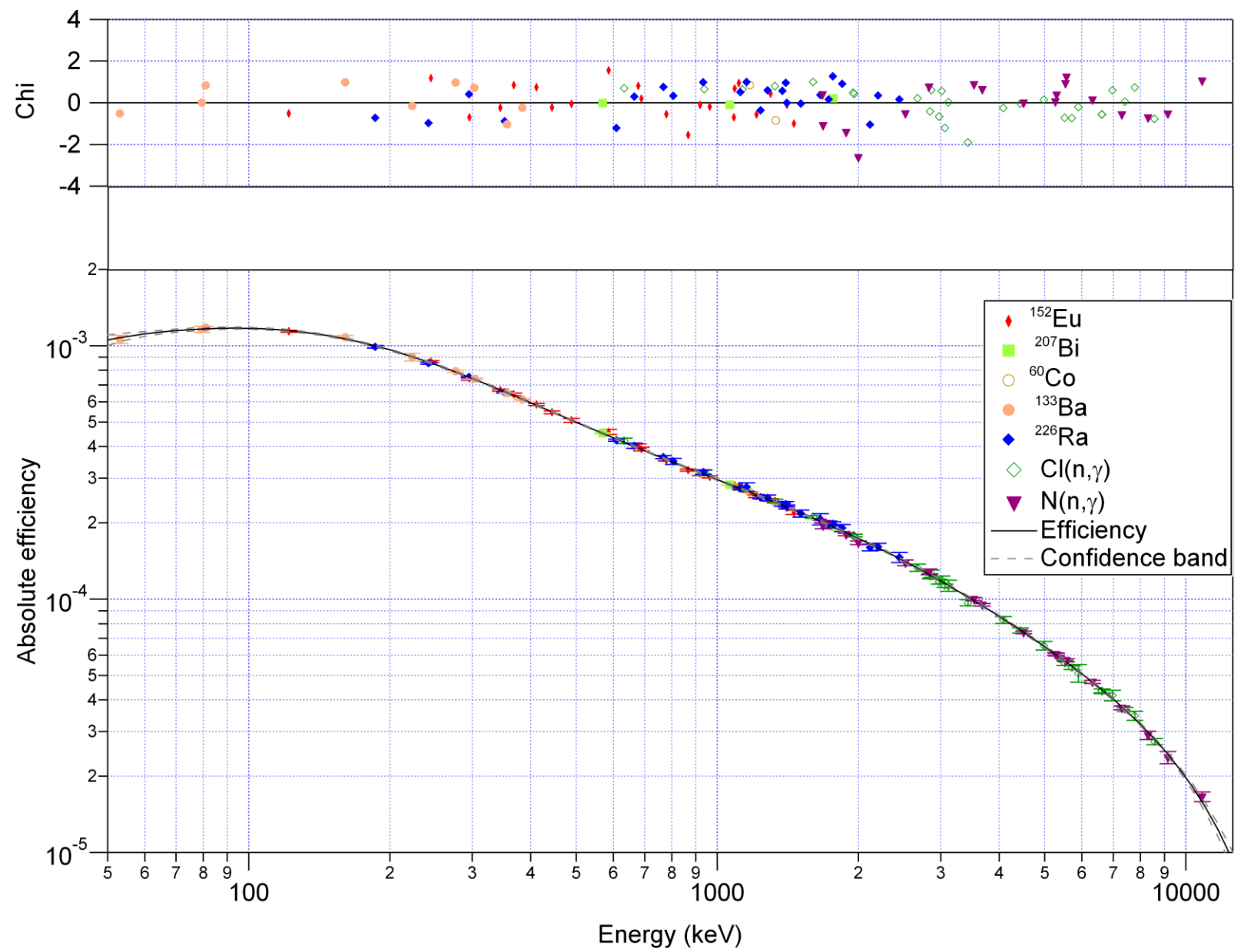
- Scintillators (high count rate)
- high-purity germanium, **HPGe** (high resolution)

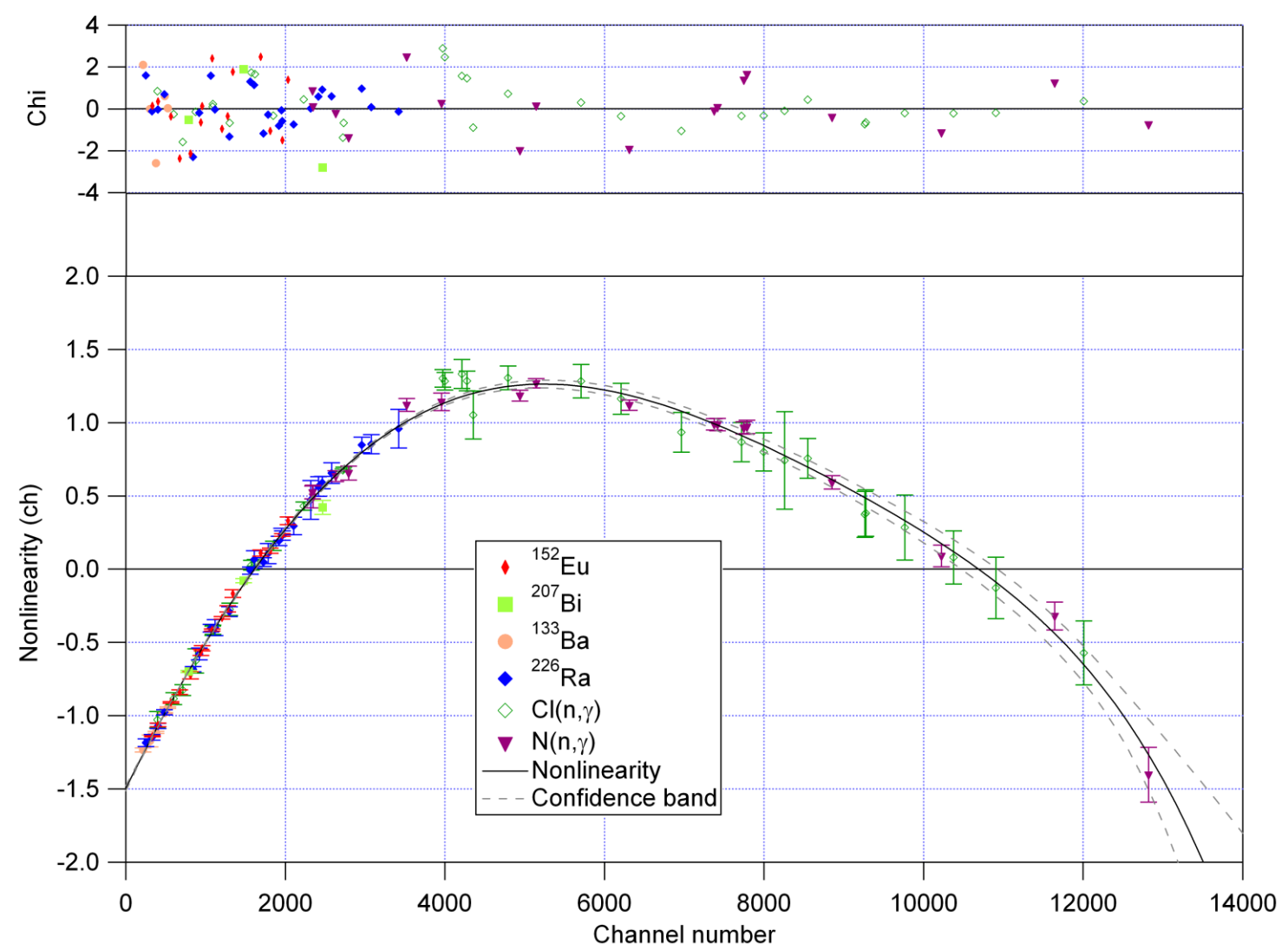
Compton-suppressed



composite detectors







16k (16384) channels
1 channel ~ 0.7 keV



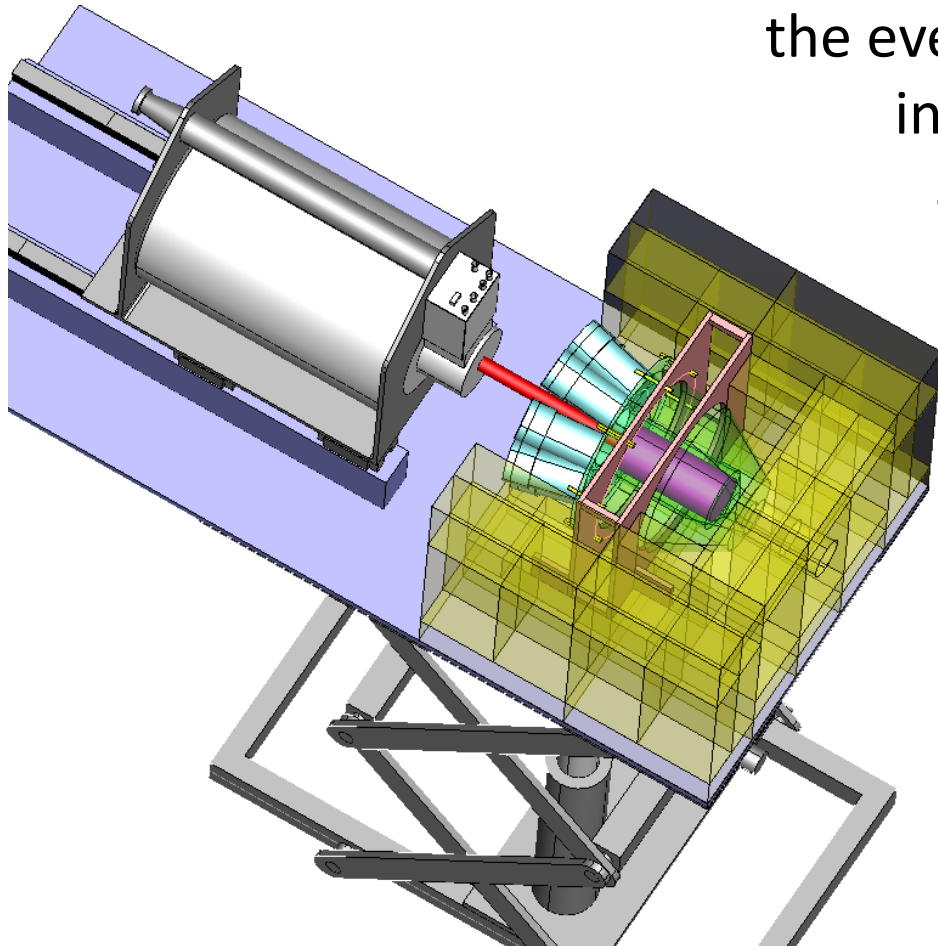
Without proper nonlinearity correction the element identification is not possible



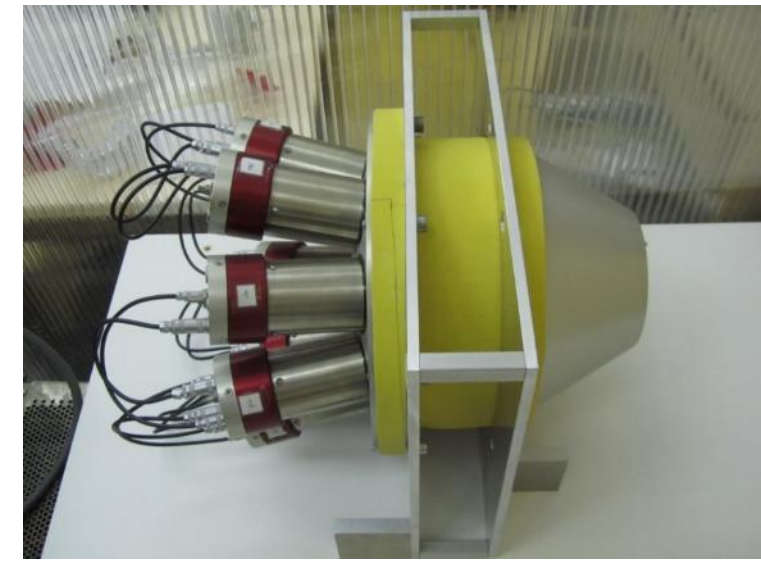
Aim: to reduce the background but not the peak!

i.e. to selectively discard
the events where interaction happened
in both HPGe and suppressor,
and keep all the events interacted
only with HPGe

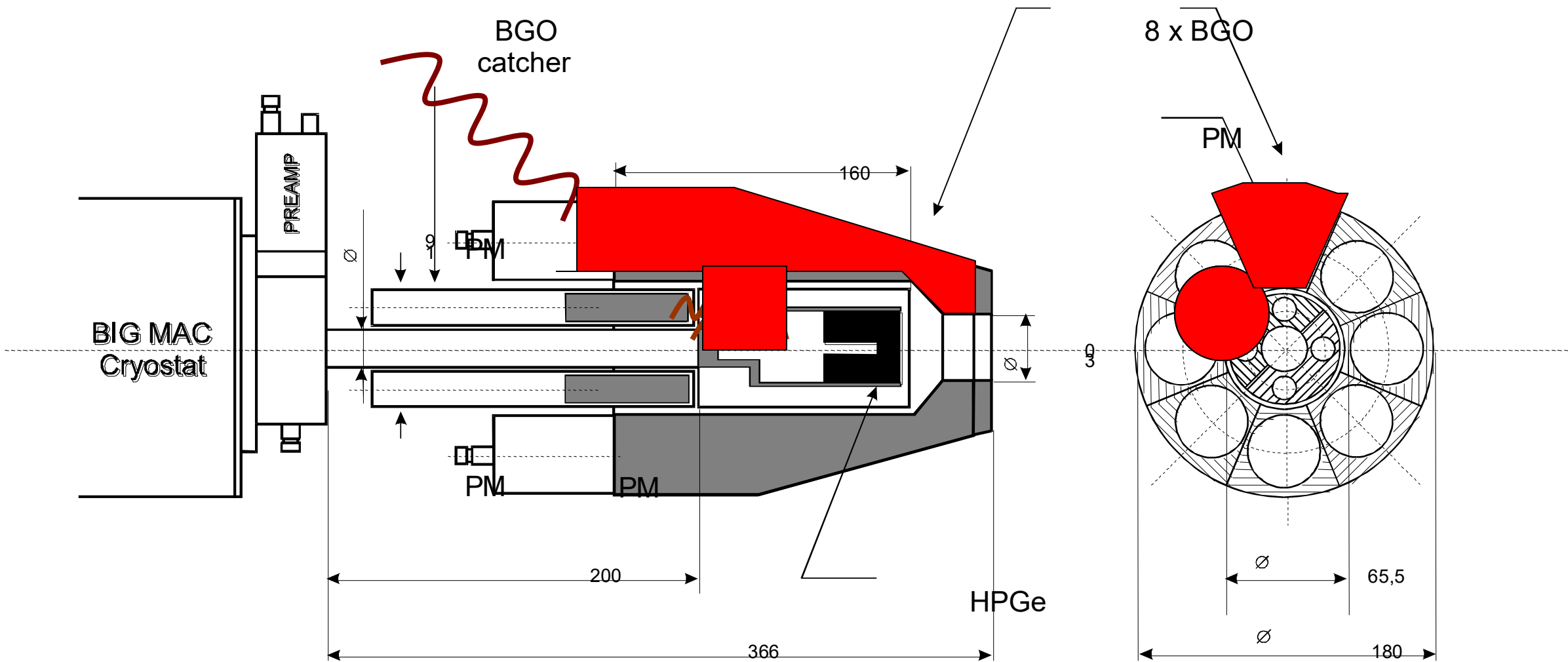
BETTER
SIGNAL-TO-NOISE
RATIO

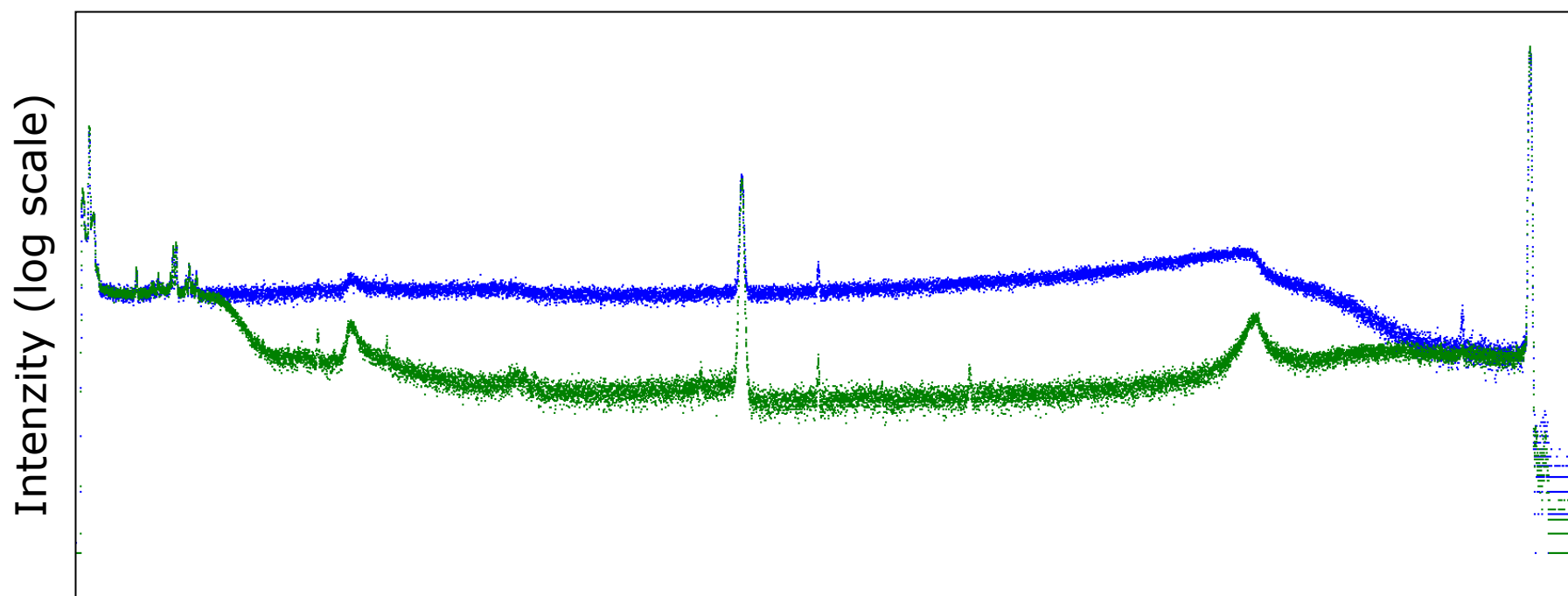


$\text{Bi}_4\text{Ge}_3\text{O}_{12}$ (BGO)



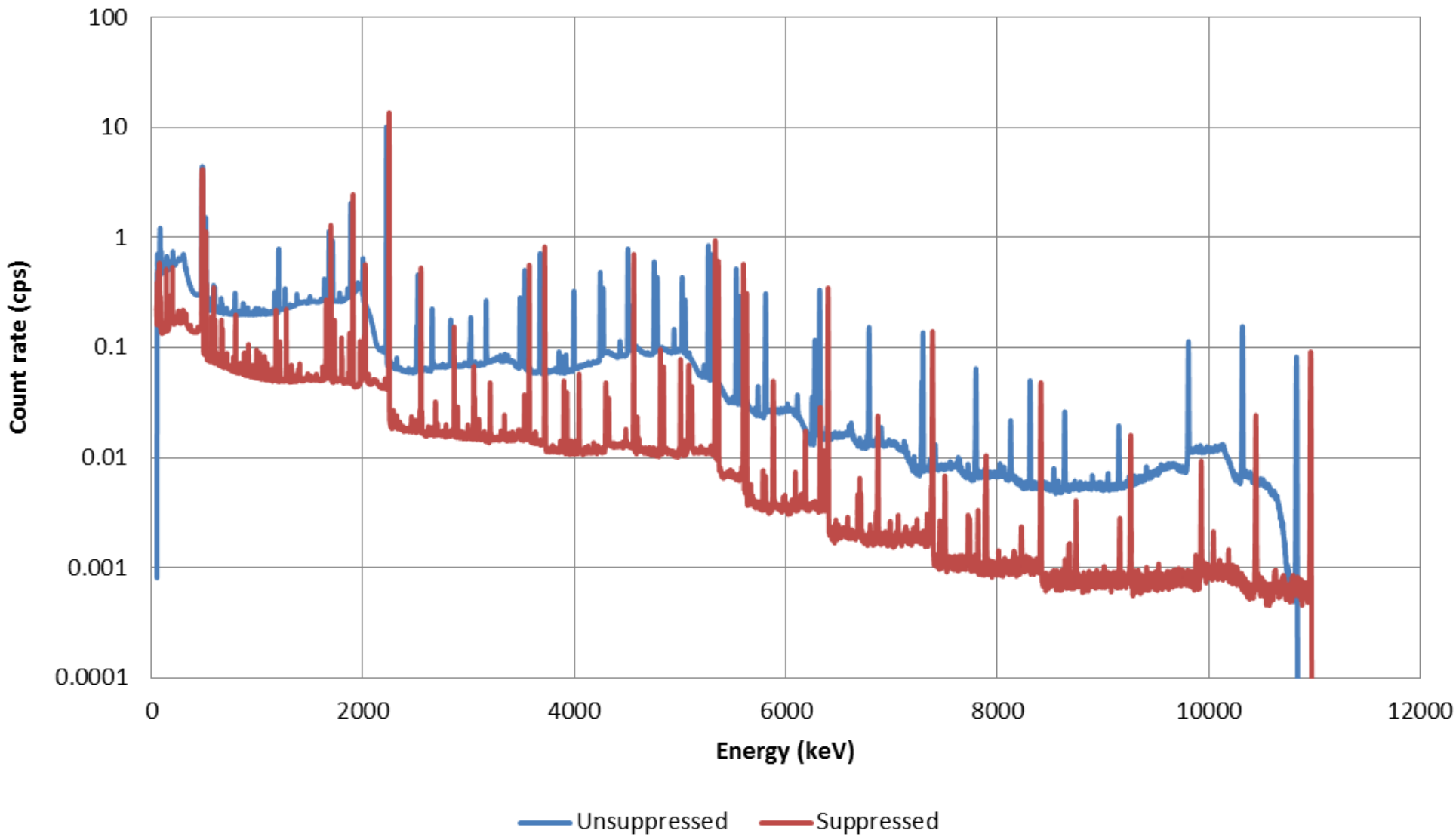
BUDAPEST COMPTON-SUPPRESSED / PAIR-MODE GAMMA SPECTROMETER





Log scale: intensity of full energy peak is the same, while the Compton-plateau is lower

Compton-suppression up to 11 MeV





$$R = \int_{E_{\min}}^{E_{\max}} N \sigma(E) \Phi(E) dE$$

R - number of captured neutron per sec

σ - differential cross section (cm^2),

Φ - flux $\text{cm}^{-2} \text{s}^{-1} \text{eV}^{-1}$

N - number of target nuclides ($\sim$ mass)

$$N = m/M \theta N_A$$

$$R_{th} = N \sigma_0 \Phi_{th}$$

For thermal neutrons



Gamma (prompt and decay) radiation is characteristic

- energy → elements (isotopes)
- intensity → quantity

Peak position

Peak area

Energy

Partial gamma-ray
production cross section

Nucl.	E (keV)	cross-sec	half-life
^1H	2223.249 keV	0.3326 b	
^{23}Na	1368.6 keV	0.500 b	14.96 h



$$A_{\gamma} = m \cdot S \cdot t; \quad S = \frac{N_A}{M} \cdot \underbrace{\theta \cdot \sigma_0 \cdot P_{\gamma}}_{\sigma_{\gamma}} \cdot \phi \cdot \varepsilon(E_{\gamma}) \cdot f(E_{\gamma})$$

Fit
from spectrum

From spectroscopic library

We always measure
concentrations, i.e. ratios
of element masses!

m : Mass of the element

S : Sensitivity

A_g : Peak area

N_A : Avogadro-number

M : Molar weight

θ : Isotopic abundance

σ_0 : Neutron capture cross-section

P_{γ} : Gamma-yield

ϕ : Neutron flux

$\varepsilon(E_{\gamma})$: Detector efficiency

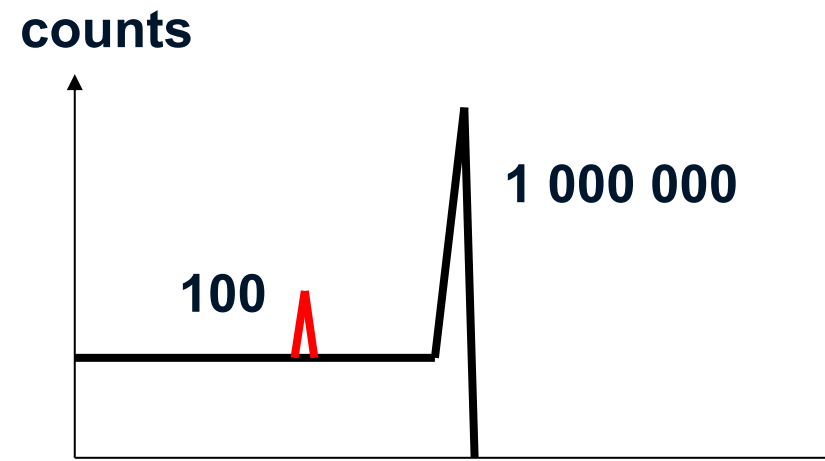


Dynamic range of gamma spectra for any nuclide pairs: 10^6 – 10^8

PGAA

1 mg H together with 1 g Cl
(10 mg water in 1 g CCl_4)

1 mg Cl together with 1 g H
(1 mg Cl in 10 g water)



NAA

it can be increased with repeated
counting after cooling, contact
counting etc.

H 2223 keV $\sigma_\gamma = 0.3326(7)$ barn

Cl 517 keV $\sigma_\gamma = 7.43(7)$ barn

C 1262 keV $\sigma_\gamma = 0.00123(3)$ barn

O 871 keV $\sigma_\gamma = 0.000193(7)$ barn



Pneumatic rabbit system

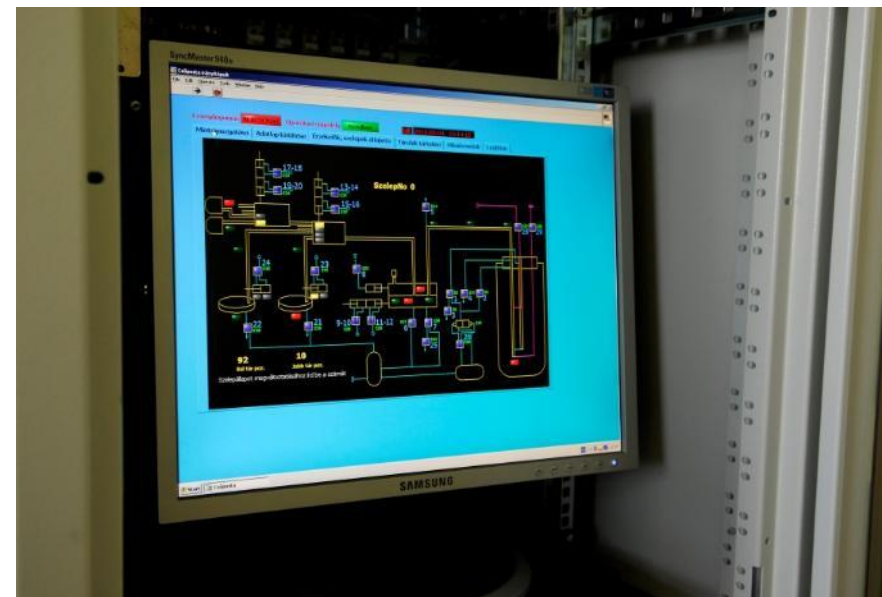
$$\Phi_{\text{th}} = 4.45 \cdot 10^{13} \text{ n/cm}^2\text{s}$$

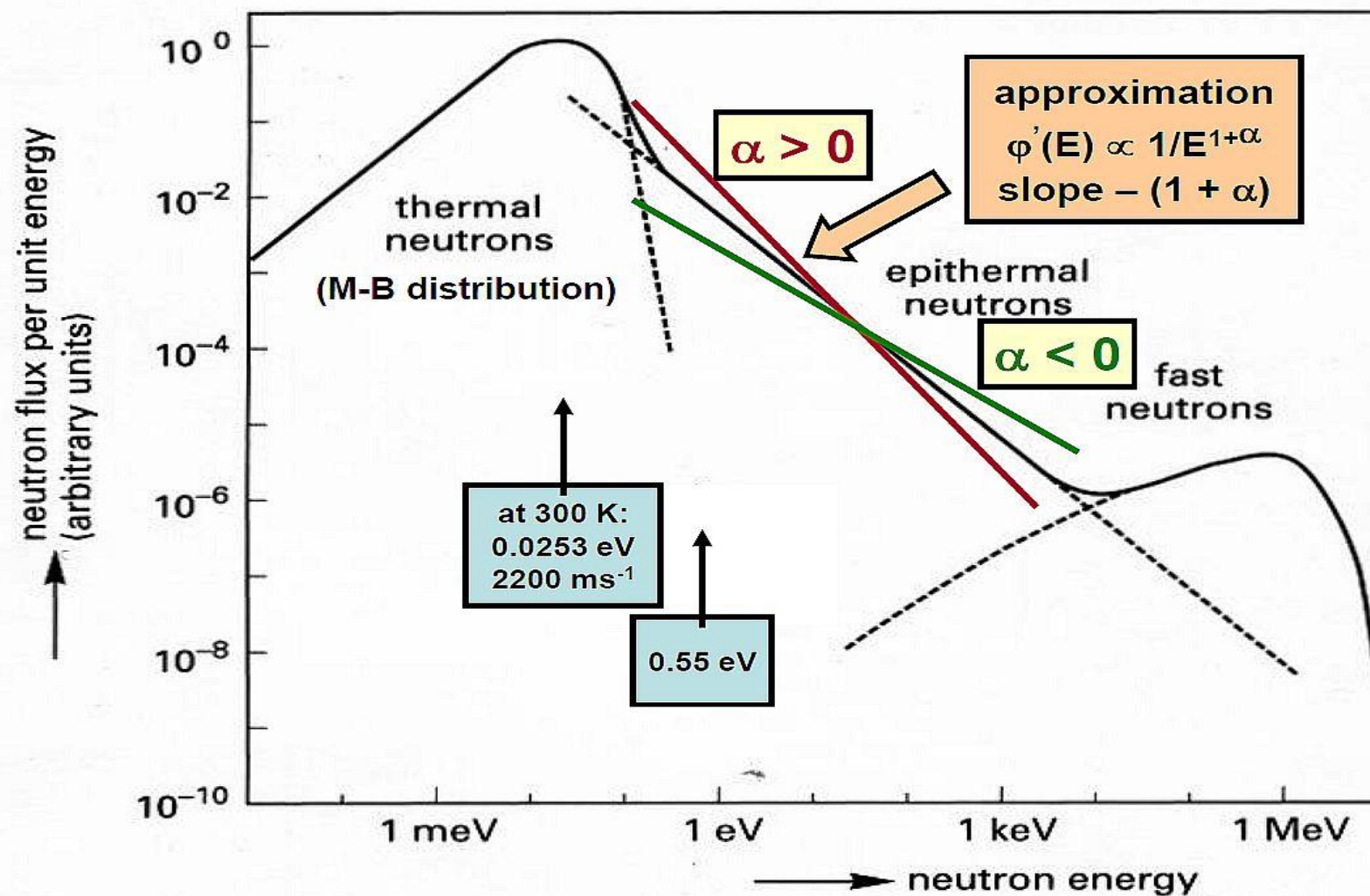
$$\Phi_{\text{epi}} = 1.28 \cdot 10^{12} \text{ n/cm}^2\text{s}$$

$$f = \Phi_{\text{th}}/\Phi_{\text{epi}} = 34.8, \alpha = 0.029$$

Measured isotopes:

^{24}Na , ^{27}Mg , ^{28}Al , ^{38}Cl , ^{49}Ca , ^{51}Ti , ^{52}V ,
 ^{56}Mn , ^{66}Cu , ^{80}Br , $^{87\text{m}}\text{Sr}$, $^{123\text{m}}\text{Sn}$, $^{125\text{m}}\text{Sn}$,
 ^{128}I , ^{139}Ba







BRR No. 17 rotating channel, ($\varnothing=54$ mm)

$$\Phi_{\text{th}} = 1.76 \cdot 10^{13} \text{ n/cm}^2\text{s}$$

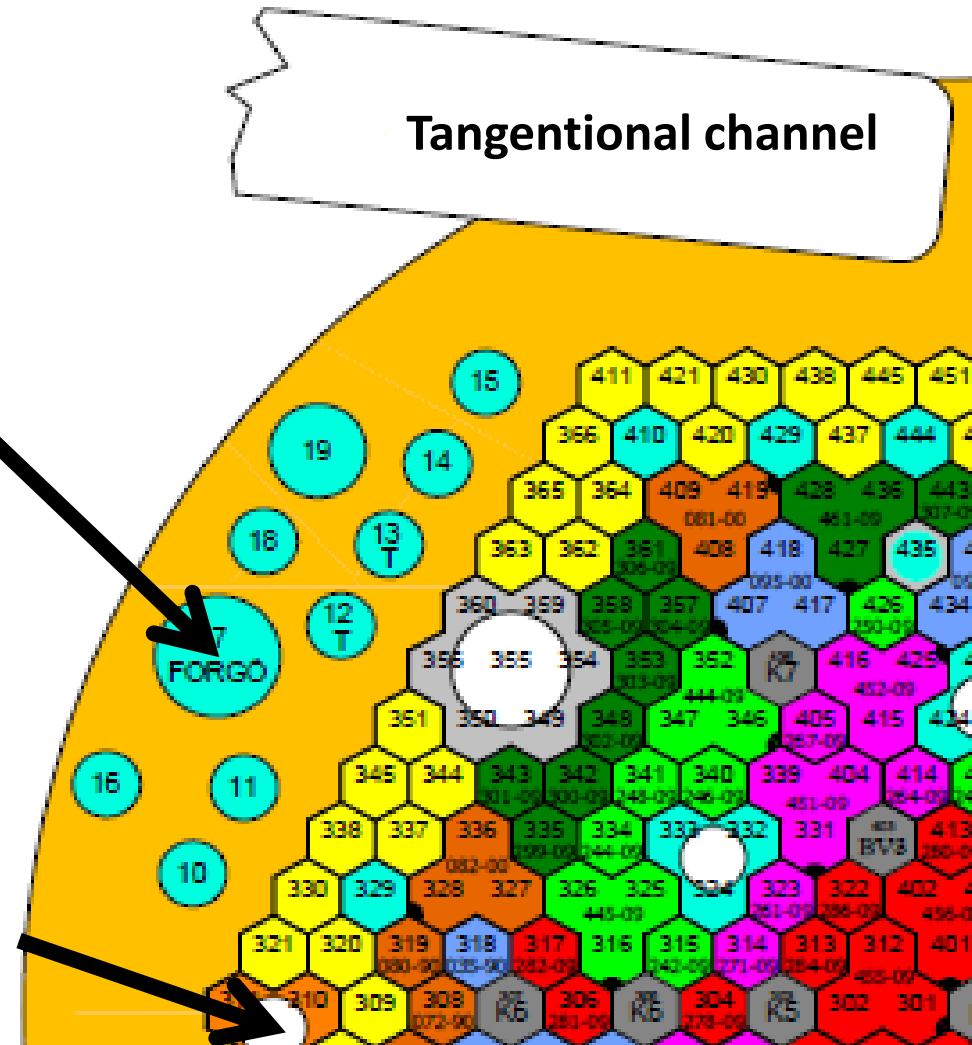
$$f = \Phi_{\text{th}} / \Phi_{\text{epi}} = 47.1$$

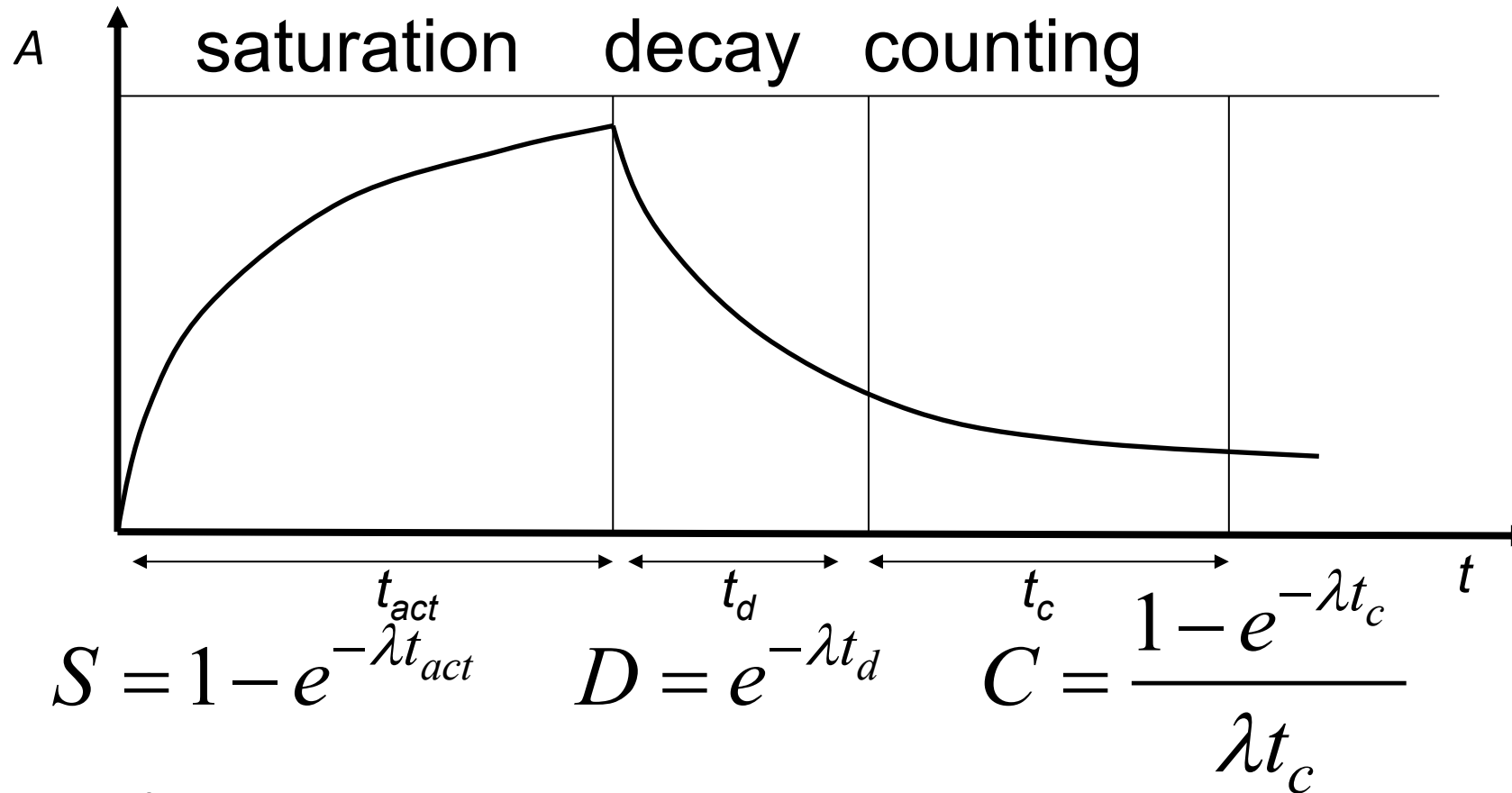
$$\alpha = 0.0249$$

Measured isotopes:

^{24}Na , ^{42}K , ^{51}Cr , ^{59}Fe , ^{60}Co ,
 $^{58}\text{Co}(\text{Ni})$, ^{65}Zn , $^{69\text{m}}\text{Zn}$,
 ^{72}Ga , ^{76}As , ^{82}Br , ^{86}Rb ,
 ^{122}Sb , $^{110\text{m}}\text{Ag}$, ^{124}Sb ,
 ^{134}Cs , ^{140}La , ^{153}Sm , ^{182}Ta ,
 ^{187}W

Pneumatic rabbit system





$\lambda = \ln(2)/T_{1/2}$, decay constant

Peak area has to be corrected for saturation: $A' = A/SDC$



- 4-8 tons
- 15—20 cm pre-war iron (ruins of the Elisabeth-bridge)
- Reduces room background by 2 order of magnitude





$$\frac{N_p/t_m}{(N_p/t_m)^*} = \frac{w}{w^*} \cdot \frac{S \cdot D \cdot C}{S^* \cdot D^* \cdot C^*} \cdot \frac{M^*}{M} \cdot \frac{\theta}{\theta^*} \cdot \frac{\gamma}{\gamma^*} \cdot \frac{\sigma_0}{\sigma_0^*} \cdot \frac{f+Q_0}{f+Q_0^*} \cdot \frac{\varepsilon_p}{\varepsilon_p^*}$$

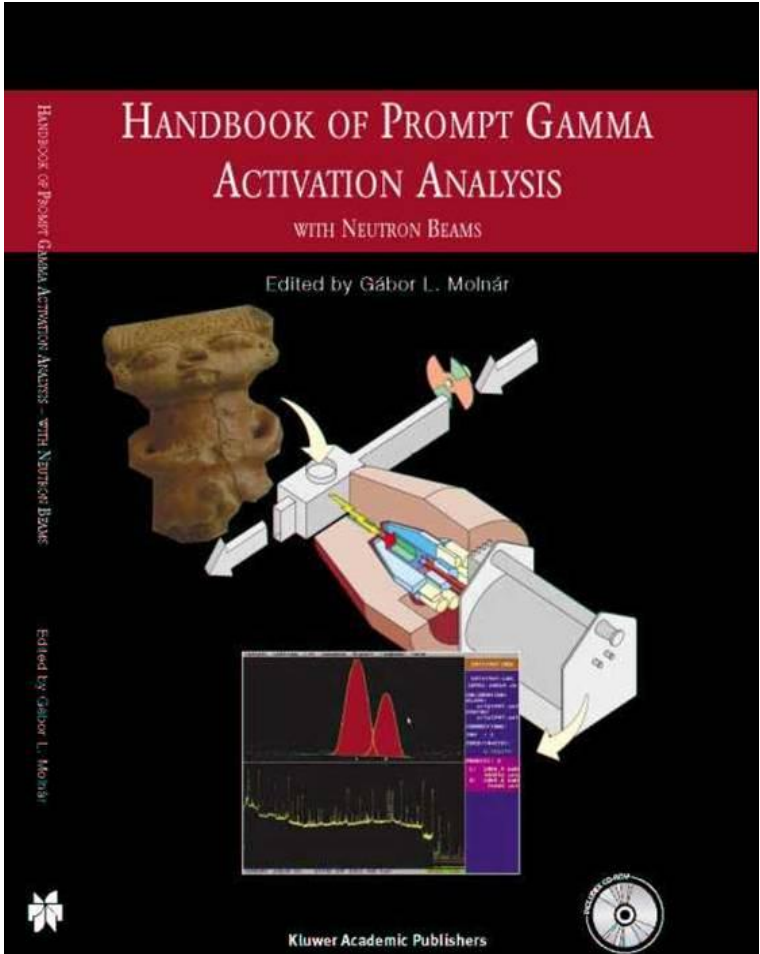
k_0 : compound nuclear constant

$$c_x(ppm) = \frac{\left[\frac{N_{p,x}}{t_m \cdot S \cdot D \cdot C \cdot W} \right]}{A_{sp,Au}} \cdot \frac{1}{k_{0,Au}(x)} \cdot \frac{f+Q_{0,Au}(\alpha)}{f+Q_{0,x}(\alpha)} \cdot \frac{\varepsilon_{p,Au}}{\varepsilon_{p,x}} \cdot 10^6$$

$$f = \frac{\Phi_{th}}{\Phi_e}, \quad Q_0 = \frac{I_0}{\sigma_{th}}, \quad A_{sp,x} = N_{p,x}/t_m \cdot S \cdot D \cdot C \cdot W$$

α : epitermal shape factor

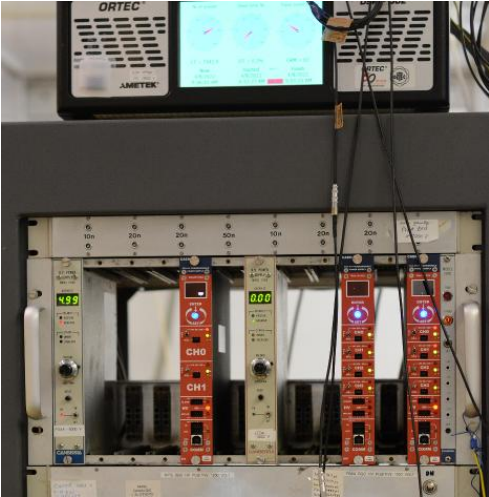
Z	EI	A	MW	#	E	dE	σ	d σ %	RI	Area	cps/g
1	H	1	1.01	1	2223.259	0.019	0.3326	0.2	100.00	100.00	64.183
1	H	2	1.01	2	6250.204	0.098	0.000492	5.0	0.15	5.00	0.0286
3	Li	6	6.94	5	477.586	0.050	0.001399	5.9	3.52	10.14	0.1218
3	Li	7	6.94	2	980.559	0.046	0.004365	5.1	10.97	18.74	0.2251
3	Li	7	6.94	3	1051.817	0.048	0.004364	5.1	10.97	17.83	0.2141
3	Li	7	6.94	1	2032.310	0.070	0.0398	5.0	100.00	100.00	1.2007
3	Li	6	6.94	6	6769.633	0.263	0.001354	6.5	3.40	0.84	0.0101
3	Li	6	6.94	4	7246.800	0.275	0.002106	8.4	5.29	1.17	0.014
4	Be	9	9.01	4	853.631	0.011	0.00165	8.9	26.69	100.00	0.0723
4	Be	9	9.01	3	2590.014	0.025	0.00188	8.9	30.41	49.08	0.0355
4	Be	9	9.01	2	3367.484	0.035	0.002924	8.9	47.30	58.96	0.0427
4	Be	9	9.01	5	3443.421	0.036	0.000993	8.9	16.06	19.54	0.0141
4	Be	9	9.01	6	5956.602	0.092	0.000146	9.1	2.36	1.41	0.001
4	Be	9	9.01	1	6809.579	0.099	0.006181	9.0	100.00	48.52	0.0351
5	B	10	10.81	1	477.600	5.000	712.5	0.3	100.00	100.00	39806
6	C	12	12.01	2	1261.708	0.057	0.00123	2.7	45.58	100.00	0.0306
6	C	12	12.01	3	3684.016	0.069	0.001175	3.5	43.53	38.02	0.0116
6	C	12	12.01	1	4945.302	0.066	0.002699	2.9	100.00	60.55	0.0186
7	N	14	14.01	22	583.567	0.031	0.000429	3.3	1.81	6.93	0.0159
7	N	14	14.01	12	1678.244	0.029	0.006254	1.5	26.34	47.15	0.1085
7	N	14	14.01	18	1681.174	0.043	0.001296	2.7	5.46	9.76	0.0225
7	N	14	14.01	21	1853.944	0.052	0.000474	4.5	2.00	3.31	0.0076
7	N	14	14.01	5	1884.853	0.031	0.0145	1.3	61.07	100.00	0.2301
7	N	14	14.01	24	1988.532	0.077	0.000294	5.8	1.24	1.94	0.0045
7	N	14	14.01	15	1999.693	0.032	0.003208	1.7	13.51	21.12	0.0486
7	N	14	14.01	13	2520.446	0.039	0.004246	1.8	17.88	22.98	0.0529



<https://www-nds.iaea.org/pgaa/databases.htm>



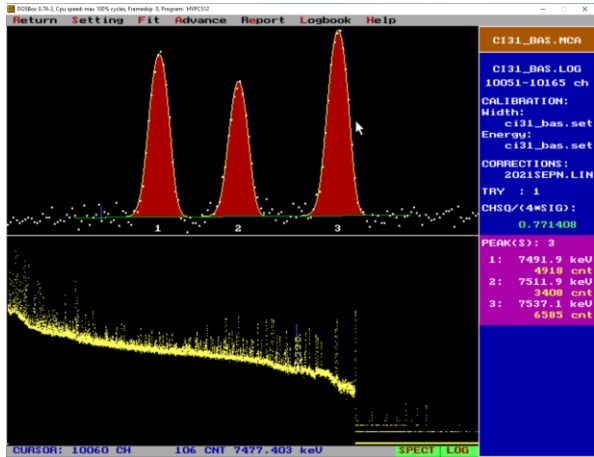
Photo of the PGAA facility



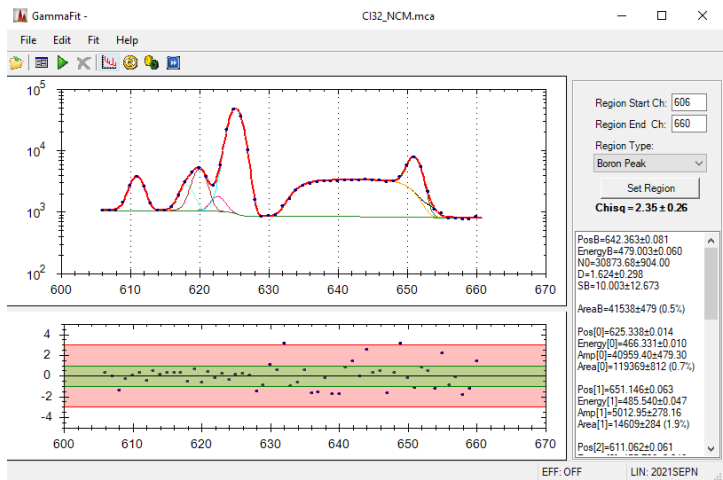
Ortec DSPEC 502A gamma ray spectrometer



The experiments took place at the PGAA facility of the Budapest Neutron Centre. An Ortec DSPEC 502 spectrometer was used for data acquisition. The spectrum evaluation was done with Hypermet PC software. The difficult regions and special shaped peaks were fitted with our inhouse software GammaFit.



Hypermet PC

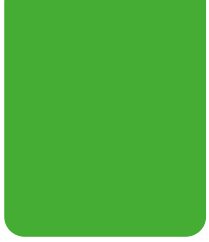


GammaFit (by L. Szentmiklósi)

The element identification and concentration calculation was done based on the formulae in the ProSpeRo software developed by Zsolt Révay.

Principle of evaluation: by fitting mathematical shape functions to the spectrum, overlapping peaks can also be evaluated (not by integration!)

Z	El	MW	E	dE	A	sigma	stat u%	tot u%	RelInt	RelArea	No.	decay	T1/2 (unc)	2020.07.22
1	H	1.008	2223.259	0.019	1	0.3326	0.20	0.20	100.00	100	1	P2002		standard
2	D	2.014	6250.243	0.003	2	0.000515	3.00	3.00	100.00	100	1			updated 2022.03
3	Li	6.941	477.586	0.05	6	0.00129	2.60	2.60	3.30	7	4			updated 2022.03
3	Li	6.941	980.559	0.046	7	0.00422	2.70	2.70	10.84	16.3	2			updated 2022.03
3	Li	6.941	1051.817	0.048	7	0.00426	2.00	2.00	10.95	15.9	3			updated 2022.03
3	Li	6.941	2032.31	0.07	7	0.0389	2.10	2.10	100.00	100	1			updated 2022.03
3	Li	6.941	6769.633	0.263	6	0.00124	2.30	2.30	3.20	1.1	6			updated 2022.03
3	Li	6.941	7246.8	0.275	6	0.00193	2.10	2.10	5.00	1.58	5			updated 2022.03
4	Be	9.012	853.631	0.011	9	0.00172	2.00	3.50	36.30	100	1			updated 2022.03

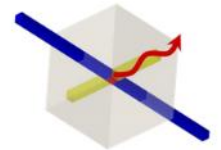
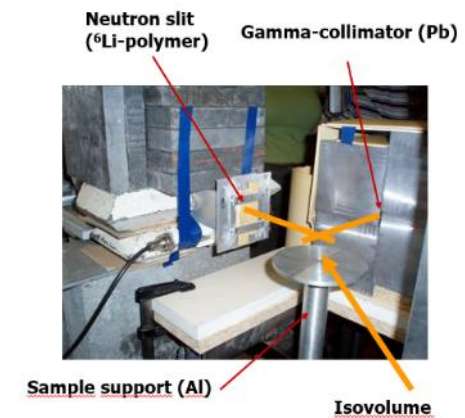


PGAA in practice



- Tomographic principle (PGI, PGA-CT, NT)
 - Detecting integrated information along the path of the neutrons, repeating this at several points and angles
 - Mathematical reconstruction is needed
 - The whole sample has to be measured
- Isovolume approach (PGAI)
 - Collimation: the source of analytical information is the so-called isovolume
 - Scanning the sample, recording localized data
 - Signal vs. position instantly gives the raw result
 - Pointwise correction for neutron self shielding and gamma self attenuation is required (Monte Carlo)
 - Measurement of the whole sample is not a must

The isovolume concept

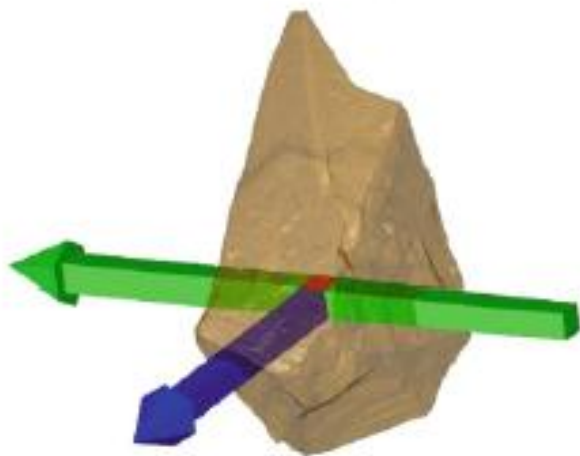




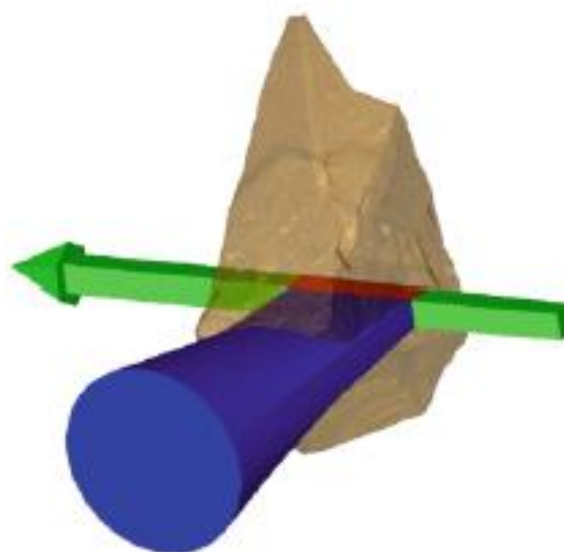
Archaeometry



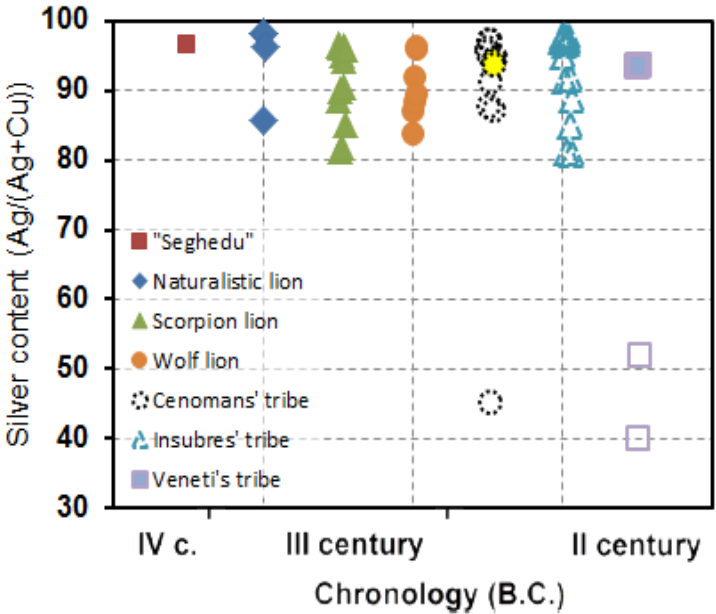
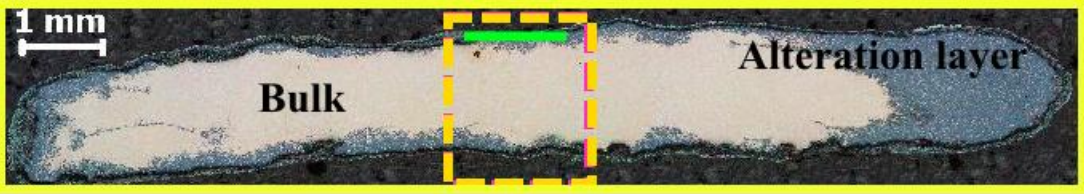
a) Isovolume geometry:
Gamma collimator size < Sample thickness



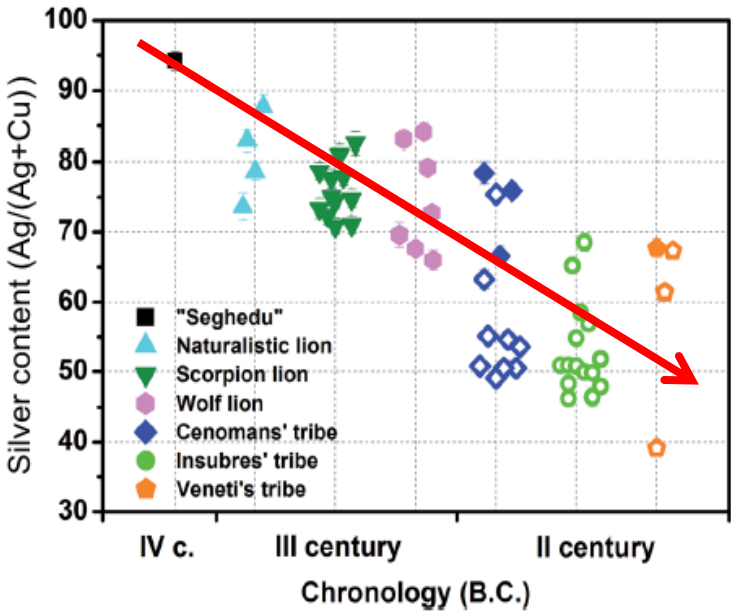
b) Chord geometry:
Gamma collimator size > Sample thickness



Presentation by
Veronika Szilágyi this
afternoon

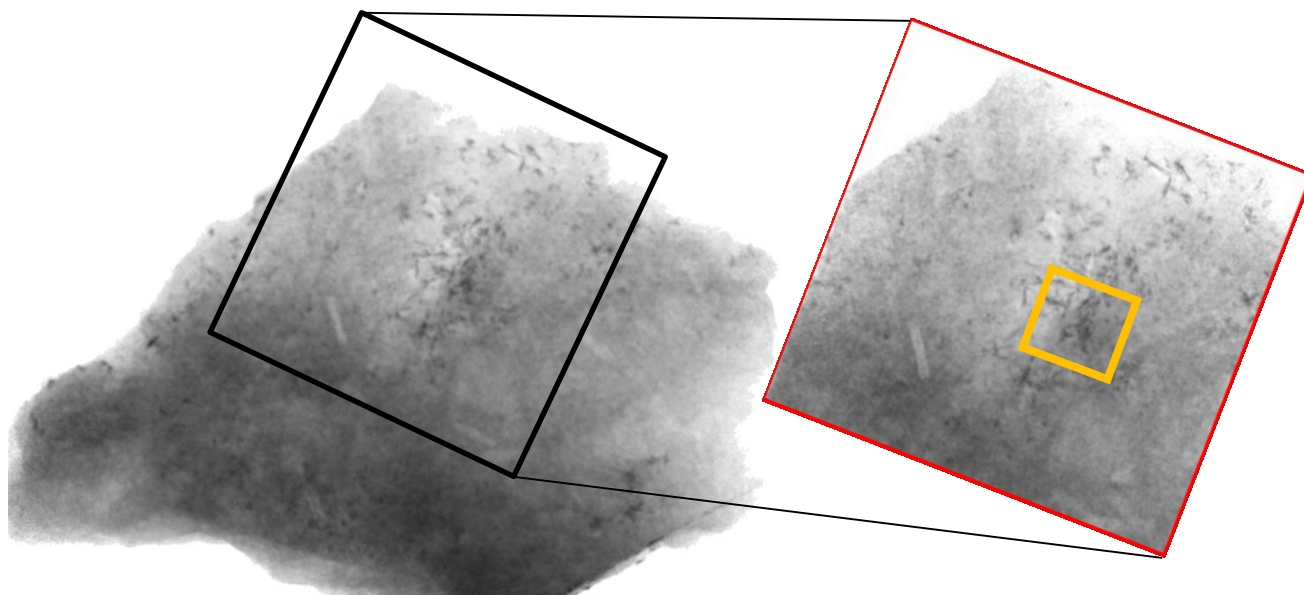


Handheld XRF results



PGAA results

- silver content determines the value of coins
- PGAA results well illustrate the process of devaluation (Punic wars)



Average boron composition

- Mantle: **<1 ppm**
- Volcanic rocks: **1-35 ppm**
- Subduction ocean plate and sediment: **80-120 ppm**

Gméling et al. J. Volcanol. Geotherm. Res. **159** (2007) 70-84

PGAA – Average boron content

1. Cross section of neutron beam 20×20 mm

160 ppm B

2. Cross section of neutron beam 5×5 mm

630 ppm B



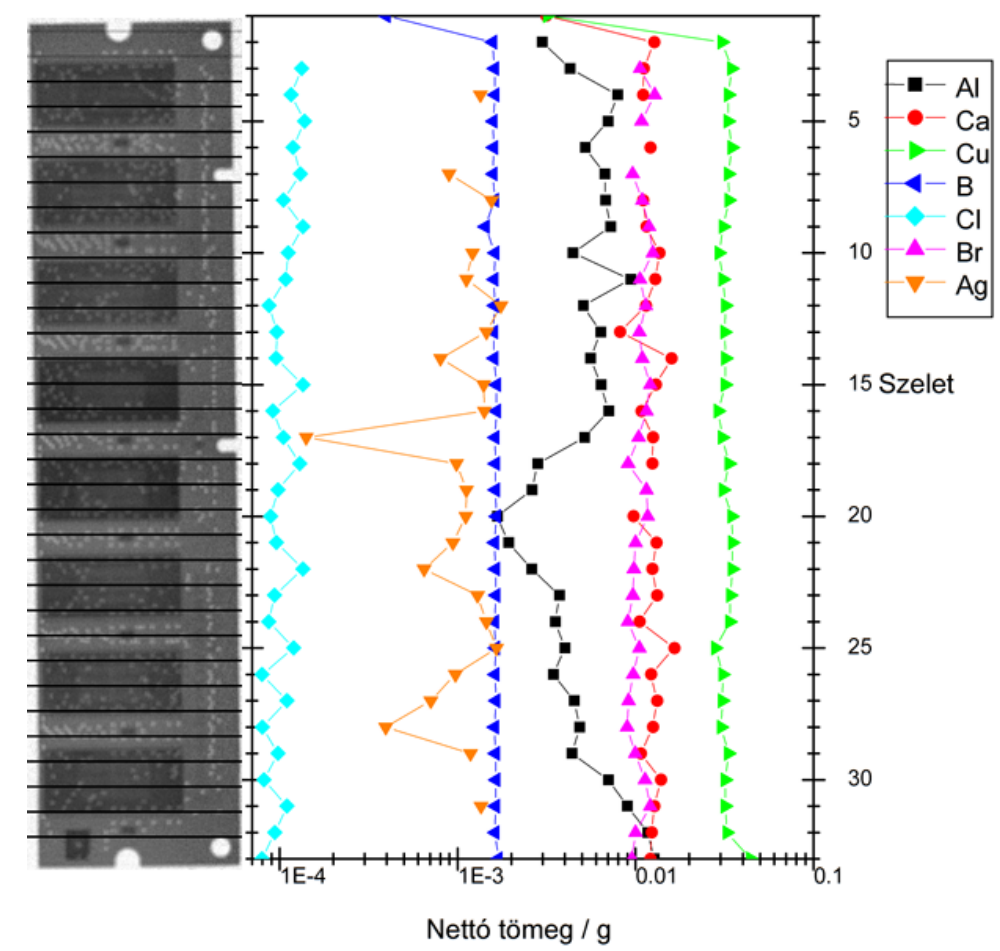
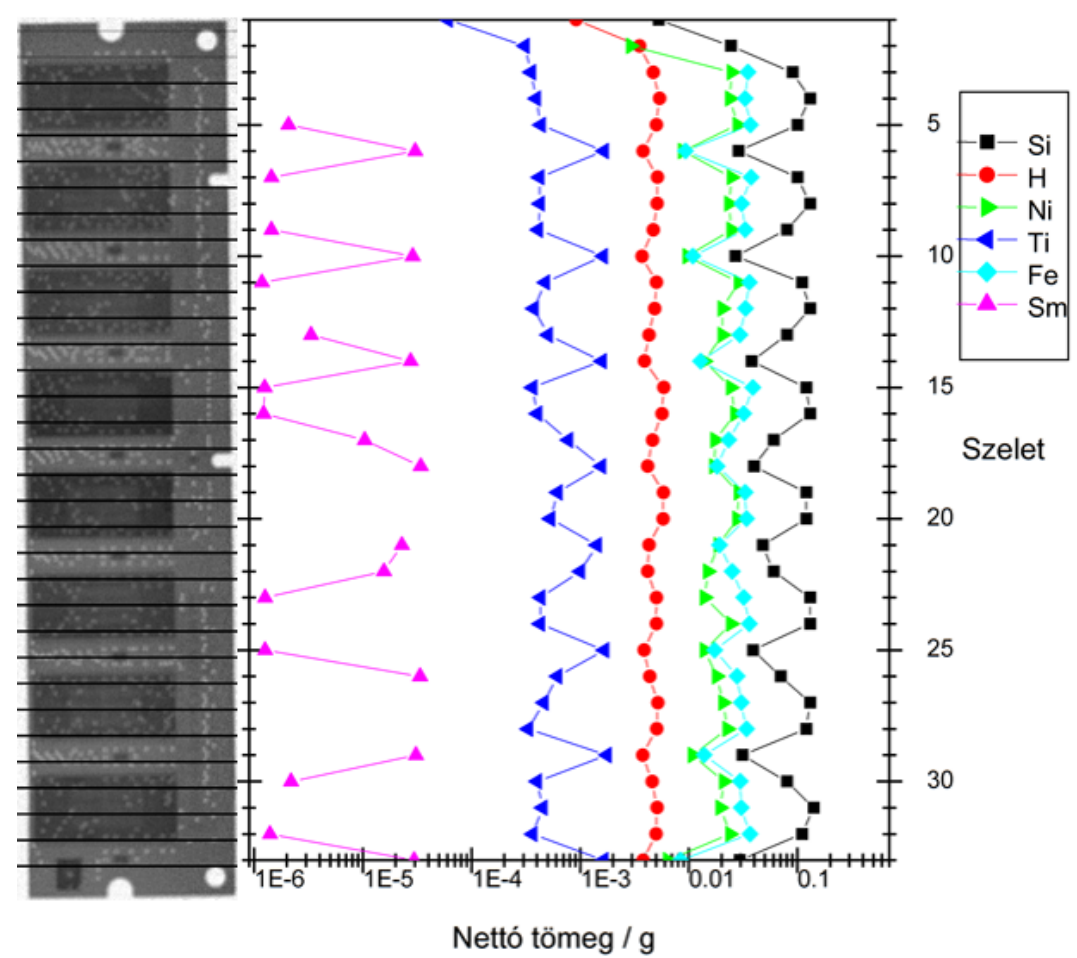
Boron

highly neutron absorbing

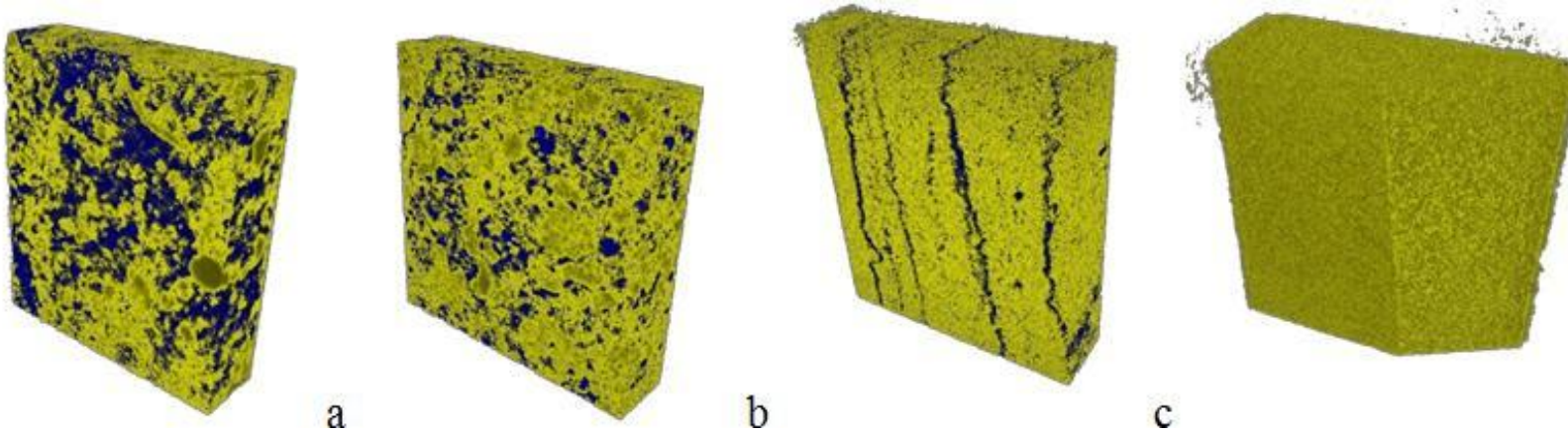
→ neutron transmission is lower

→ darker pixels

Szakáll S., Kristály F., Kasztovszky Zs., Maróti B.: Vulkáni eredetű borátásvány a Cserhátból, VI. Kőzettani és Geokémiai Vándorgyűlés Ópálos, 2015. szeptember 10-12.



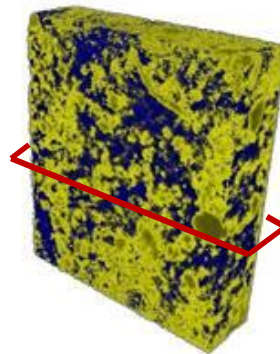
Next presentation by
Noémi Buczkó



- a) Vicenza limestone
- b) Trachite
- c) Istria limestone
- d) Carrara marble

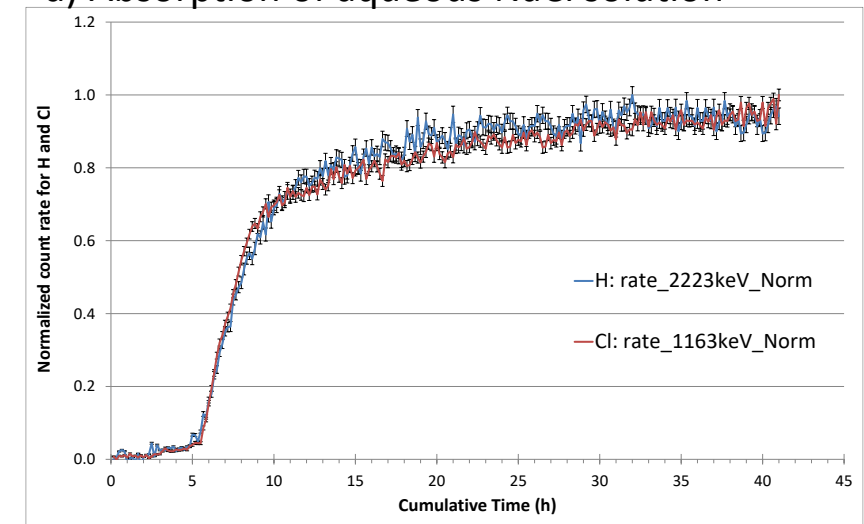
Cross section of neutron beam:
2x43 mm (V×H)

Position of the measurement:
48 mm from the bottom of the stone



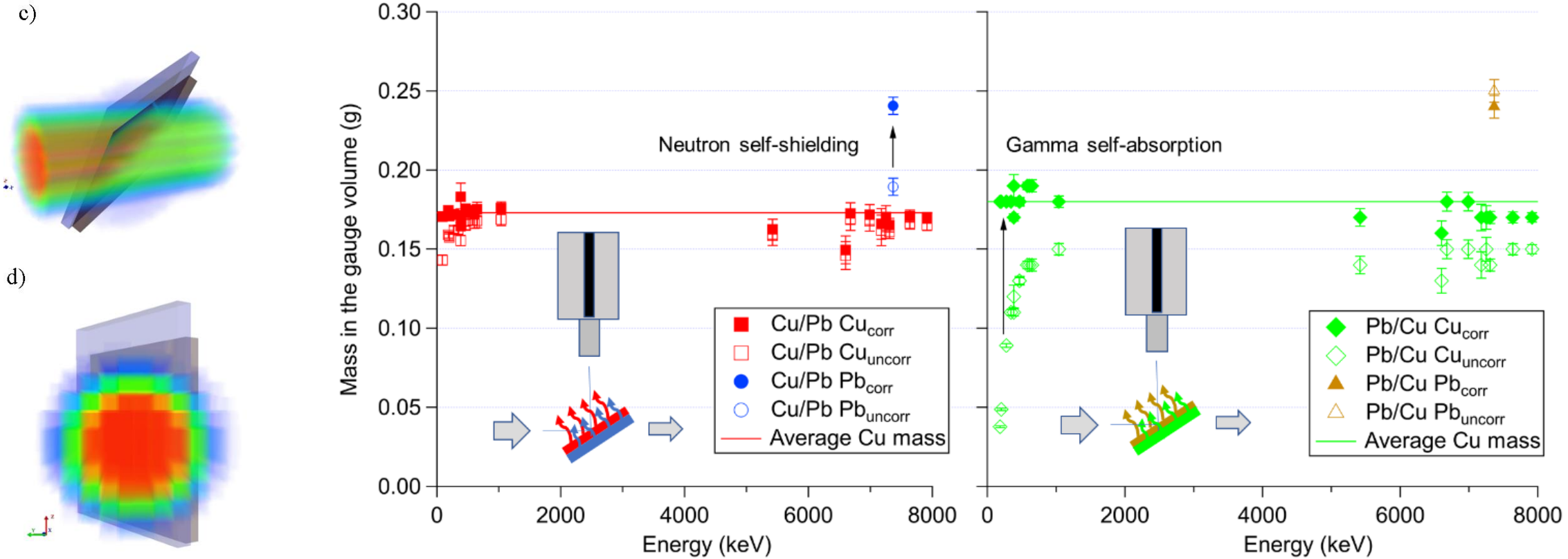
a

a) Absorption of aqueous NaCl solution



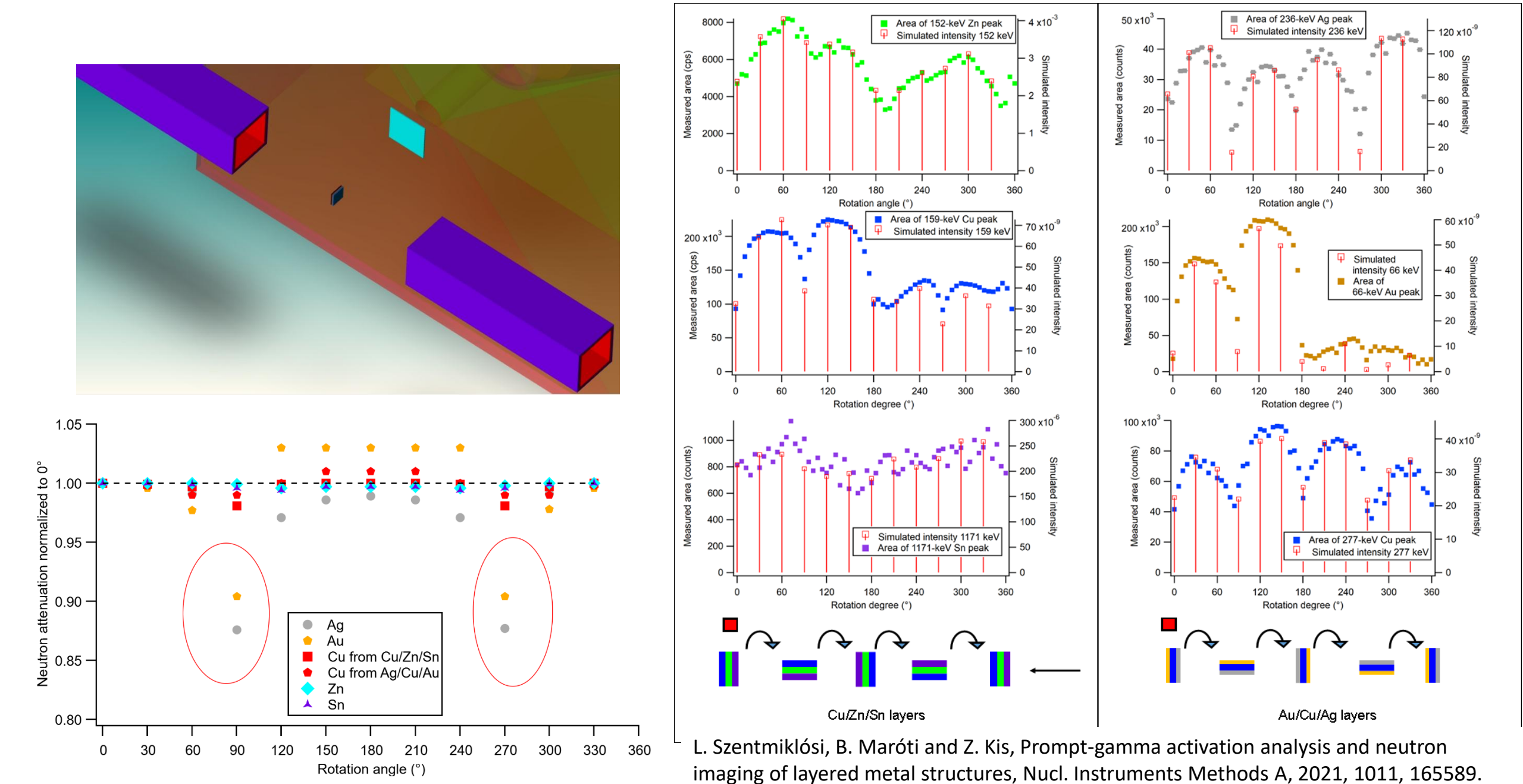
Kis, Z., 2015. Report on water uptake experiments carried out for various stones from Venice by neutron imaging and PGAI methods. MTA EK-NAL-2015-441-1-2-M0.

	whole slab	Cu/Pb, gauge volume				Pb/Cu, gauge volume			
	balance mass (g)	uncorrected mass (g)	unc %	corrected mass (g)	unc %	uncorrected mass (g)	unc %	corrected mass (g)	unc %
Cu	1.5480	0.164	2.2	0.173	2.1	0.141	2.7	0.180	2.0
Pb	2.0146	0.189	2.9	0.241	2.9	0.249	2.9	0.238	2.9
	nominal m%	uncorrected m%	unc %	corrected m%	unc %	uncorrected m%	unc %	corrected m%	unc %
Cu	43.5	46	2.0	42	2.1	36	2.5	43	2.0
Pb	56.5	54	1.7	58	1.5	64	1.4	57	1.5





- The intensities of detected signals from each element depend on the sample orientation
- Front layers tend to absorb neutrons, meaning fewer neutrons reach the rear layers
- Gammas from (n, γ) reaction go through different layers while reaching the HPGe detector
- Real samples: metals with surface patina, an oxide layer, paint, gilding, coins, glazed ceramics
- Principal disagreement between data from PGAA and surface-analytical techniques (portable XRF, PIXE) due to the different probing volumes. If we do both, we can turn this into an advantage.





200 Hungarian Forint, 1993

Handheld XRF:

74.7(2) m% Ag – 25.3(2) m% Cu

PGAA raw composition:

51.0(7) m% Ag – 49.0(7) m% Cu

PGAA, based on two peaks
of similar energies

50(2) m% Ag – 50(2) m% Cu

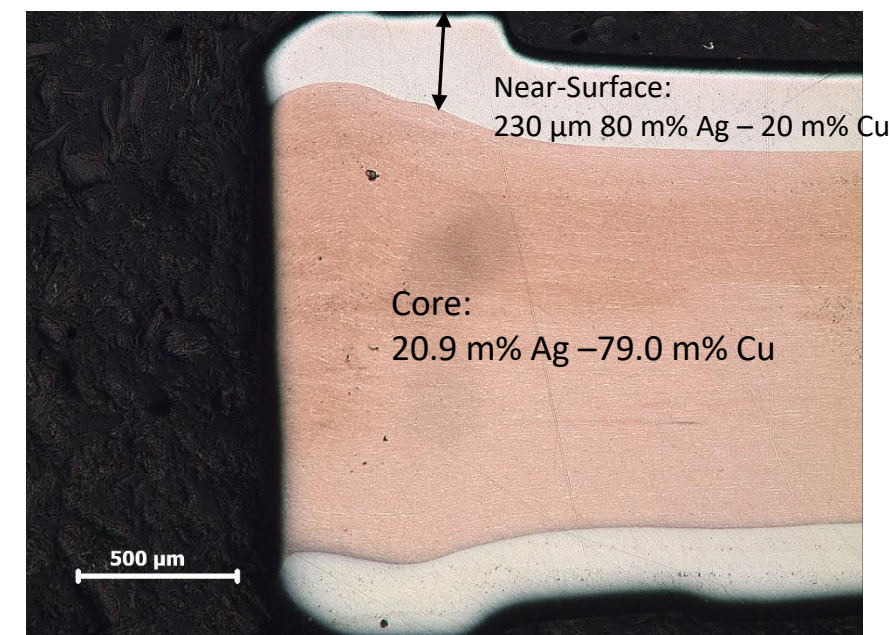
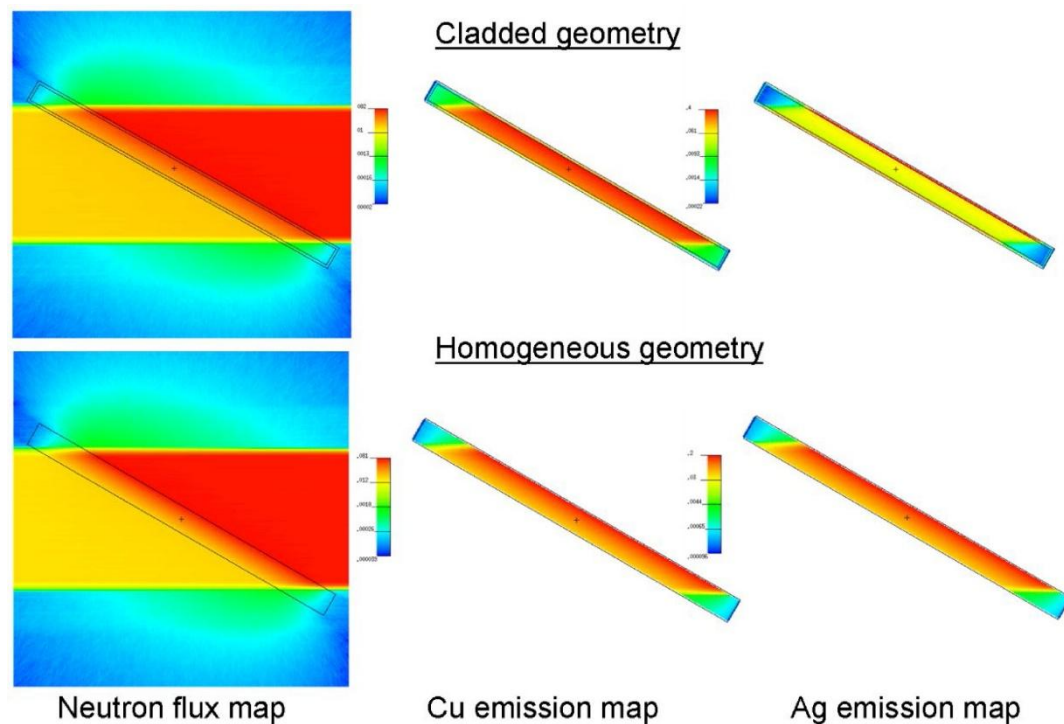
(Ag-206 keV; Cu-202 keV):

PGAA Monte-Carlo
corrected composition:

50.0(7) m% Ag – 50.0(7) m% Cu

Nominal: 50% Ag - 50% Cu

- Minted in the USA, 1967-ban $m=11.5$ g.
- Thickness=2.15 mm, diameter= 30.6 mm
- Core composition Cu 79.1m% - Ag 20.9m%
- Near-surface 230 μm cladding rich in silver:
Ag 80m% - Cu 20m%.
- From material feeds, the avg. composition is
known: Cu 60m% - Ag 40m%.



Takeaway

Main benefits of PGAA

- Non-destructive
- Bulk (average composition)

PGAA is the method of choice for

- Expensive and valuable objects
(material science & heritage science)
- In-depth composition needs to be measured
(complementary to XRF, PIXE, LIBS, LA-ICP-MS)
- Samples that are difficult to dissolve or sample preparation would be time consuming
(chemical analysis of geological samples)
- In-situ monitoring of dynamic processes
(catalysis experiments)

