DETERMINING PARTIAL GAMMA-RAY PRODUCTION CROSS-SECTIONS AT BUDAPEST

Zsolt Révay, Tamás Belgya, László Szentmiklósi,
Zoltán Kis, Gábor Molnár

Institute of Isotopes, Budapest

Prompt Gamma Activation Analysis

- Based on radiative neutron capture, or (n,γ) reaction
- Prompt gamma radiation is characteristic
  - Energy identifies the nuclide (element)
  - Intensity proportional to mass
- Very linear: $\frac{A}{\varepsilon t} = \frac{m}{M} N_A \Phi \sigma_\gamma$ $\sigma_\gamma = \sigma_0 P_\gamma \theta$
- Large number of non-characteristic lines...

PGAA facilities at reactors

- Budapest, Japan
- Korea, India, Munich (earlier Switzerland)
- USA: Washington, Texas, Missouri
- Argentina, China?, Brazil?, Portugal?, Morocco?

Earlier PGAA databases

- 1969-70: MIT (Rasmussen-Orphan)
  - 75 elements measured with Ge(Li)
- 1981: Lone table (Chalk River)
  - Compilation of mainly Rasmussen's data
- 1993: IAEA – Lone table as an attachment to a report
- 1995: Tuli database (Alfassi's PGAA book)
  - ENSDF data for nuclides, where available
  - The rest is Lone table (Z<20)
  - Only energies and relative intensities

- No analytical database until 1996!!!

Relative method

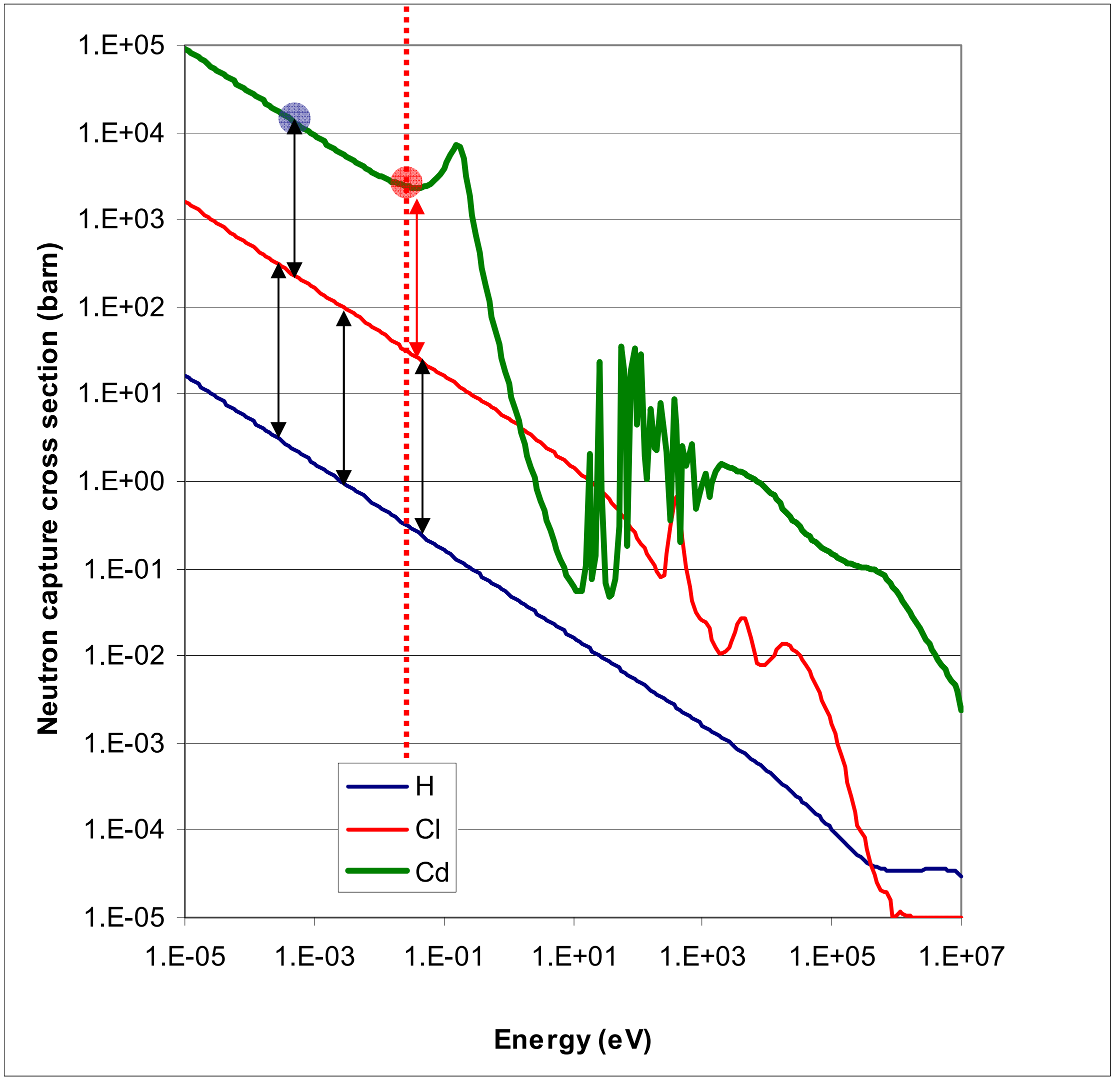
- Peak area ratios
  - Relative efficiency: $A/(\varepsilon t) = aP_\gamma$
    - Cross-section ratios
      - Mass ratios
      - Concentrations, composition

$$\frac{A_1/\varepsilon_1}{A_2/\varepsilon_2} = \frac{n_1}{n_2}\frac{\sigma_{\gamma,1}}{\sigma_{\gamma,2}}$$

- Comparator: earlier Cl, now H
  - 2223 keV   0.3326 barn ±0.2%

Thermal cross section

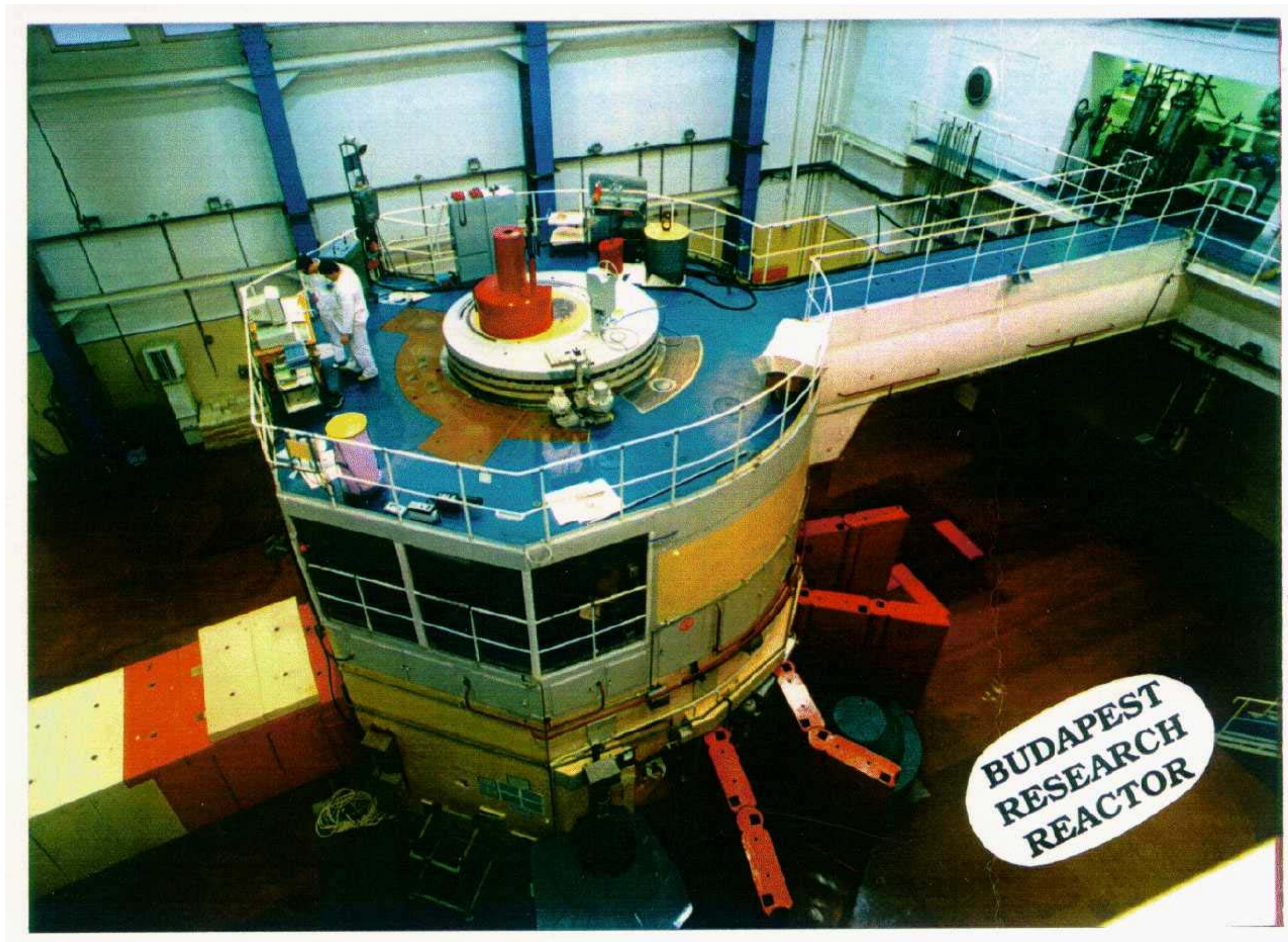
- at low E: 1/*v* law
  - highest reaction rate for **cold neutrons**
- Thermal cross-section taken for 25meV neutrons
- Westcott *g* factor is used to transform between different T-s, distributions




Budapest PGAA
facility

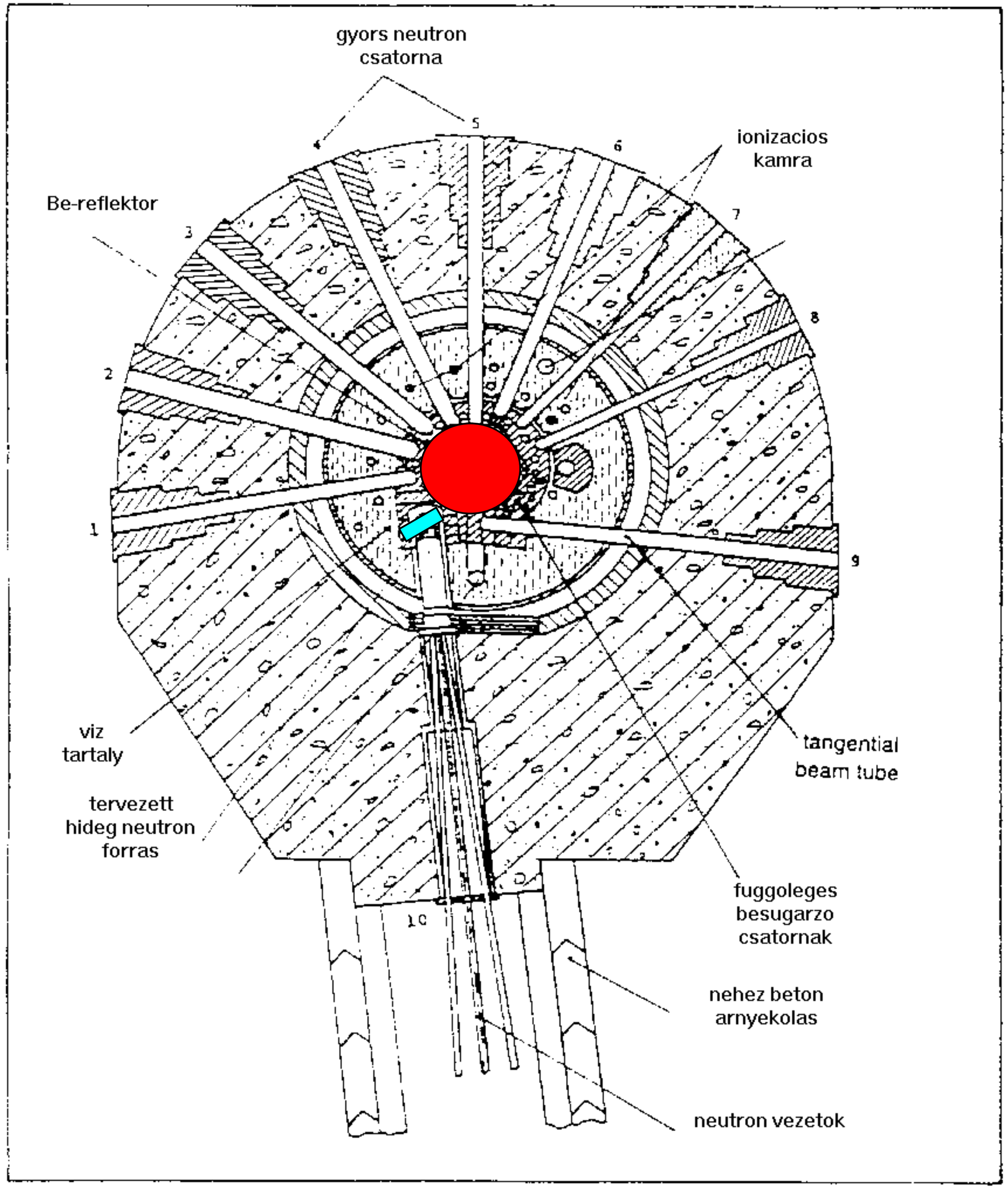
Research Reactor

- 20 MW
- water cooled
- water moderated
- thermal flux 10^{14} cm^{-2} s^{-1}

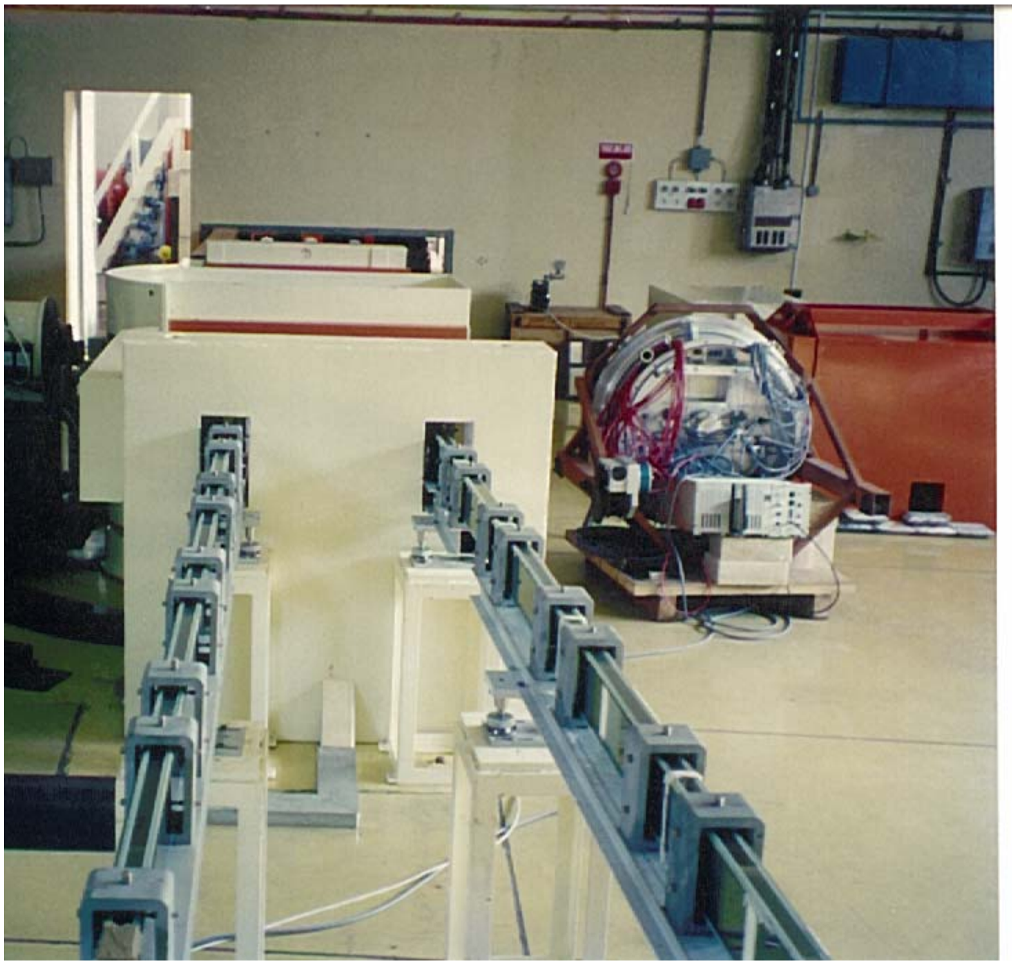


Cold neutron source at Budapest

$400\ cm^3$
20 K liquid H_2



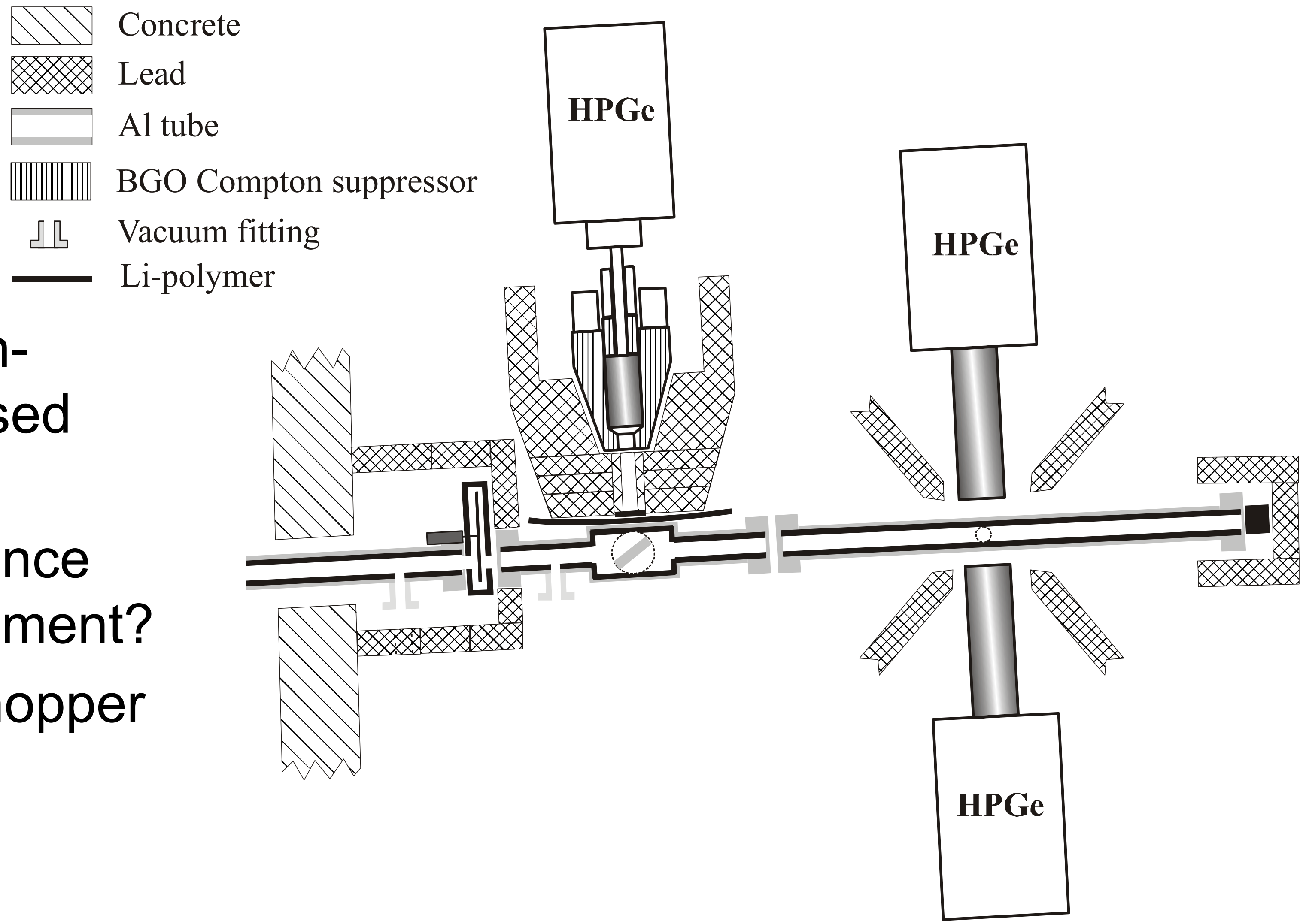
Neutron guides



- Curved supermirror guides
- relatively small losses
- **low background**

- **ONLY cold neutrons!!!**

Budapest PGAA facility

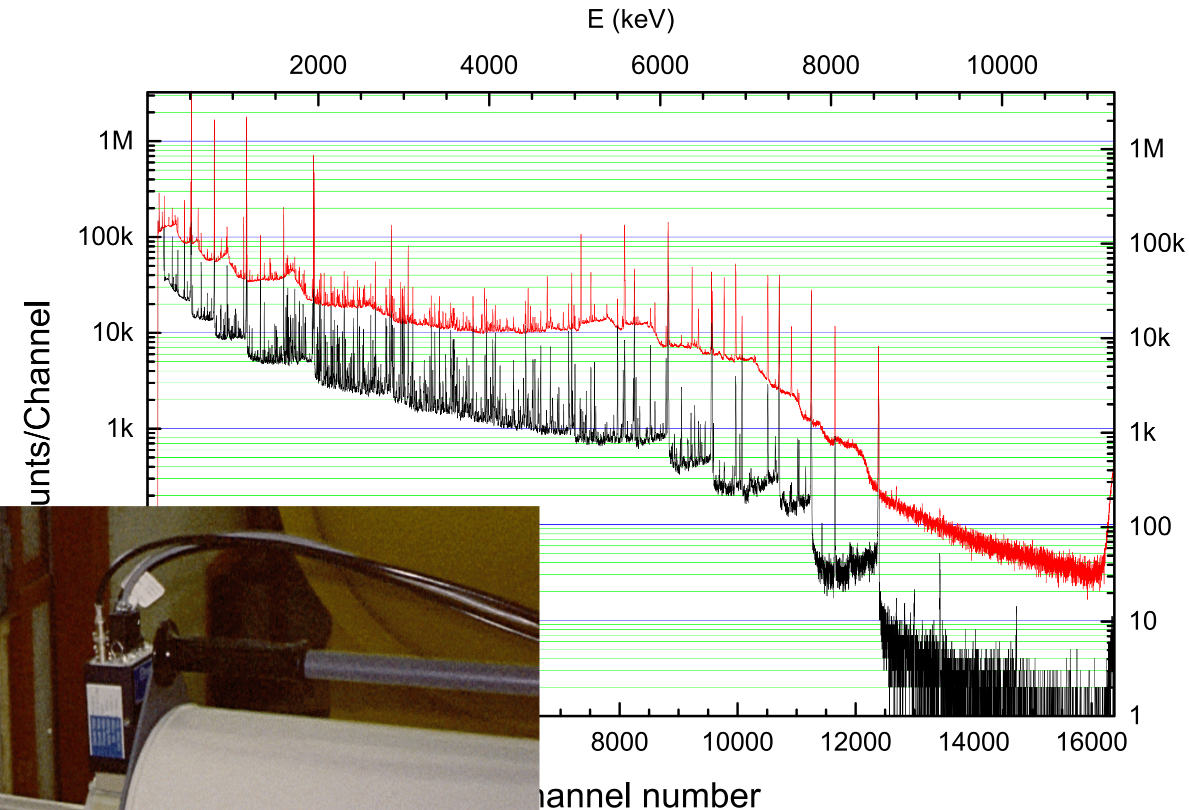


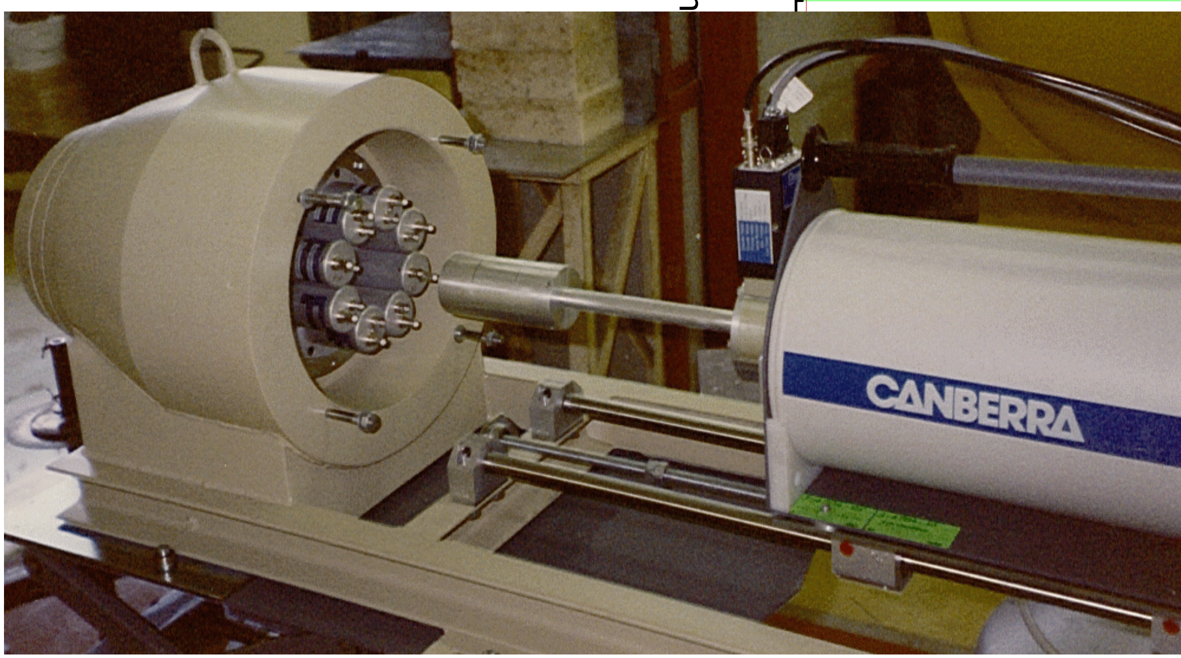
- Compton-suppressed HPGe
- Coincidence measurement?
- Beam chopper

Compton-suppressed HPGe

Room background:

0.6 cps instead of 100cps





Flux and background

- Budapest 1997: 2.5×10^{6} cm^{-2} s^{-1} 3 cps
- Budapest 2001: 3×10^{7} cm^{-2} s^{-1} 5 cps
- Budapest 2009: 1.5×10^{8} cm^{-2} s^{-1} 10 cps
- (Garching 2009: 3×10^{9} cm^{-2} s^{-1} 300 cps)

- Max count-rate: >10,000 cps!!!

The PGAA project in the Institute of Isotopes

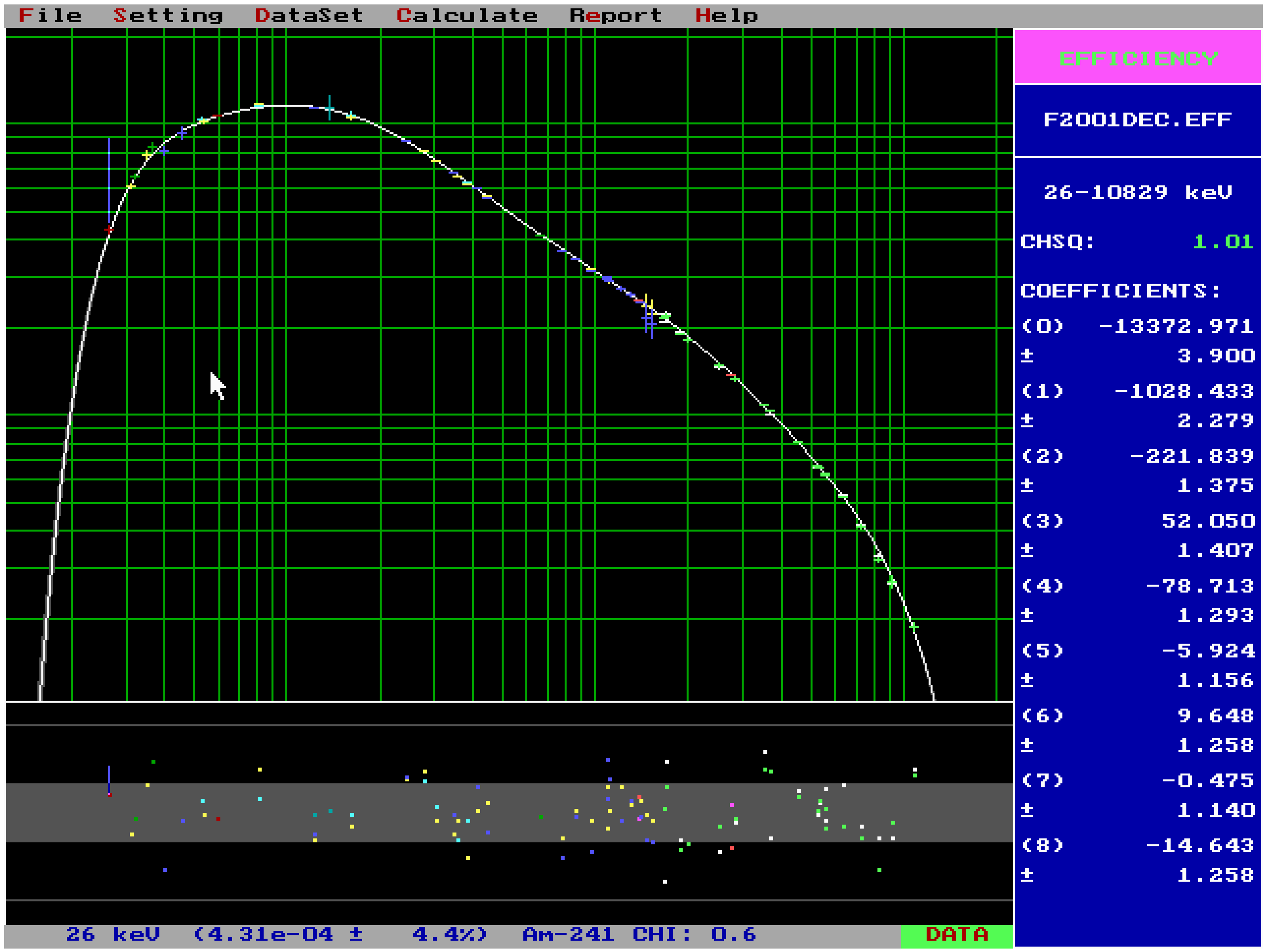
History of the PGAA facility

- 1992 upgraded reactor starts
  1995 first PGAA measurement on the thermal beam
  1997–1998 establishment of PGAA data library
  1999 – 2000 applications
  2001 new cold beam
  2002 – 2004 Handbook and Atlas
  2006 – revision of data

Calibration

- Efficiency
- Non-linearity
- Peak-shape calibration (asymmetric parts)
- Backgrounds (room, beam-on, etc.)

Efficiency fitted to measured data (^{152}Eu, ^{133}Ba, Cl(n,γ)) 50 keV—11 MeV



Non-linearity

- ~ ± 1channel = ± 0.7 keV
- It is relatively constant in time
- measured using crystal-spectrometer data of ^{152}Eu, $^{35}Cl(n,\gamma)$
- after correction the uncertainty of the peaks is mainly determined by the peak statistics
  - 0.01 keV below 2 MeV
  - 0.1 keV around 8 MeV

Library measurements

1. Elemental spectra: to obtain ...
   - Relative positions
   - Relative intensities
2. Energy calibration: to absolutize energy scale
   - 2 energies det-d for the 2-point E-calibration
   - Non-linearity
3. Standardization: to absolutize intensity scale
   - efficiency ratios
   - compounds or mixtures

1. Elemental spectra

- Elements
  - Metals
- Oxides
  - Nonmetals
  - Metals
- Other simple compound with low-Xsec elements
  - Carbonates, hydroxides, carbides, nitrates etc.

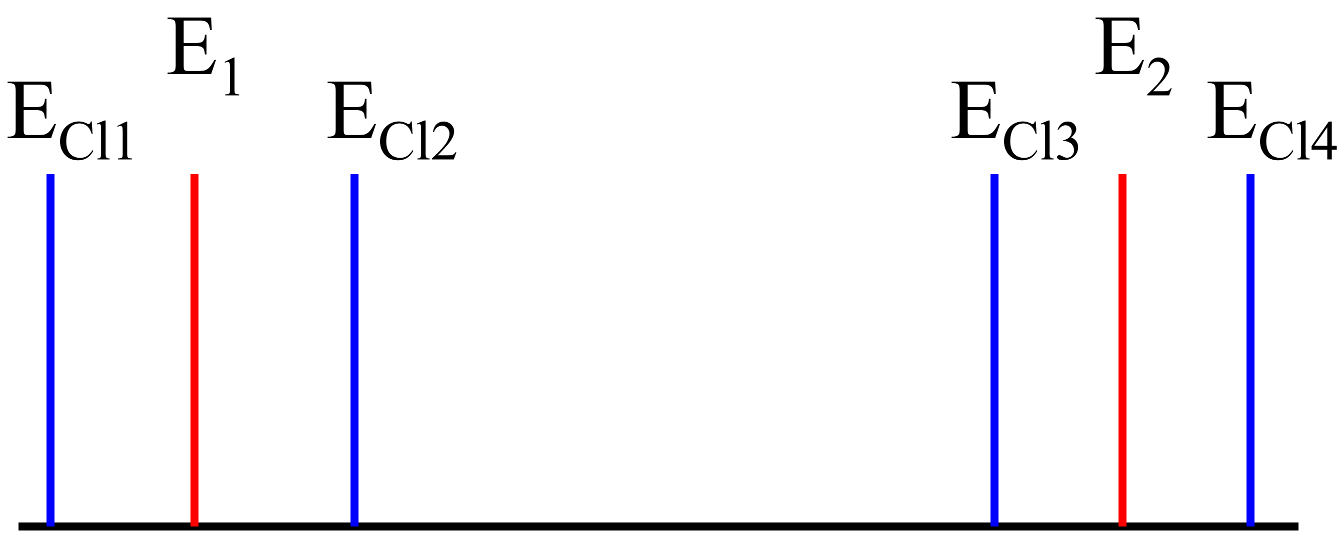
1. Measurements of elements

1 H O			1 D O														2 He
3 Li CO_3,C-F	4 Be * O											5 B C, H-O	6 C ** H	7 N C-D-O, NO_3	8 O H, Be	9 F C	10 Ne
11 Na * CO_3,C-H-O	12 Mg *											13 Al ** O	14 Si * O N	15 P * **O**	16 S **	17 Cl **C,C-H**	18 Ar *
19 K HCO_3	20 Ca * O CO_3	21 Sc O	22 Ti ** O	23 V O	24 Cr * O-H	25 Mn * O	26 Fe **	27 Co *	28 Ni **	29 Cu * O	30 Zn * O	31 Ga **	32 Ge * O	33 As O	34 Se * O-H	35 Br * C-H	36 Kr *
37 Rb O CO_3	38 Sr CO_3	39 Y O	40 Zr O	41 Nb O	42 Mo **	43 (Tc)	44 Ru **	45 Rh * C-H	46 Pd *	47 Ag **	48 Cd **	49 In *	50 Sn **	51 Sb O	52 Te **	53 I * **C-H**	54 Xe F
55 Cs O	56 Ba OH,CO_3	57 La O	72 Hf * O	73 Ta * O	74 W O	75 Re *	76 Os * O C-H	77 Ir * O	78 Pt *	79 Au *	80 Hg ** O	81 Tl *	82 Pb **	83 Bi **	84 (Po)	85 (At)	86 (Rn)
87 (Fr)	88 (Ra)	89 (Ac)															

58 Ce O C-H-O	59 Pr O	60 Nd O	61 (Pm)	62 Sm **O**	63 Eu **O**	64 Gd **O**	65 Tb O	66 Dy *	67 Ho O	68 Er **O**	69 Tm O	70 Yb O	71 Lu O
90 Th NO_3	91 (Pa)	92 U ***O*** C-H-O											

2. Energy calibration

- Energy difference method
- Crystal spectrometer data for ^{35}Cl
- Element measured in presence of Cl



3. Standardization

- Stoichiometric compounds
  - Chlorides, nitrates
- Homogeneous mixtures
  - Water solution
  - Water-TiO_2-XO suspension
- Relative to the comparator

$$\sigma_{\gamma,x} = \frac{n_c}{n_x} \frac{A_x / \varepsilon(E_x)}{A_c / \varepsilon(E_c)} \sigma_{\gamma,c}$$

Intermediate comparators

element	compounds	Comparators	σ_γ(barn)	Statistical unc. (%)	Total unc. (%)
H^*			0.3326		0.2
N – 1884	Pyridine, NH_4NO_3, NH_4Cl, melamine	H – 2223	0.01452	0.2	0.4
C – 4945	Polyethylene, melamine, urea, pyridine	H – 2223 N – 1884	0.00259	0.6	0.8
S – 841	$(NH_4)_2SO_4$	H – 2223	0.353	0.9	1.0
Cl – 1951	NH_4Cl, NaCl solutions	H – 2223	6.5095	0.3	0.4

3. Standardization measurements

1 **H** alap			1 D H														2 He
3 Li C,N	4 Be N, O											5 B *H*	6 C H N	7 **N** H Cl	8 O H	9 F K,C,Ca	10 Ne
11 Na *H* Cl S **B**	12 Mg H Cl S,*Fe** **B**											13 Al H Cl S,*Fe** **B**	14 Si N O *Fe**	15 P H Na	16 **S** H Na, Al	17 **Cl** 3H **B**	18 Ar absz: Cl
19 K H Cl **B**	20 Ca Cl *Fe**	21 Sc *H* S,*Ti* **B**	22 Ti Cl	23 V *H* **B**	24 Cr H Cl	25 Mn H Cl **B**	26 Fe 2Cl	27 Co H Cl **B**	28 Ni H Cl **B**	29 Cu H Cl	30 Zn Cl **B**	31 Ga H N **B**	32 Ge Co **B**	33 As H Na **B**	34 Se *H* **B**	35 Br H *Cl* **B**	36 Kr
37 Rb Cl **B**	38 Sr Cl **B**	39 Y Cl **B**	40 Zr Cl N	41 Nb Cl	42 Mo Cl	43 (Tc)	44 Ru *H* Cl	45 Rh H Cl	46 Pd Cl	47 Ag *H* Cl	48 Cd *H* Cl	49 In Sb **B**	50 Sn H Cl	51 Sb S	52 Te H Cl	53 I H *Cl*	54 Xe F
55 Cs Cl	56 Ba H Cl	57 La Cl	72 Hf H Cl	73 Ta H *Ti,H*	74 W H Na	75 Re Cl	76 Os H	77 Ir Cl	78 Pt Cl	79 **Au** H Cl	80 Hg Cl	81 Tl S	82 Pb Cl N	83 Bi Cl	84 (Po)	85 (At)	86 (Rn)
87 (Fr)	88 (Ra)	89 (Ac)															

58 Ce H C	59 Pr *H* *S*	60 Nd *H* *S*	61(Pm)	62 Sm *H* *S*	63 Eu *H* *S* **B**	64 Gd *H* *S*	65 Tb *H* *S*	66 Dy *H* *S*	67 Ho *H* *S*	68 Er *H* *Cl*	69 Tm *H* *S*	70 Yb *H*	71 Lu *H* *S*
90 Th H N **B**	91 Pa	92 U H C **B**											

PGAA library

Z	El	A	MW	#	E	dE	σ	dσ %	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

Verification

- SRM, CRM
- Samples with partly known composition

Verification

1 H **komp** oldatok			1 D H														2 He
3 Li	4 Be											5 B *H*, üveg GEO	6 C karbo- nátok	7 N **komp**	8 O oxidok	9 F Ca	10 Ne
11 Na komp üveg	12 Mg üveg											13 Al cem, kat, GEO	14 Si üveg, kat, GEO	15 P H Na	16 S komp cem GEO	17 Cl **komp**	18 Ar
19 K SRM	20 Ca SRM, cem	21 Sc	22 Ti Cl GEO	23 V kat	24 Cr SRM, kat	25 Mn SRM GEO	26 Fe SRM, GEO	27 Co oldat	28 Ni kat, fémüveg	29 Cu Ag-Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr fémüveg	41 Nb kat	42 Mo kat	43 (Tc)	44 Ru	45 Rh	46 Pd fémüveg	47 Ag Ag-Cu	48 Cd SRM GEO	49 In	50 Sn Sn-Cd	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt kat	79 Au komp	80 Hg	81 Tl	82 Pb Pb-Cd	83 Bi	84 (Po)	85 (At)	86 (Rn)
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58 Ce	59 Pr	60 Nd	61(Pm)	62 Sm	63 Eu	64 Gd üveg, GEO	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
90 Th	91 Pa	92 U											

Calculation of uncertainties

$$\sigma_{\gamma,x} = \frac{n_c}{n_x} \frac{A_x / \varepsilon(E_x)}{A_c / \varepsilon(E_c)} \sigma_{\gamma,c}$$

- Uncertainty= statistical + systematic

$$\delta\sigma_{\gamma,x} = \sqrt{\left(\delta A_{\gamma,x}\right)^2 + \left(\delta A_{\gamma,c}\right)^2 + \left(\delta \frac{\varepsilon(E_{\gamma,c})}{\varepsilon(E_{\gamma,c})}\right)^2 + \left(\delta\sigma_{\gamma,c}\right)^2}$$

(mainly) thermal PGAA library

- 1997—2000 measurement, evaluation
  - 5, 25, 100 lines/element
- IAEA CRP (finished 2000)
  - TECDOC (2007)
- 2004 Handbook of PGAA with neutron beams
  - Atlas and catalog: 100 lines/element

Cold PGAA library

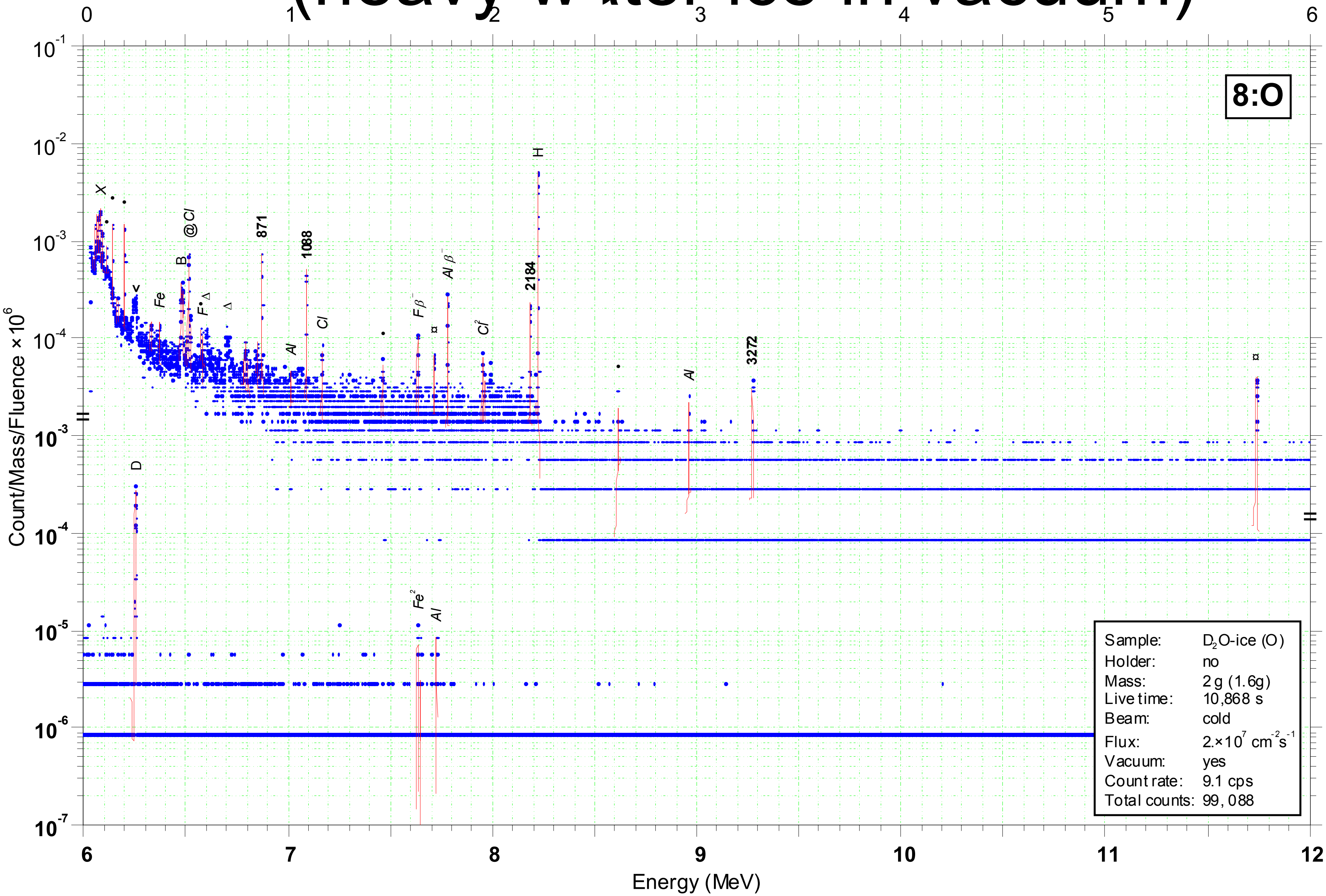
- In progress
- 2004 Budapest: 16 elements
- 2009 Budapest + Munich: 19 elements
- Complete revision planned

- Statistical and systematic uncertainties handled separately

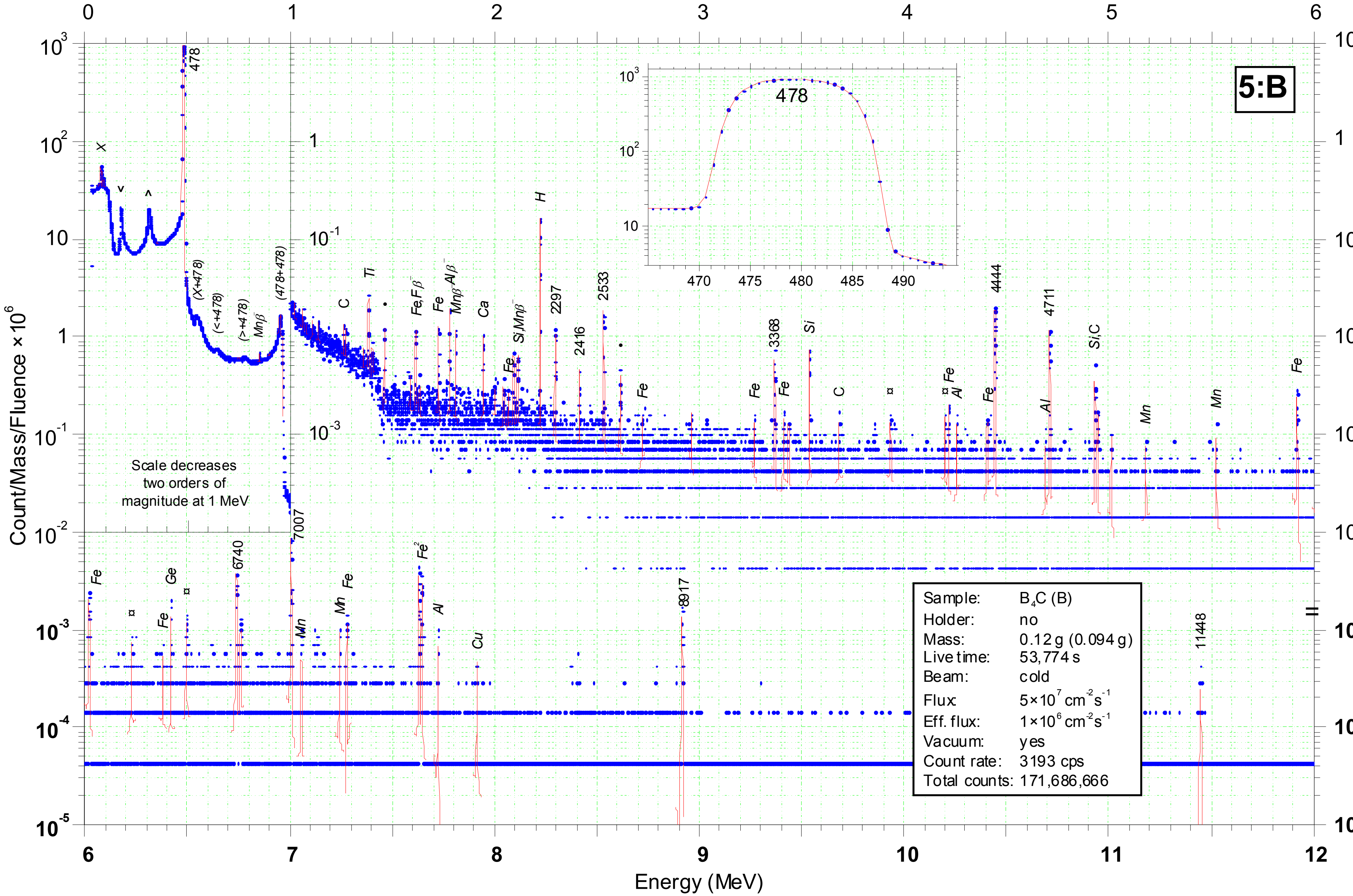
Spectra for the atlas

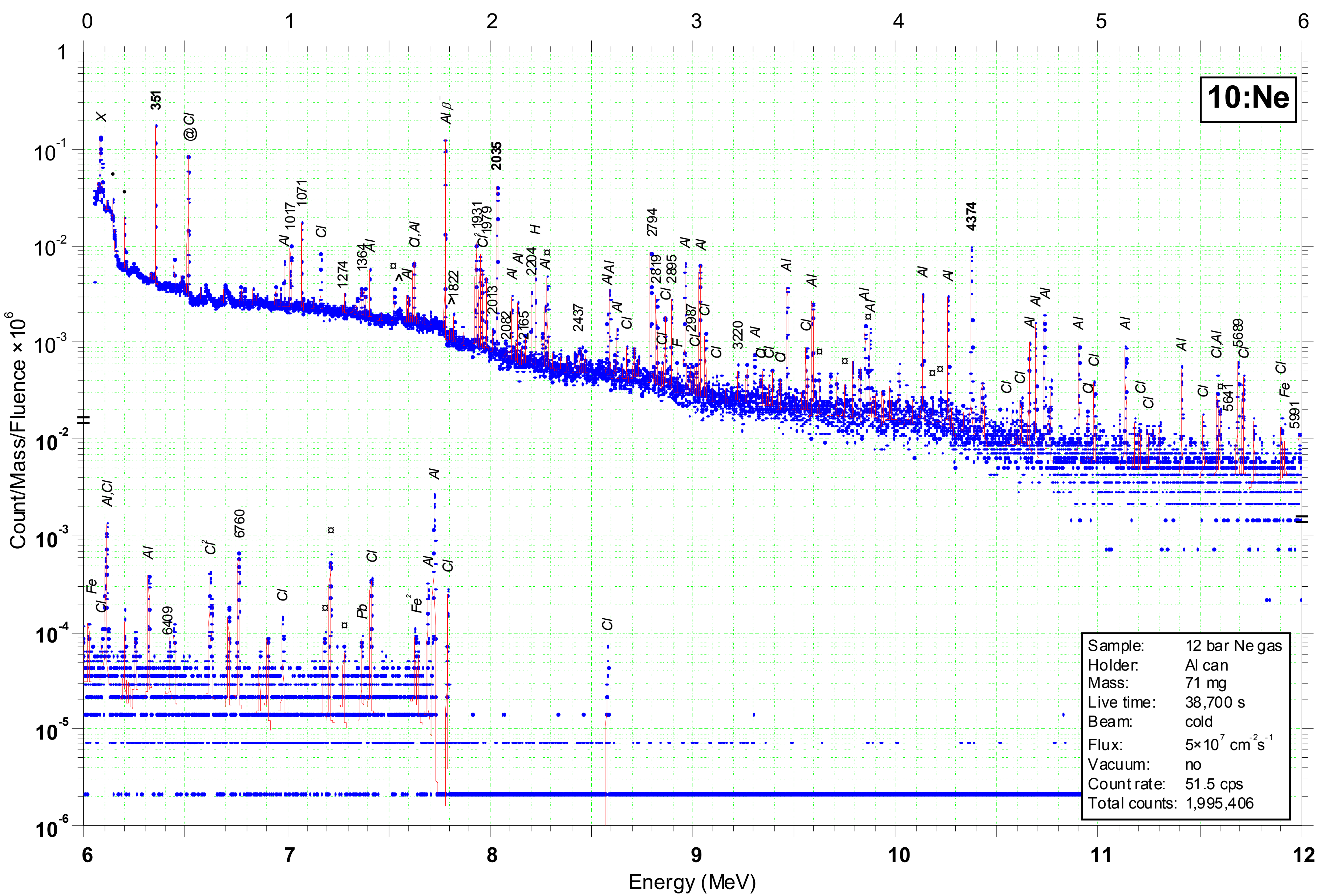
- No visible (or weak) pollution
- Statistics is not so important
- normalization:
  - for 1 g of element
  - for 1 s of acquisition time
- every characteristic peak is fitted

PGAA spectrum of O
(heavy water ice in vacuum)



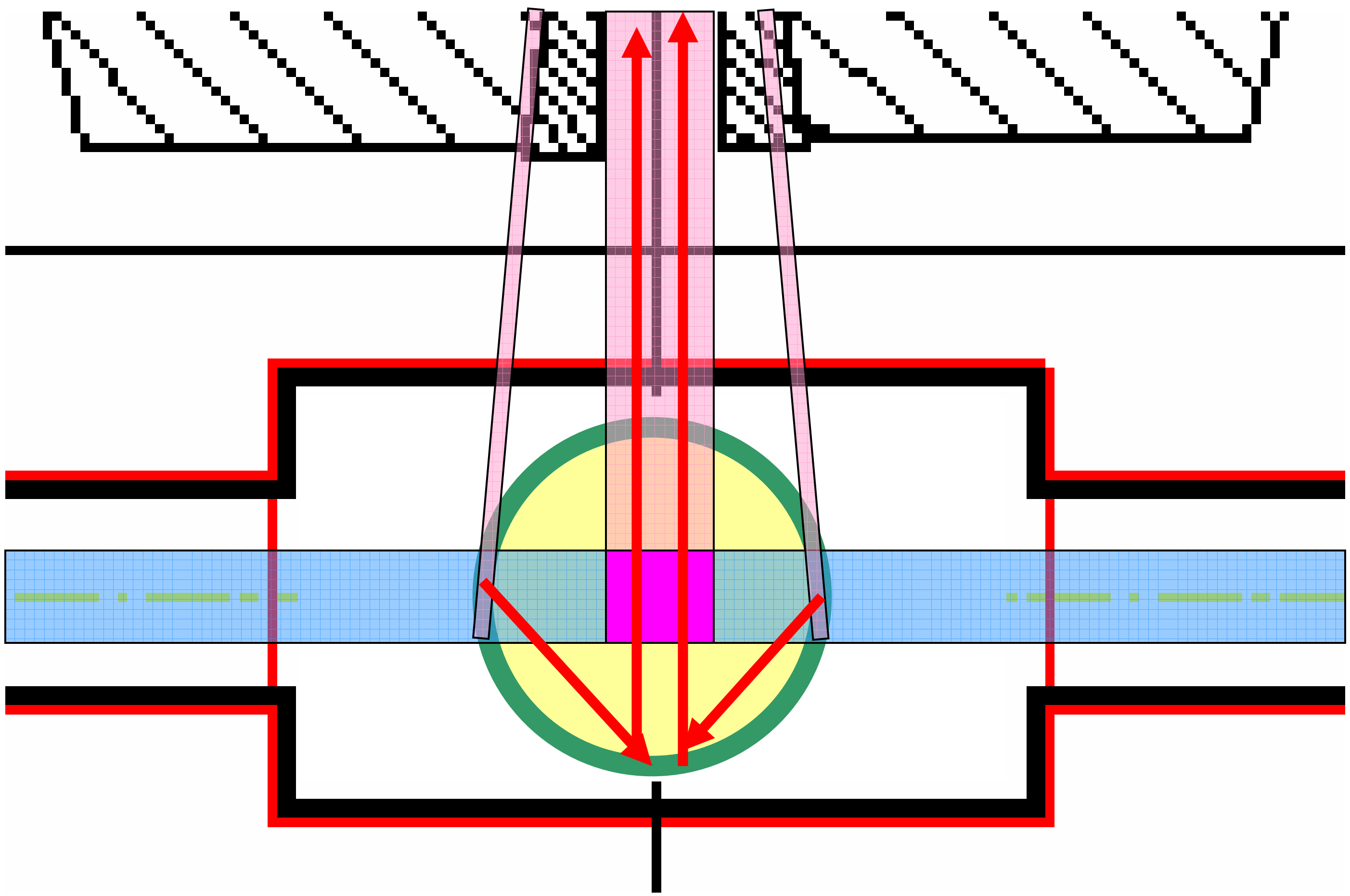
Boron (carbide)



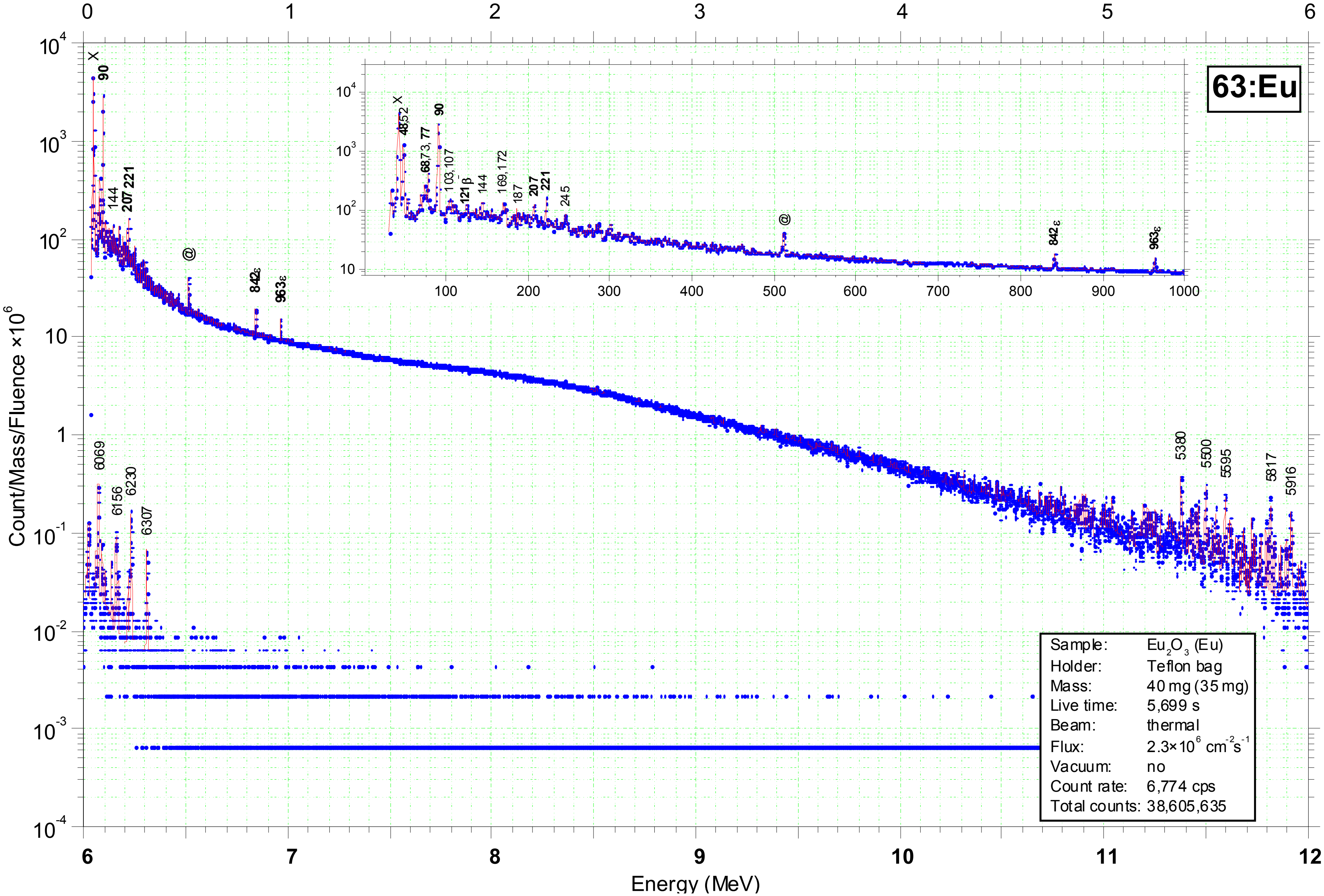


10:Ne
Count/Mass/Fluence × 10^6
Energy (MeV)
Sample: 12 bar Ne gas
Holder: Al can
Mass: 71 mg
Live time: 38,700 s
Beam: cold
Flux: 5×10^7 cm^-2s^-1
Vacuum: no
Count rate: 51.5 cps
Total counts: 1,995,406

“Invisible container” method



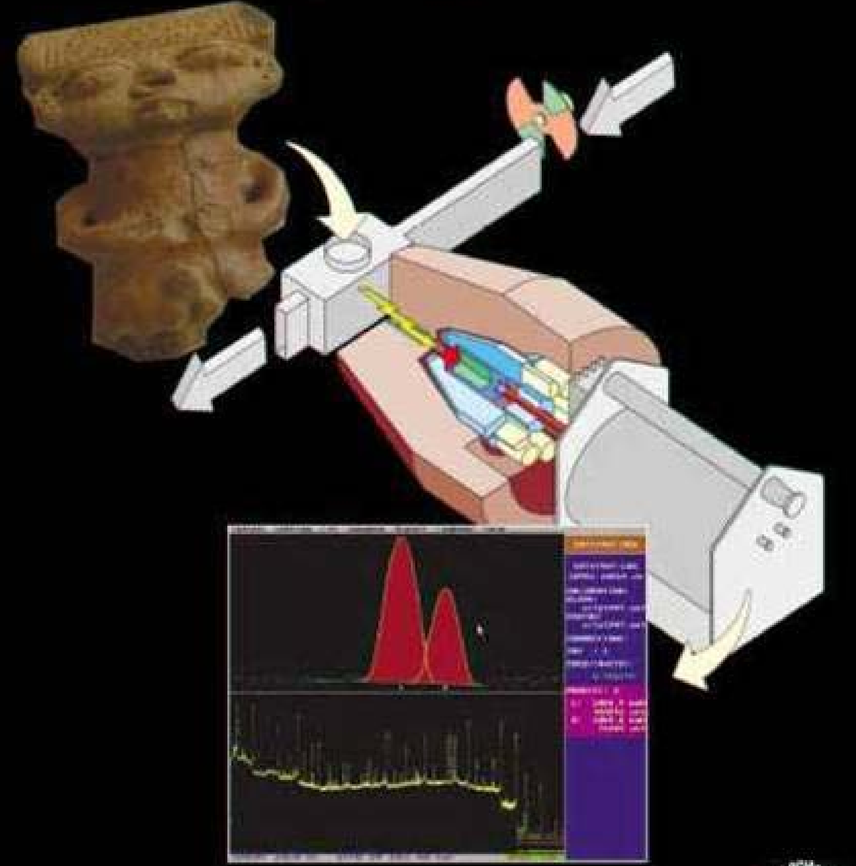
Eu, huge continuum



HANDBOOK OF PROMPT GAMMA ACTIVATION ANALYSIS

WITH NEUTRON BEAMS

Edited by Gábor L. Molnár



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