(n,n'γ) transitions Induced by D(d,n)He neutrons

Preliminary results

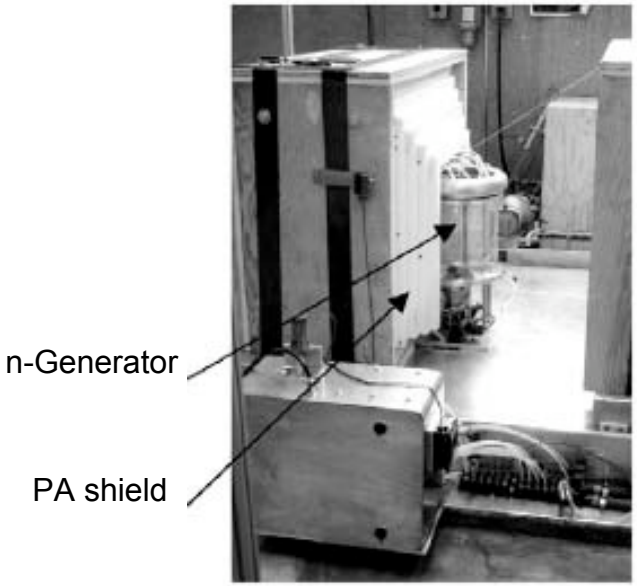
Yair Ben-Dov

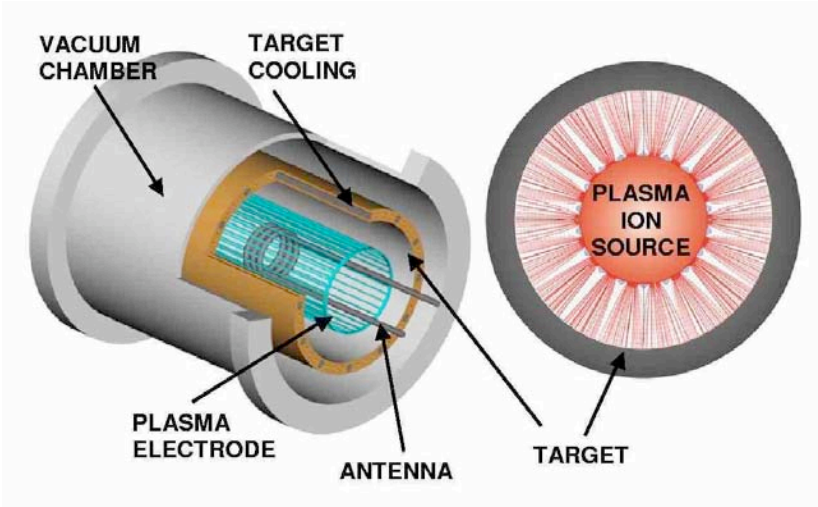
Outline

- The AFRD n-generator
- Previous studies
- Current setup & goals
- MCNP simulations
- Cross sections measurements
- LLD's
- What's next

The AFRD neutron generator

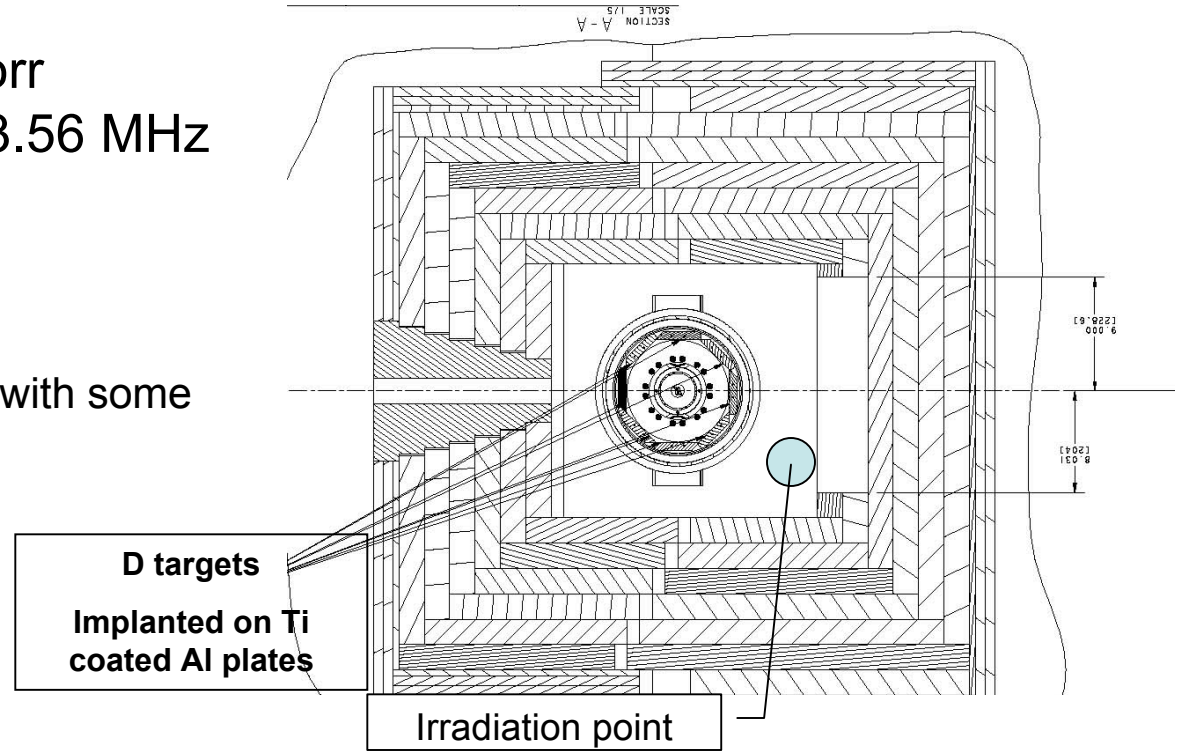
- RF induction discharge generates deuterium plasma
- ~100 kV potential difference accelerates D ions towards the D target
- Co-axial target design to maximize target area and n flux





Operation specifications

- D gas pressure: 5-6 mTorr
- RF power: 1200 Watt, 13.56 MHz
- HV: 80-100 kV
- D Ion current: 60 mA
- Neutron yield: 10^9 n/s
- Long durable operation (with some limitations)



Previous publications

Analysis of Fissile Materials by High-Energy Neutron-Induced Fission Decay Gamma Rays[a]

R.B. Firestone[1*], G.A. English[1], J. Reijonen[1], F. Gicquel[1], K-N. Leung[1], D.L. Perry[1], G. Garabedian[2], B. Bandong[2], Zs. Revay[3] and G.L. Molnar[3]

[1]*Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley, CA 94720, USA*

[2]*Lawrence Livermore National Laboratory, Livermore, CA 94550, USA*

[3]*Institute for Isotope and Surface Chemistry, Budapest, Hungary*

NSD 2003

PGAA/NAA Analysis with the Lawrence Berkeley National Laboratory (LBNL) D+D Neutron Generator

R.B. Firestone, G.A. English , D.L. Perry, J.P. Reijonen, F. M. Gicquel, S. Basunia, and K.N. Leung

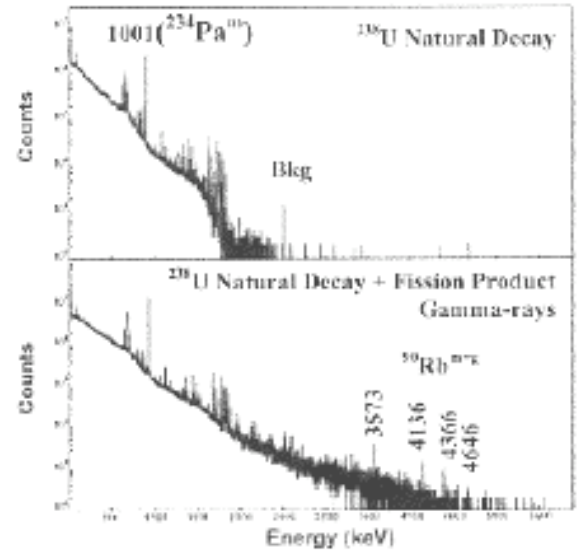
Lawrence Berkeley National Laboratory, Berkeley, CA 94720

G.F. Garabedian and B.B. Bandong

Lawrence Livermore National Laboratory, Livermore, CA 94550-9234

G.L. Molnár, L. Szentmiklosi and Zs. Révay

Institute of Isotope and Surface Chemistry, Budapest H-1525, Hungary



AccApp 2003

Neutron Flux and Cadmium Ratio Measurement of the Co-axial Cylindrical Neutron Generator

M. S. Basunia[1], H. A. Shugart[2], J. Reijonen[3], F. Gicquel[3], and R. B. Firestone[1]

[1] *Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720*

[2] *Department of Physics, UC Berkeley, Berkeley, CA 94720*

[3] *Accelerator and Fusion Research Division, Berkeley National Laboratory, Berkeley, California 94720*

NSD 2004

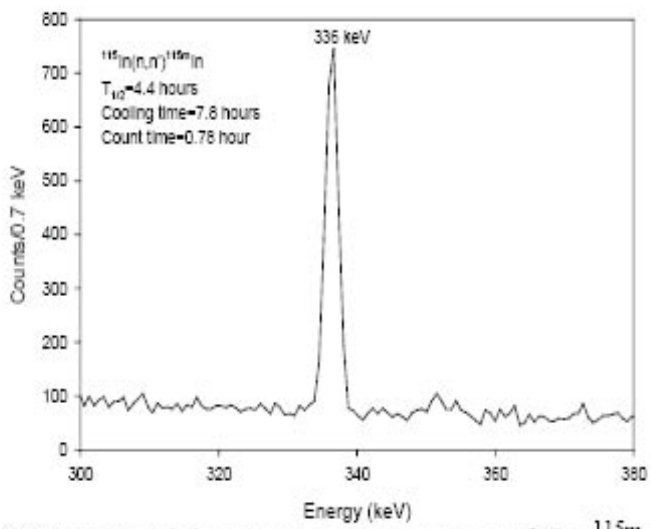


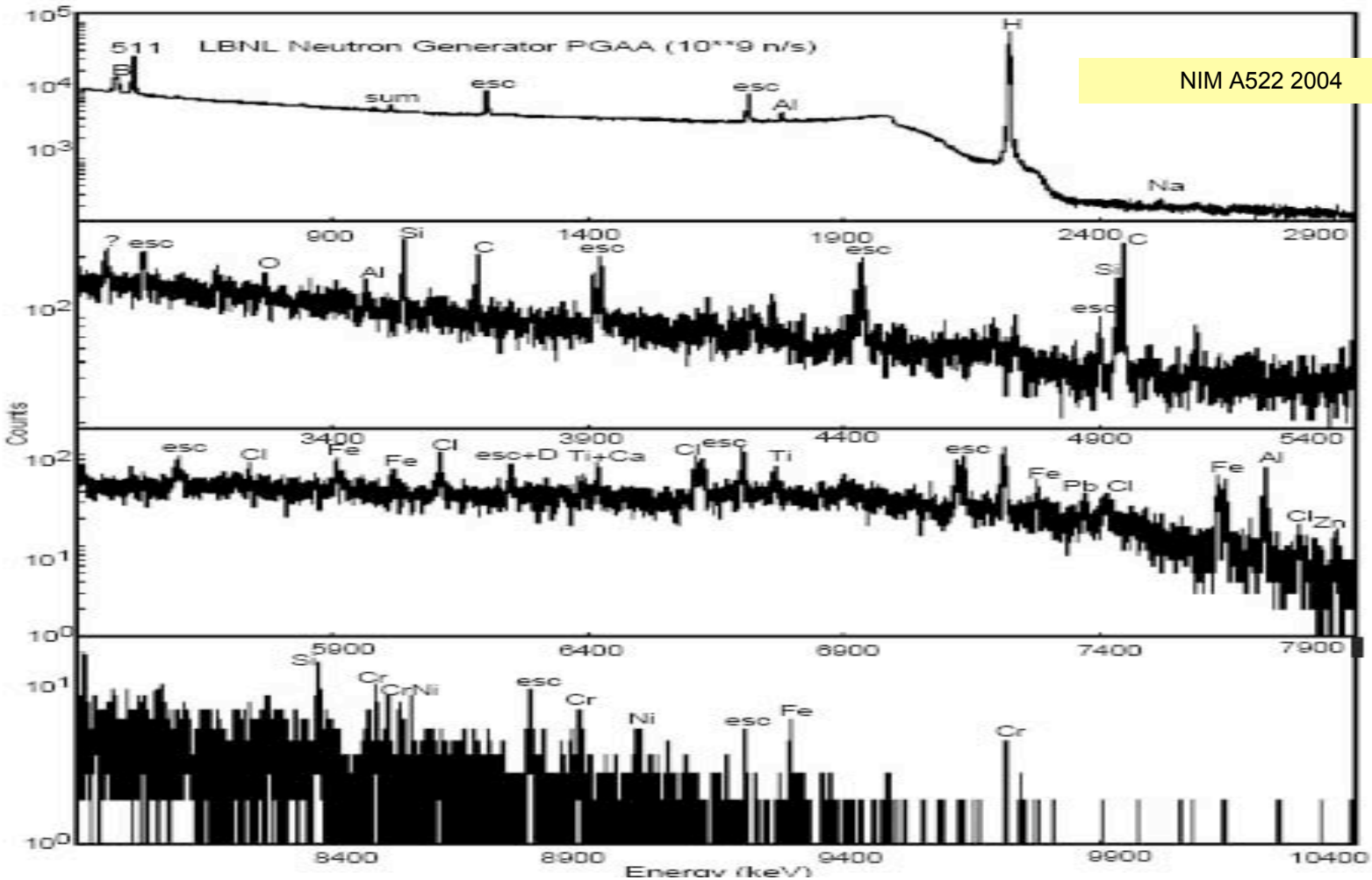
FIG. 1: Partial gamma ray spectrum of the ^{115m}In decay.

Table 1: Neutron flux (n.cm^{-2}.sec^{-1}) at different locations around the NG, uncertainty ≈20%.

Sample location	Distance from the center of the NG cylinder	Thermal neutron flux	Fast neutron (>0.5 MeV) flux
Pneumatic terminal	≈28.2 cm	1.1E+04	1.4E+05
Inner shielding	≈26.2 cm	1.3E+04	1.0E+05
Top of the NG		1.1E+04	1.5E+05

First PGAA and NAA experimental results from a compact high intensity D–D neutron generator ☆

J. Reijonen[a,*], K.-N. Leung[a], R.B. Firestone[a], J.A. English[a], D.L. Perry[a],



NIM A522 2004

Available online at www.sciencedirect.com
SCIENCE DIRECT®
ELSEVIER
Annals of Nuclear Energy 31 (2004) 1883–1900
annals of NUCLEAR ENERGY
www.elsevier.com/locate/anucene

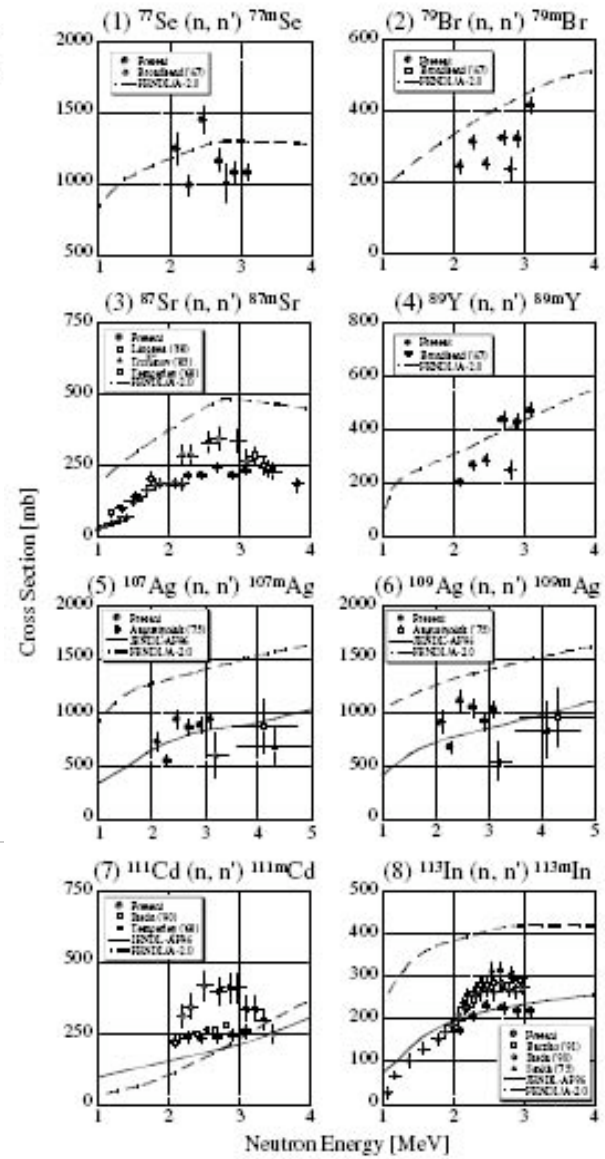
Measurements of activation cross-sections of (n, n′) reaction with d-D neutrons in the energy range of 2.1–3.1 MeV

Toshiaki Shimizu [a,*], Hitoshi Sakane [a,1], Michihiro Shibata [a,2], Kiyoshi Kawade [a], Takeo Nishitani [b]

[a] *Department of Energy Engineering and Science, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan*

[b] *Department of Fusion Engineering Research, Naka Fusion Research Establishment, Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken 319-1195, Japan*

Received 23 March 2004; accepted 15 May 2004
Available online 20 July 2004



77Se	79Br
87Sr	89Y
107Ag	109Ag
111Cd	113In

Motivation and goals

- Need for accurate experimental cross sections at 2.5 MeV for :
  - DD generator applications such as:
    - Neutron generator calibration
    - Nuclear reactor calculations
    - Industrial / Medical (n-interrogation, BNCT)
    - Homeland security (nonproliferation, contraband)
    - Neutron Activation Analysis (prompt, delayed)
  - Comparison to evaluated data bases
- Determine quantitative capabilities of the LBNL n-generators
- Determine (n,n'γ) cross sections at En ≈ 2.5 MeV
- Determine current setup LLD's for the elements investigated

Current setup: The Rabbit system

Current setup: Detection stand

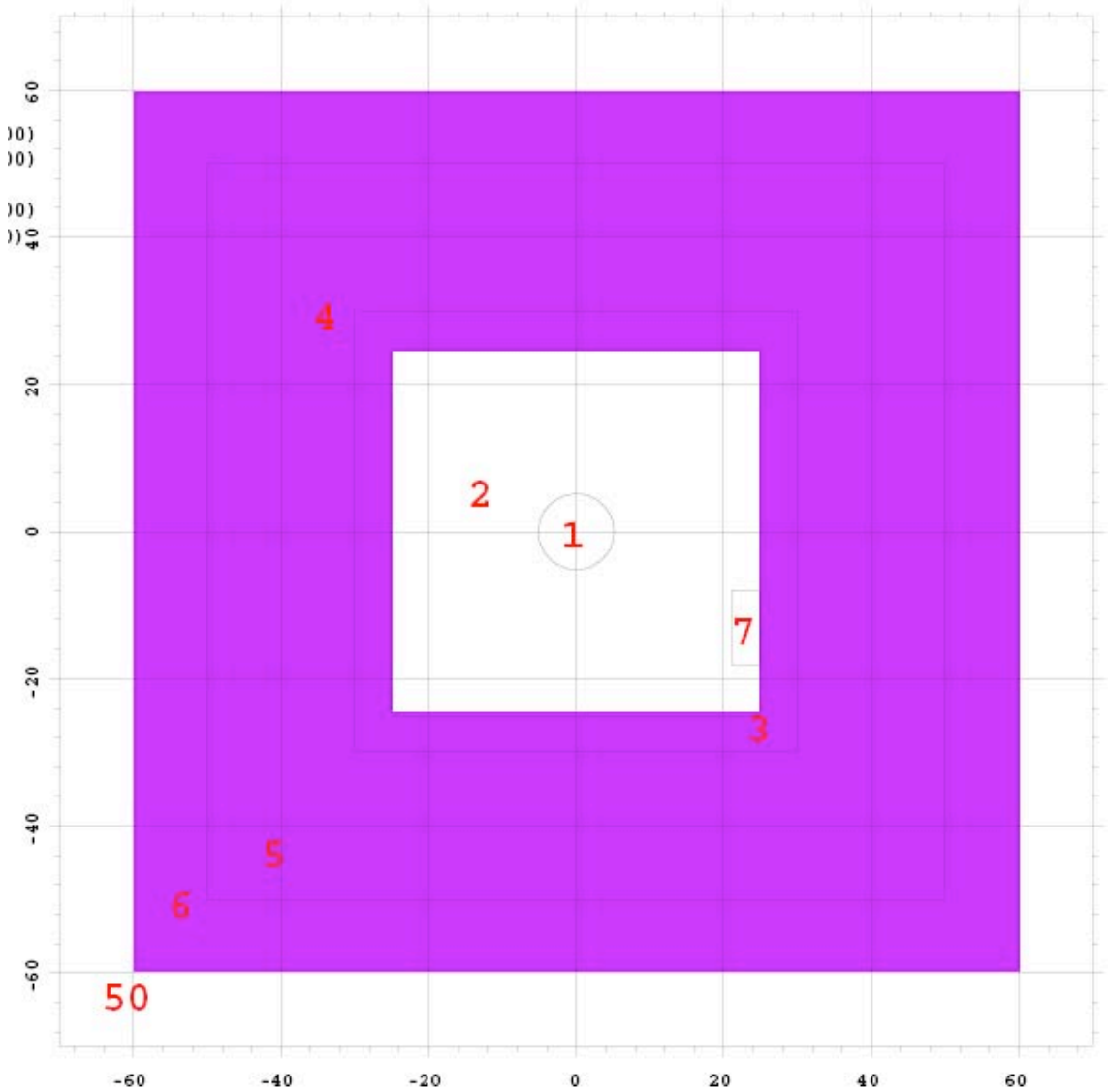
detector: 25% coaxial HPGe

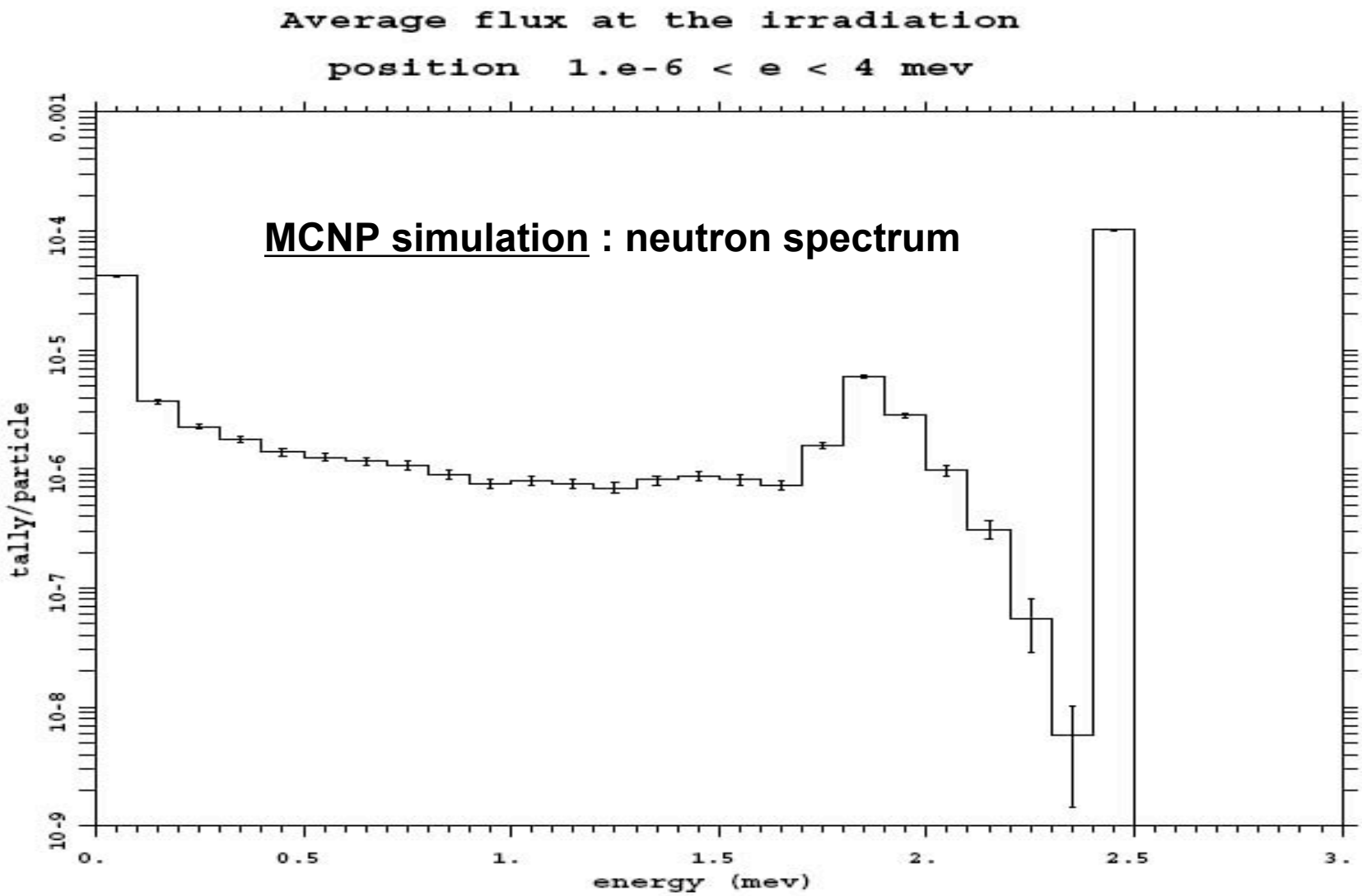
Electronics: DSPEC by Ortec

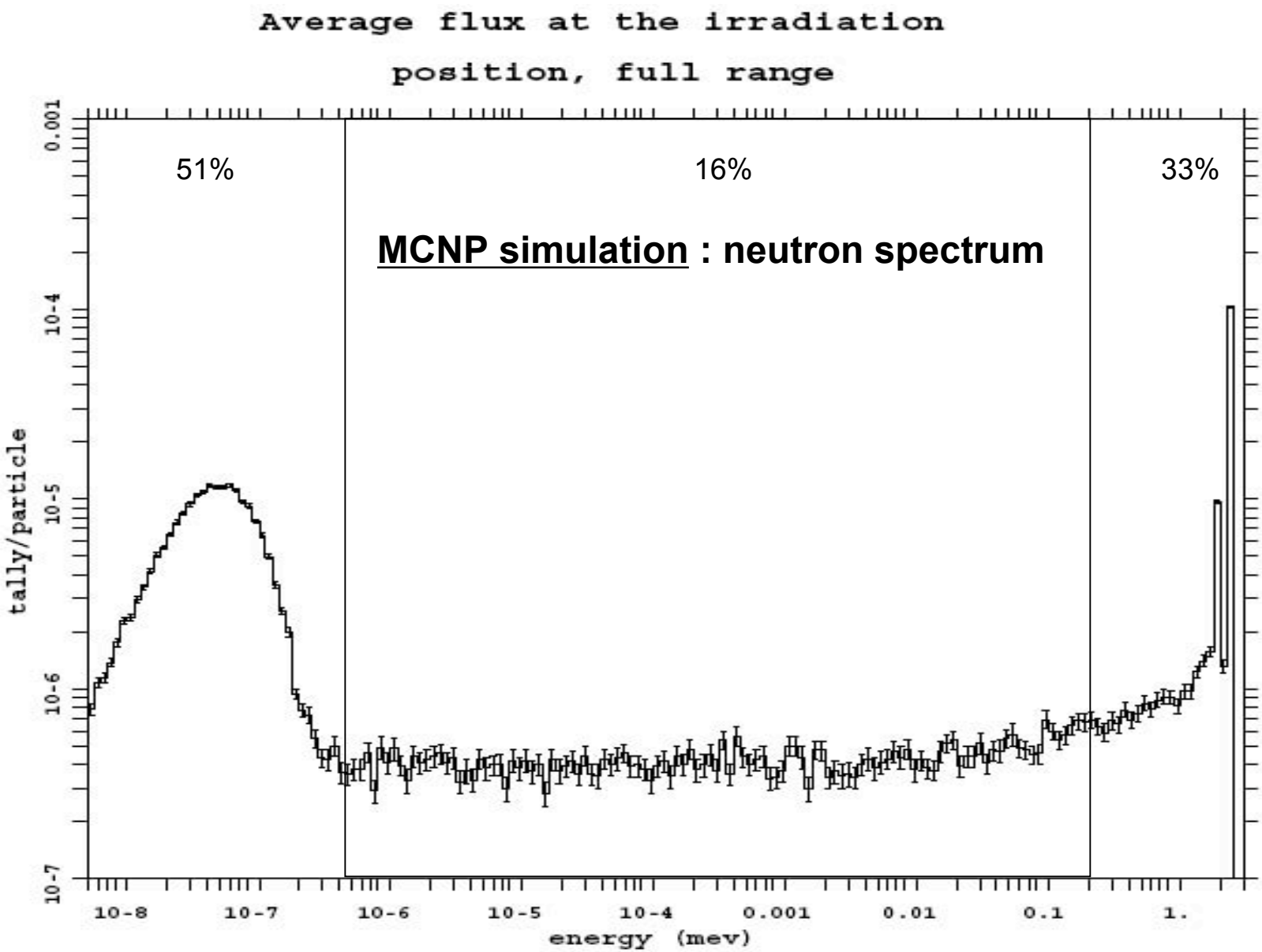
Acquisition: Gamma Vision by Ortec

OS: windows 2000

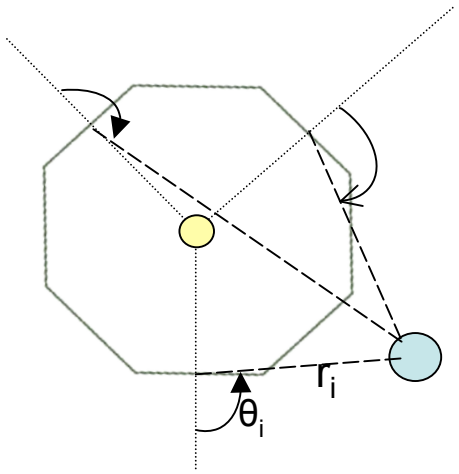
MCNP simulation : geometry

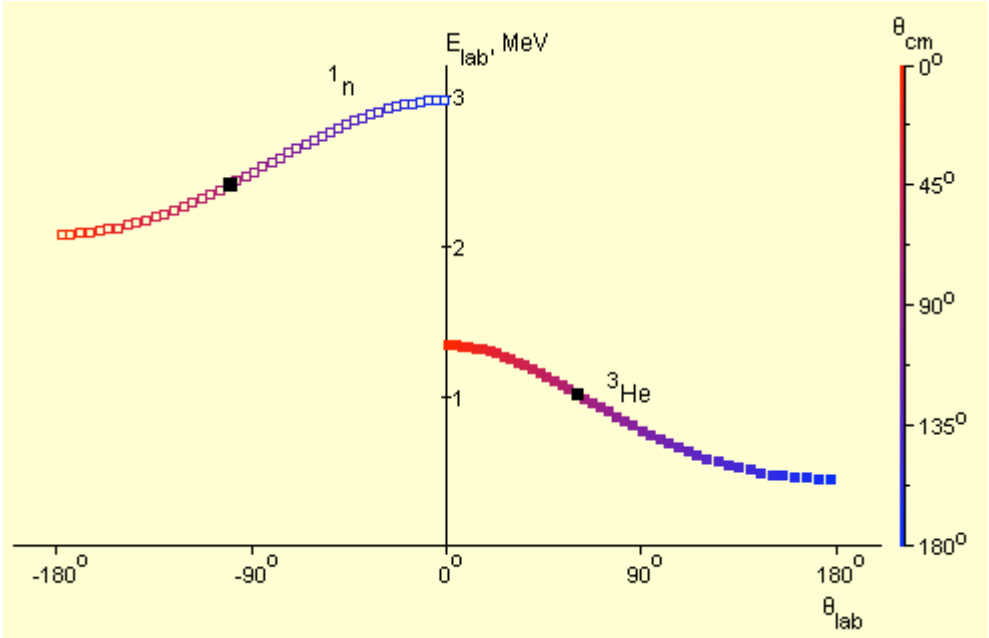



Average flux at the irradiation
position 1.e-6 < e < 4 mev
MCNP simulation : neutron spectrum
tally/particle
0.001
10-4
10-5
10-6
10-7
10-8
10-9
0.
0.5
1.
1.5
2.
2.5
3.
energy (mev)


Average flux at the irradiation position, full range
51%
16%
33%
MCNP simulation : neutron spectrum
tally/particle
0.001
10-4
10-5
10-6
10-7
10-8
10-7
10-6
10-5
10-4
0.001
0.01
0.1
1.
energy (mev)

Weighted average neutron energy

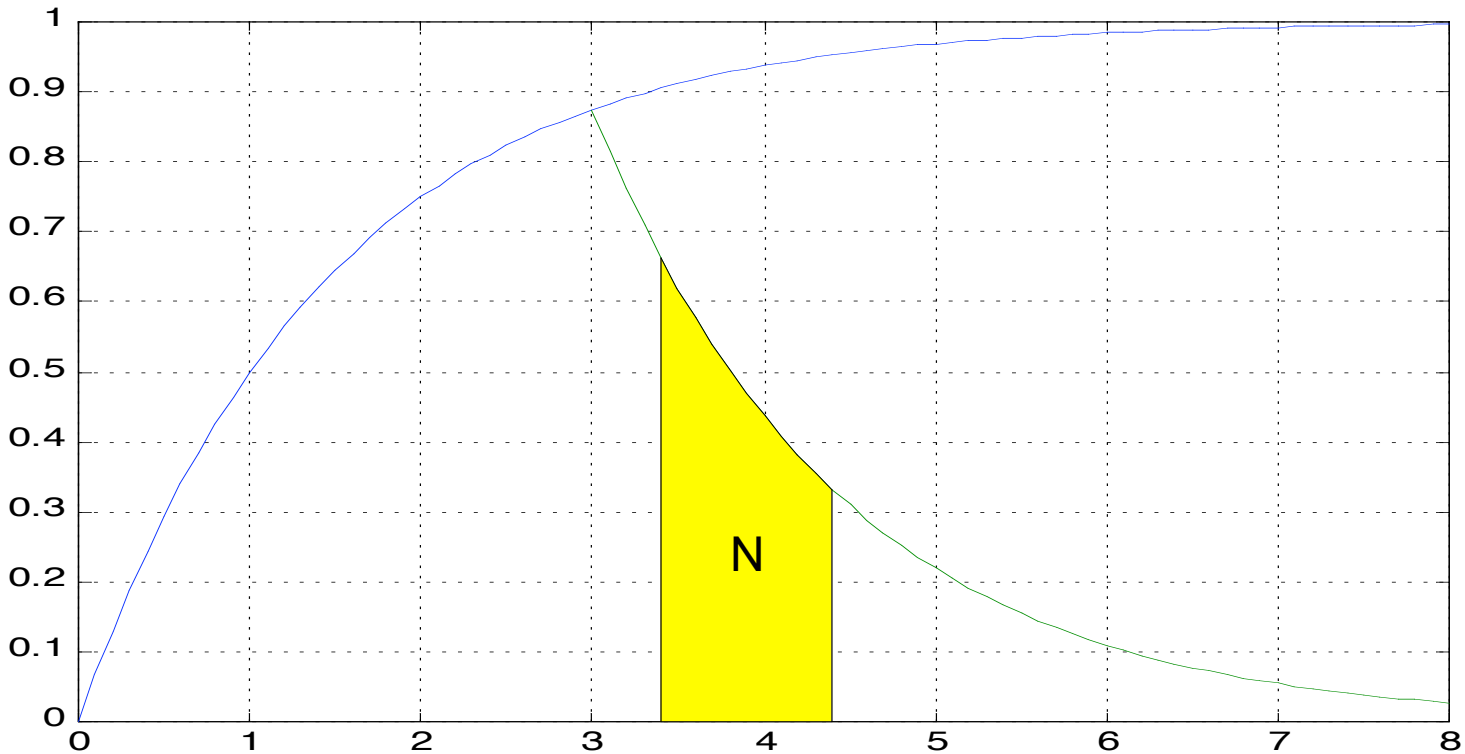




$$\langle E_n \rangle = \frac{\sum_1^8 \frac{1}{r_i^2} En(\theta_i)}{\sum_1^8 \frac{1}{r_i^2}} = 2.48 MeV$$

Activation / Detection procedure

$$A_{sat} = \phi n \sigma = \frac{\lambda N}{(1-e^{-\lambda t_{Irr}})e^{-\lambda t_{Irr}}(e^{-\lambda t_{AccStart}} e^{-\lambda t_{AccStop}})}$$



The Samples studied

Element	comp.	W[gr]	Diam.[cm]	Height [cm]	Density [g/cc]
Ag	AgNO3	52.0	30.0	22.0	3.35
Al	Al2O3	39.4	30.0	45.0	1.24
Ba	BaCO3	41.2	30.0	45.0	1.30
Br	KBr	40.0	30.0	30.0	1.89
Cd	CdF2	29.8	30.0	10.0	4.21
Hf	metal	6.9	5.0	13.0	27.16
Hg	Hg	498.8	30.0	50.0	13.55
In	metal	1.4	10.0	1.0	17.24
Se	Se	15.9	30.0	14.0	4.70
Te	Te	30.5	22.0	14.0	6.24
V	V2O5	46.2	30.0	47.0	1.39
W	WC	254.1	30.0	42.0	8.56
Y	metal	50.9	25x21	25.0	4.47
Zn	ZnO	12.2	30.0	20.0	0.86

Data taking

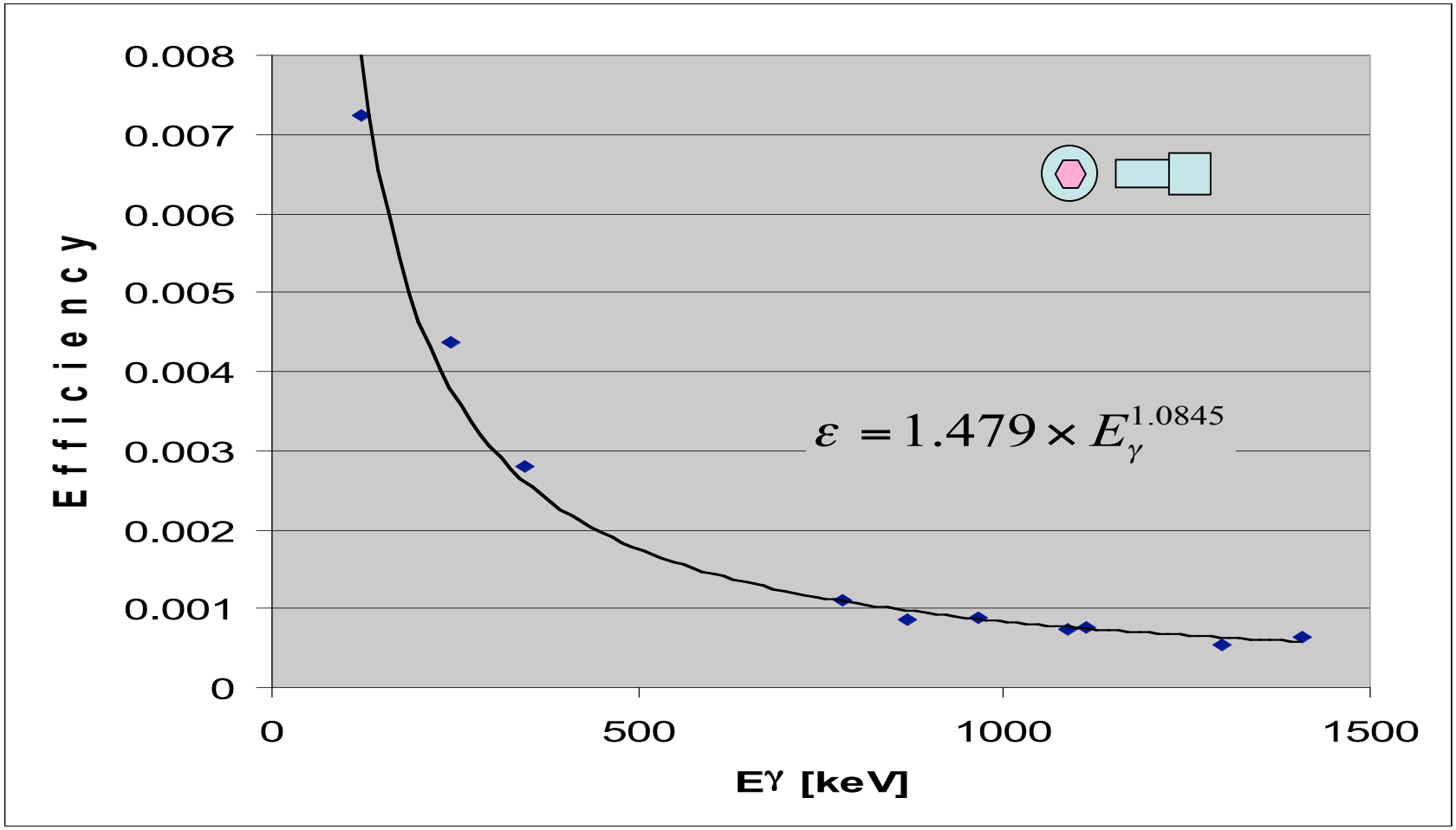
- Background optimization (~3 cps @ 50-3000 keV)
- Detection efficiency calibrated with a ^{152}Eu source **(0.12 – 1.4 MeV)**
- ^{115}In(n,n'γ)^{115}In (4.3h) data used to estimate fast n flux
- ^{115}In(n,γ)^{116}In (54.2m) data used to estimate thermal n flux
- Irradiate samples for 4-8 $T_{½}$'s (n-generator limitations)
- Accumulate γ spectra for ~ 6-10 $T_{½}$'s, saving data every ~½-¼ $T_{½}$
- Irradiate with and without Cadmium cover
- Repeat the above 2 steps if necessary
- Record irradiation/accumulation times
- Record mR/h meter reading continuously for n-yield monitoring (verified by frequent Indium irradiation measurements)

Data analysis

- Identify all γ line (energies, isotopes and transitions).
- Evaluate net peak area and apply corrections for:
  - Isotope Abundance
  - γ Branching
  - γ Detection efficiency
  - γ self absorption
  - n-yield (mR meter)
- Verify identification by $T_{½}$ determination
- Evaluate $A_{sat} = \Phi n \sigma$ for the given irradiation and accumulation times.
- Evaluate cross section on the basis of known n and Φ (from In).
- Compare to data bases (ENDF, JENDL) and exp. data

Full energy peak efficiency (± 10-15%)

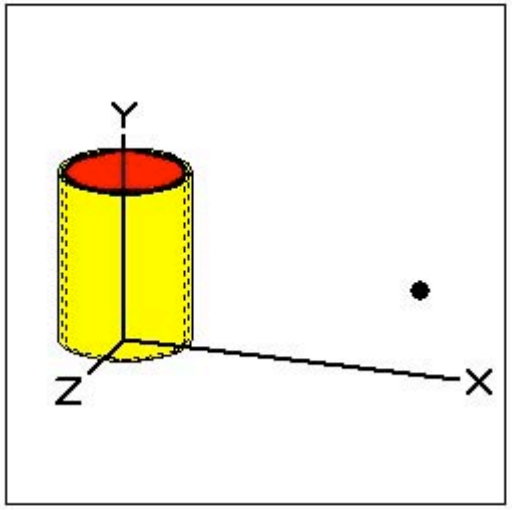
(^{152}Eu calibration)



Self absorption Correction

File Ref: _____
Date: _____
By: _____
Checked: _____

Case Title: agno3
Description: Case 2
Geometry: 7 - Cylinder Volume - Side Shields



Source Dimensions

Height	4.5 cm	1.8 in
Radius	1.5 cm	0.6 in

Dose Points

A	X	Y	Z
# 1	8 cm	2.25 cm	0 cm
	3.1 in	0.9 in	0.0 in

Shields

Shield Name	Dimension	Material	Density
Source	31.809 cm³	BaCO3	1.3
Shield 1	.1 cm	Cd	8.65
Transition		Air	0.00122
Air Gap		Air	0.00122
Wall Clad	.1 cm	Plastic	0.9

Source Input
Grouping Method : User Defined Energies

Group #	Energy (MeV)	Activity Photons/sec	Volume Source Photons/sec/cm³	% Energy Activity
1	0.6617	1.0000e+000	3.1438e-002	100.000

Buildup
The material reference is : Transition

Integration Parameters

Radial	10
Circumferential	10
Y Direction (axial)	20

Point source geometry: 5.84e-04 Mev/cm^2/sec

'net' self Abs factor : 0.87

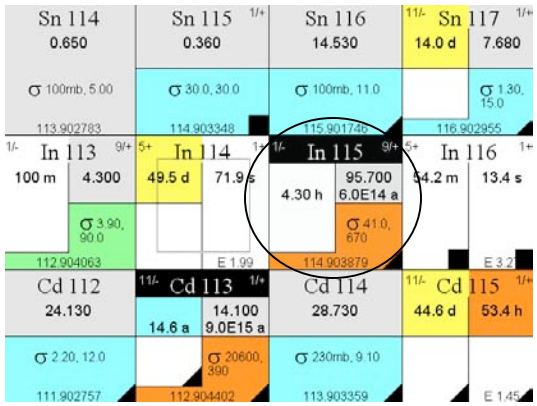
Results

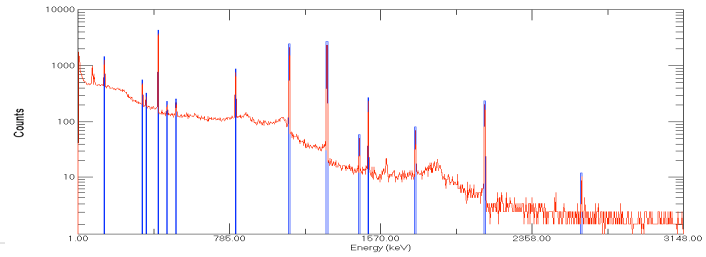
Energy MeV	Activity photons/sec	Fluence Rate MeV/cm²/sec No Buildup	Fluence Rate MeV/cm²/sec With Buildup	Exposure Rate mR/hr No Buildup	Exposure Rate mR/hr With Buildup
0.6617	1.000e+00	6.674e-04	7.928e-04	1.294e-06	1.537e-06
TOTALS:	1.000e+00	6.674e-04	7.928e-04	1.294e-06	1.537e-06

Self absorption Correction

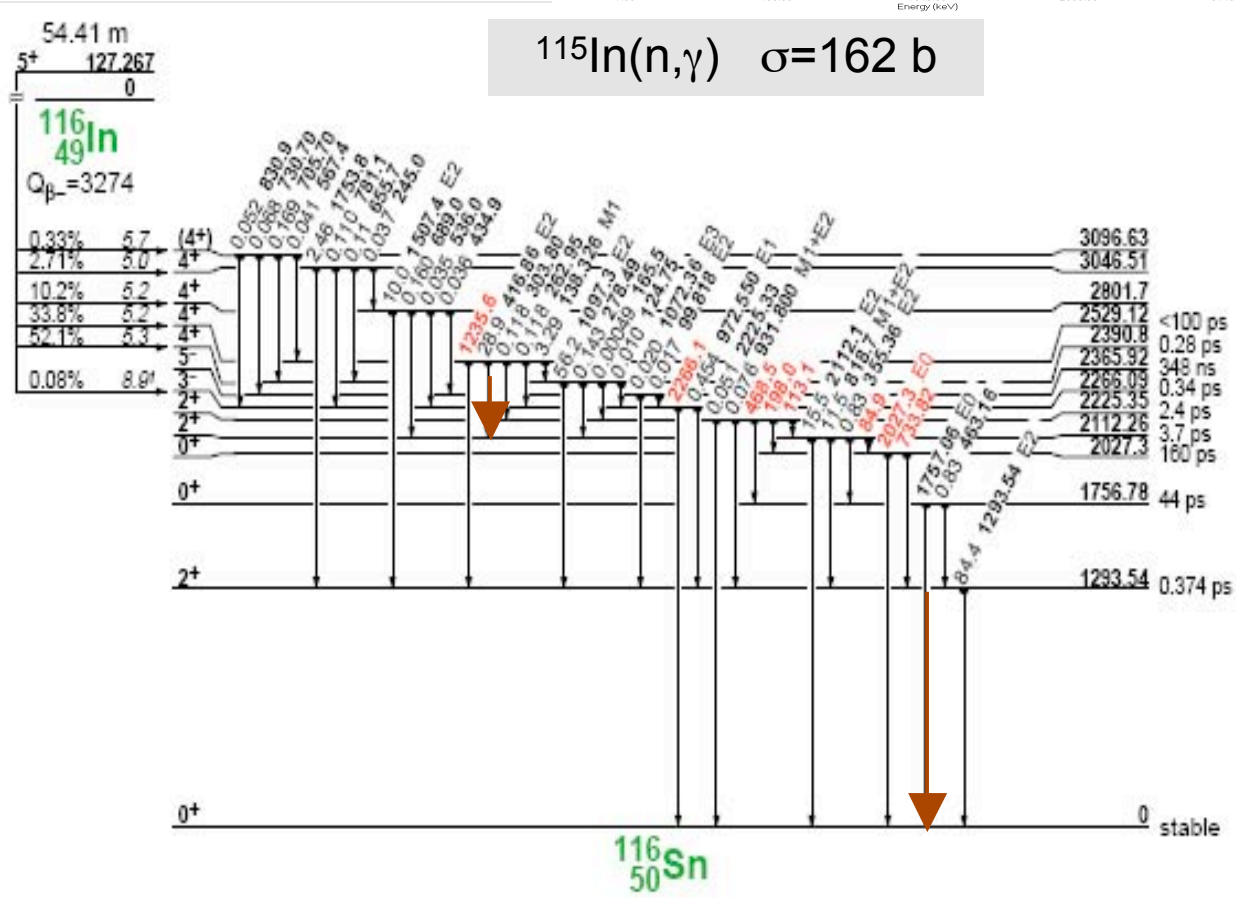
Element	γ energy	SelfAbs
Ag	93	0.13
Ag	632.9	0.73
Ag	657.8	0.74
Ba	661.7	0.87
Br	207	0.75
Cd	150.8	0.28
Cd	245.4	0.5
Hf	214	0.246
Hf	215	0.248
Hf	333	0.48
Hf	443	0.61
Hg	158.4	0.023
In	336	0.68
Se	162	0.38
Te	151	0.21
Te	456	0.71
Y	909	0.58

Indium measurement

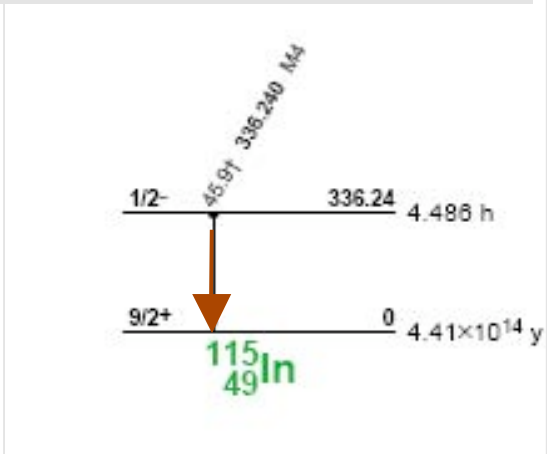




^{115}In(n,γ) σ=162 b

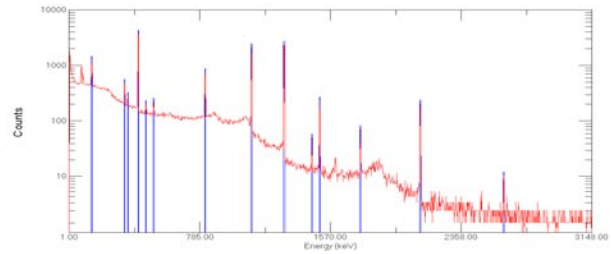


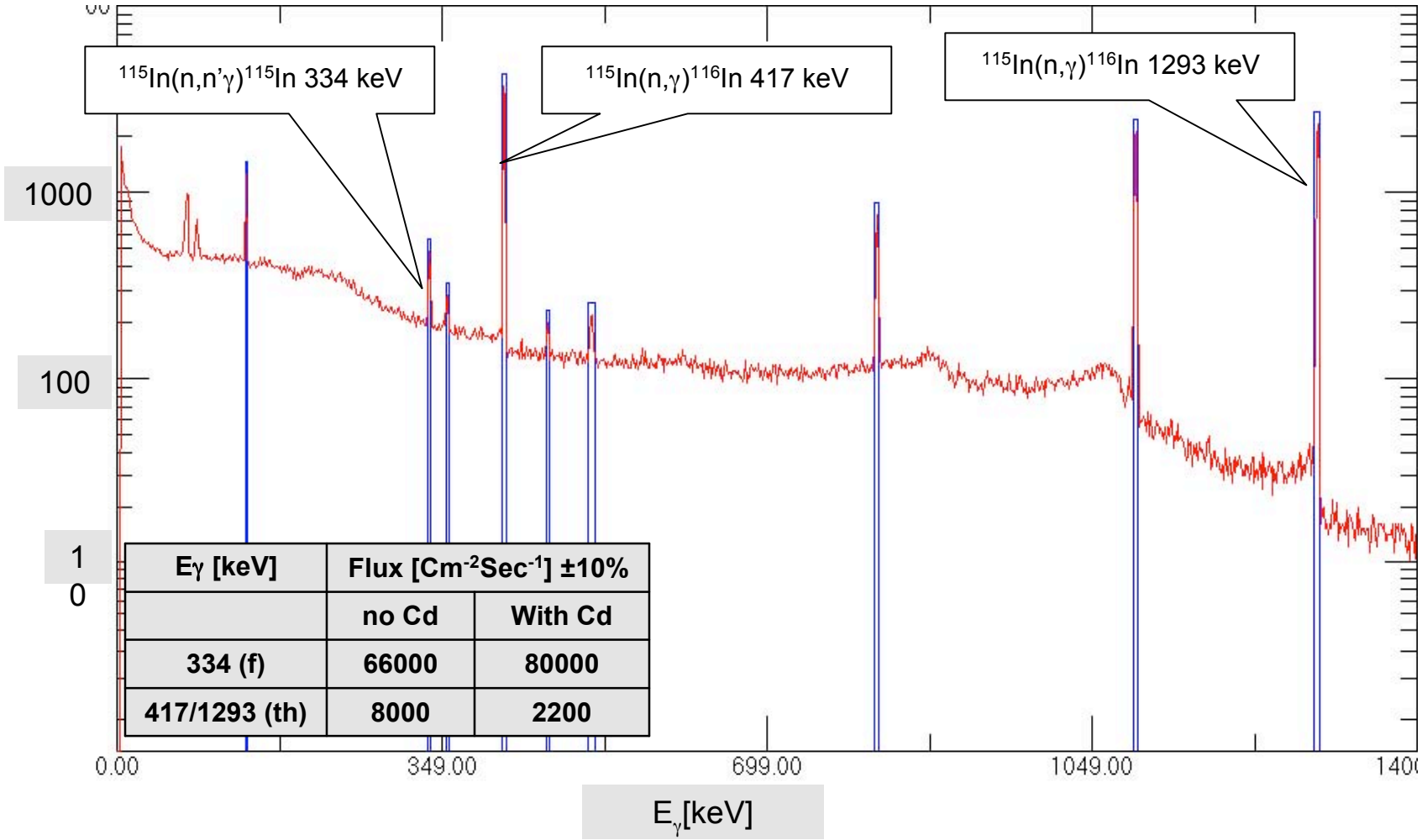
^{115}In(n,n'γ) σ= 344 mb



Indium measurement

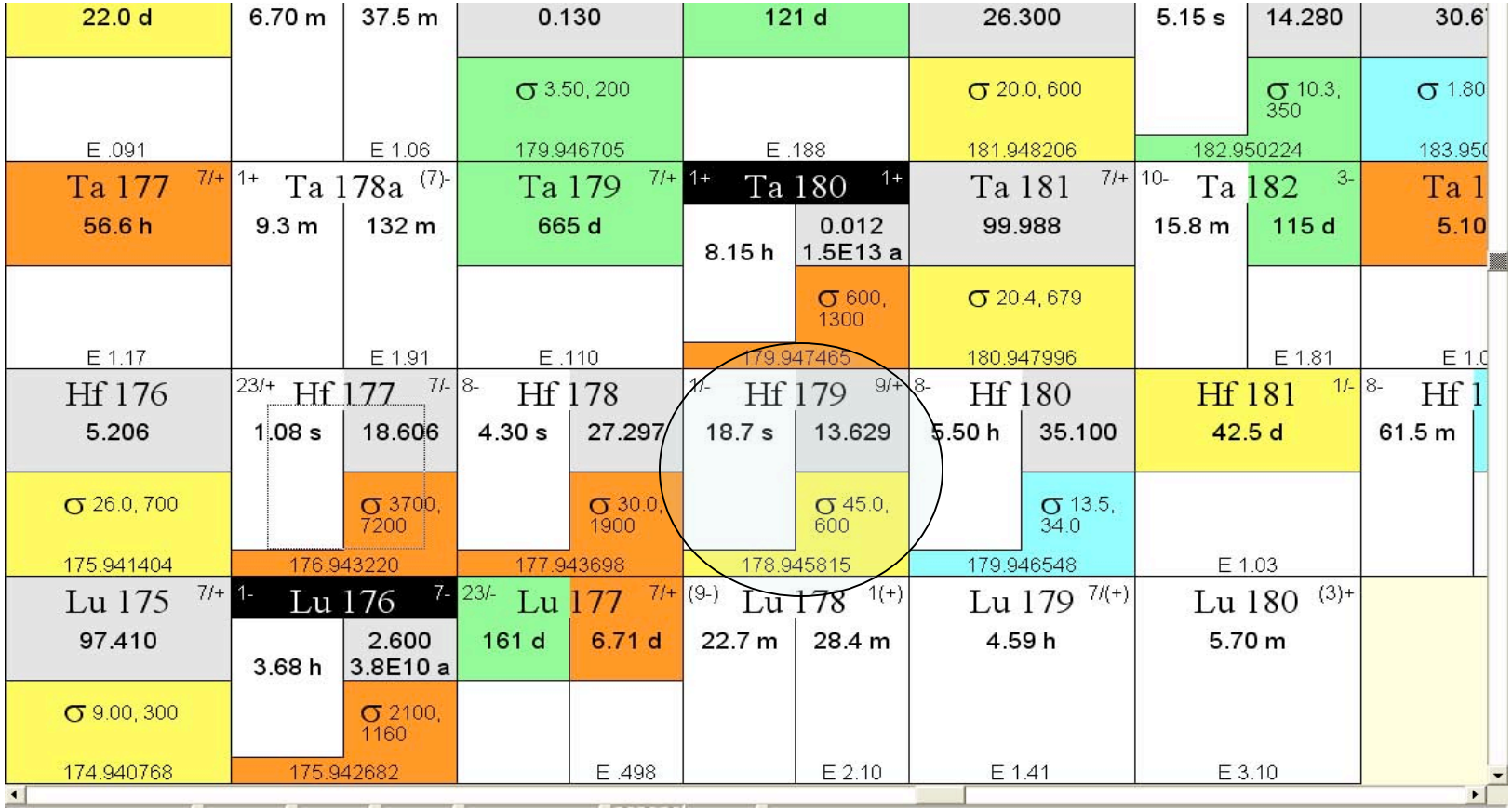
lt=50000 sec



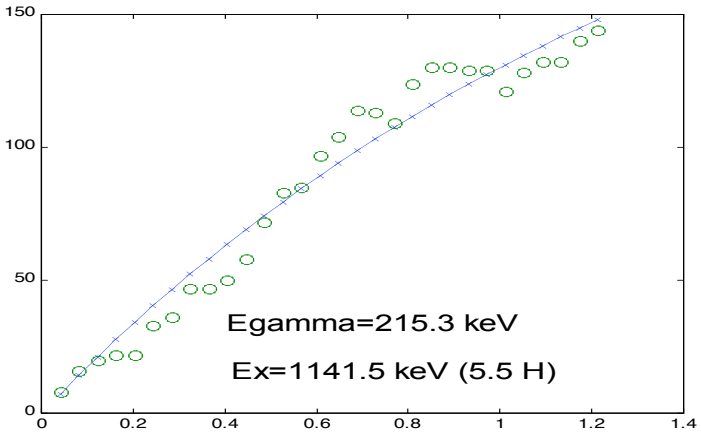


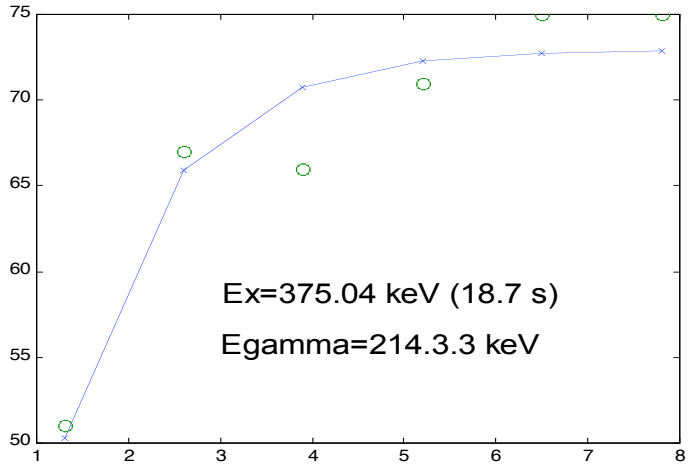
Eγ [keV]	Flux [Cm^{-2}Sec^{-1}] ±10%	
	no Cd	With Cd
334 (f)	66000	80000
417/1293 (th)	8000	2200

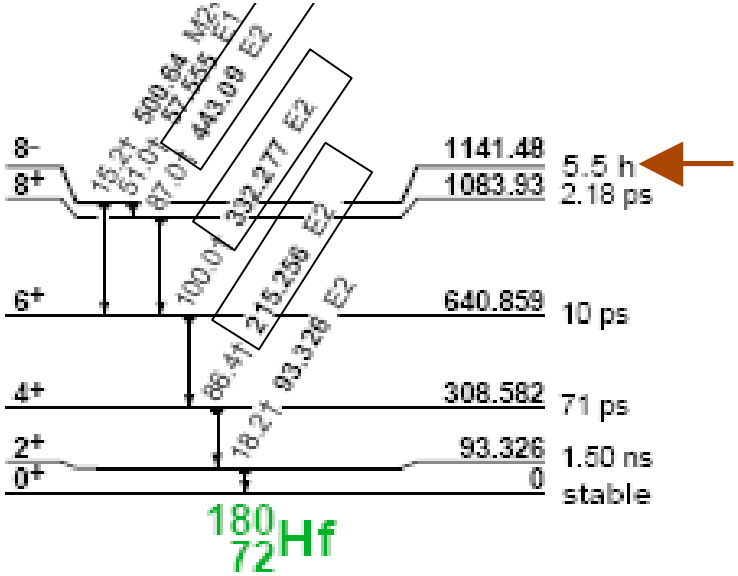
Identification conflicts (1)

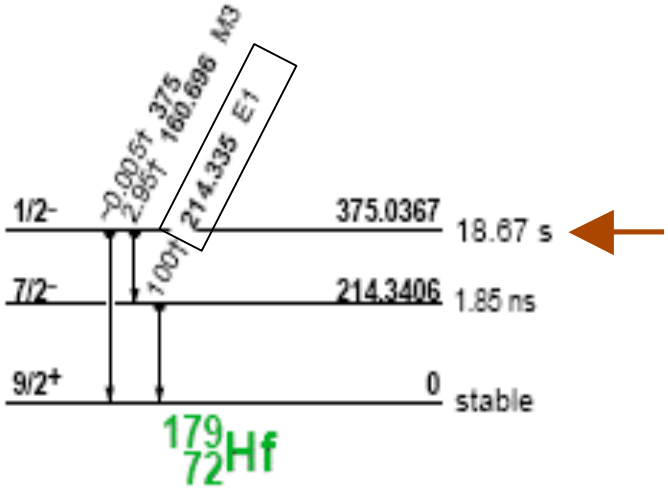


Identification conflicts (2)









Combined (n,n'γ)+(n,γ) transition

$$A_{sat} = \sum_A n_A \int \phi(E)\sigma(E)dE \approx \phi_{th} n_{A-1}\sigma_{n,\gamma} + \phi_f n_A \sigma_{n,n'\gamma}$$

No Cd: $$A_{sat} = \phi n \sigma = 8000 n_{A-1}\sigma_{n,\gamma} + 66000 n_A \sigma_{n,n'\gamma}$$

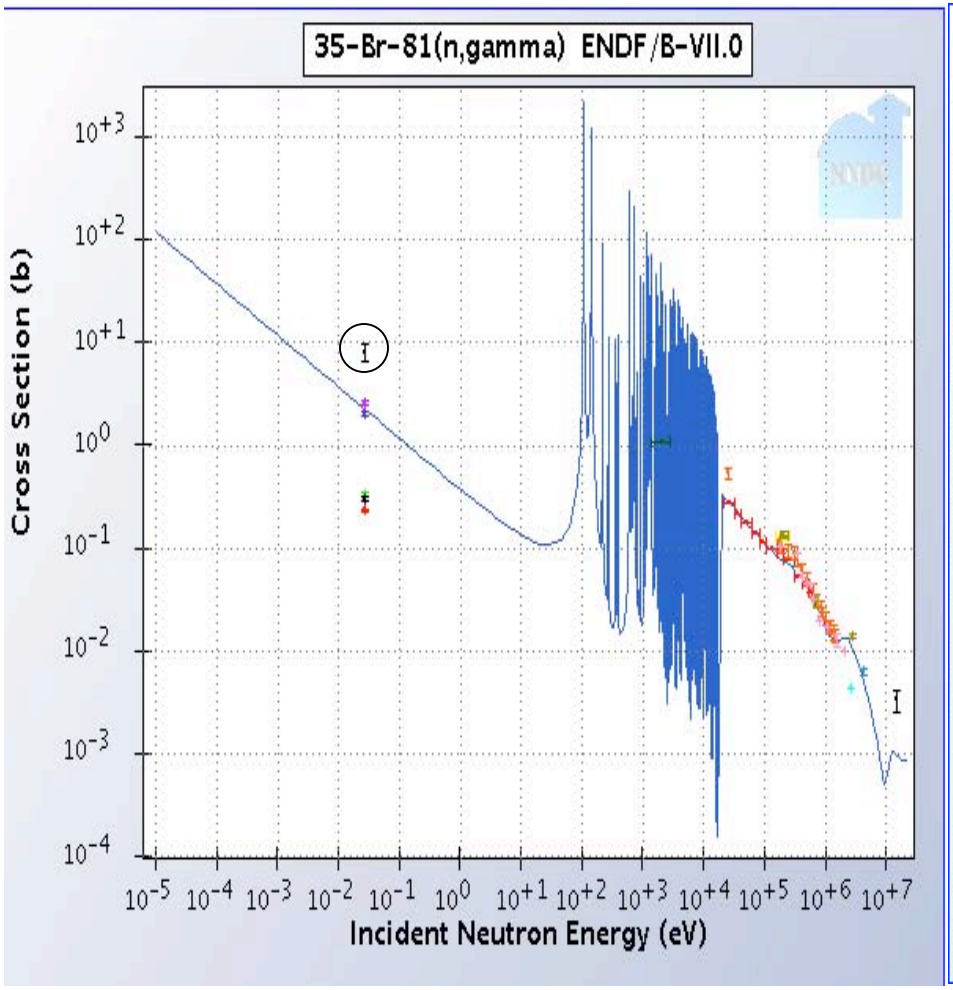
With Cd: $$A_{sat} = \phi n \sigma = 2200 n_{A-1}\sigma_{n,\gamma} + 80000 n_A \sigma_{n,n'\gamma}$$

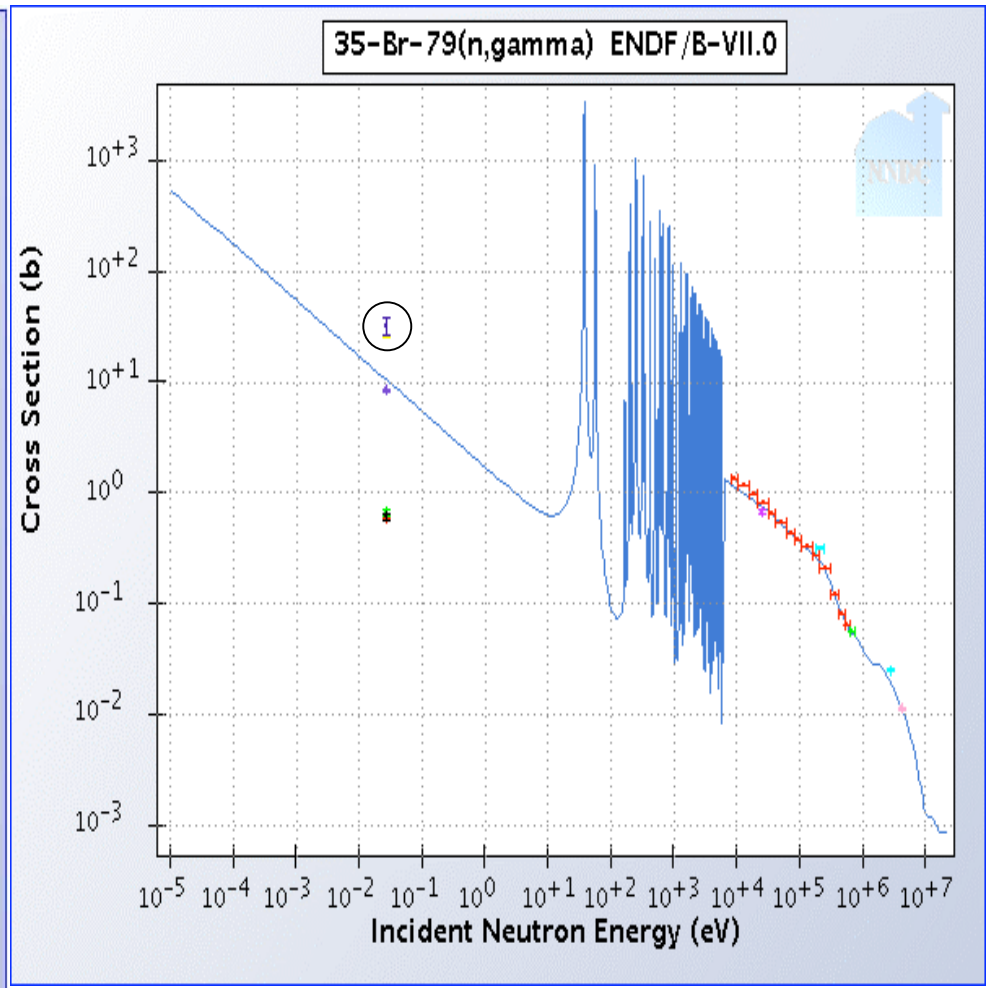
... Pure transitions exist

(n,γ) transitions

Pure
combined

A		Eγ	σ	δσ	ENDF/B-VII
		keV	barns		barns
27	Al	1779	0.46	0.10	0.23
51	V	1434	7.9	2.0	4.9
76	Se	162	55.0	15.0	22
79	Br	666	33.6	5.1	10.3
81	Br	554	8.6	1.3	2.4
81	Br	776	8.4	1.2	2.4
107	Ag	633	34.0	5.0	37.6
109	Ag	658	82	13	86
110	Cd	150	0.7	0.5	0.14
110	Cd	245	0.7	0.5	0.14
115	In	417	161.0		162
115	In	1293	161.0		162
136	Ba	662	0.93	0.80	0.01
178	Hf	214	45.0	15.00	53.0
179	Hf	332	0.75	0.30	0.445
179	Hf	443	0.75	0.30	0.445
198	Hg	158	0.018		0.018
198	Hg	374	0.018		0.018


35-Br-81(n,gamma) ENDF/B-VII.0
Cross Section (b)
Incident Neutron Energy (eV)


35-Br-79(n,gamma) ENDF/B-VII.0
Cross Section (b)
Incident Neutron Energy (eV)

(n,n'γ) transitions

Pure
combined

A		Eγ	σ	δσ	ENDF/B-VII	Nagoya 2004
		keV	barns		barns	barns
77	Se	162	2.50	1.00	1.3	1.3
79	Br	207	0.20	0.04	0.4	0.28
89	Y	909	0.10	0.02	0.4	0.37
107	Ag	93	0.9	0.2	0.8	0.9
111	Cd	150	0.40	0.06	0.2	0.24
111	Cd	245	0.30	0.10	0.2	0.24
115	In	336.2	0.34		0.34	0.34
137	Ba	662	0.40	0.10	0.75	0.4
179	Hf	214	1.30	0.70	0.65	0.63
180	Hf	332	0.03	0.03	0.003	0.002
180	Hf	443	0.03	0.03	0.003	0.002
199	Hg	158	0.004	0.001	0.0055	
199	Hg	374	0.004	0.001	0.0055	

LLD evaluation

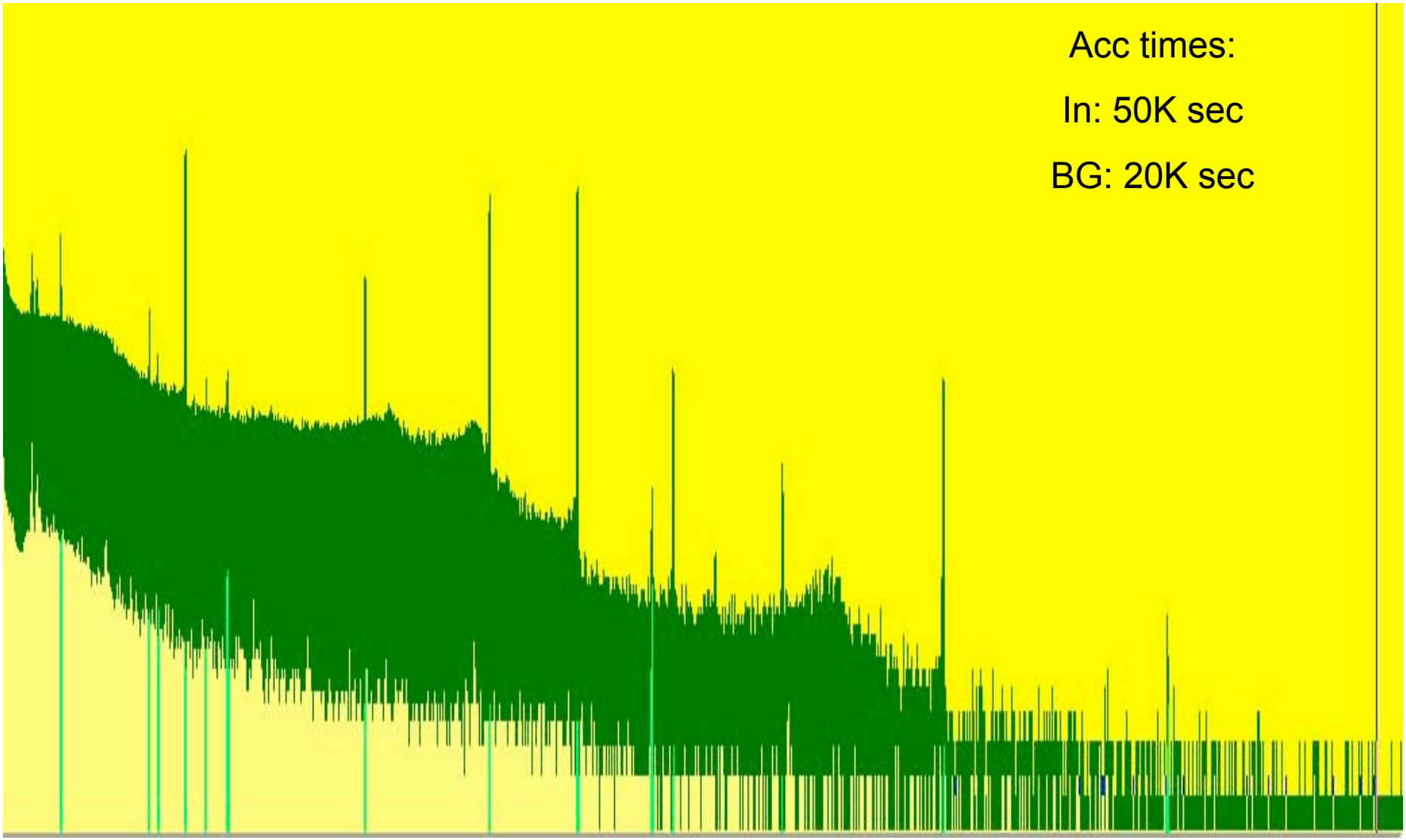
activation and accumulation for 3 half lives

$$Cnts(3T_{1/2}) = 0.766 \times N_{sat} = \frac{0.766 \times N(t_{irr}, t_{acc})}{SelfAbs \times Decay(irr) \times Decay(acc)}$$

$$LLD = \frac{3\sigma_{BG}}{Cnts(3T_{1/2})} \times W_{sample}$$

- Background rate was evaluated for each γ line separately

Indium target / background spectra



Lowest Level of Detection

A	LLD[mg]	Reaction
27 Al	2	n,g
51 V	5	n,g
77 Se	6	n,n'
79 Br	16	n,n'
89 Y	42	n,n'
107 Ag	40	n,n'
111 Cd	125	n,n'
115 In	1	n,n'
137 Ba	235	n,n'
179 Hf	4	n,n'
199 Hg	130	n,n'

Estimated detection limits for INAA using decay gamma rays

(irradiation in a reactor neutron flux of 1×10^{13} n/cm^{-2}s^{-1})

Sensitivity (mg)	Elements
1e-3	Dy, Eu
1e-3 – 0.01	**In**, Lu, Mn
0.010 – 0.1	Au, Ho, Ir, Re, Sm, W
0.1 – 1	**Ag**, Ar, As, **Br**, Cl, Co, Cs, Cu, Er, Ga, Hf, I, La, Sb, Sc, **Se**, Ta, Tb, Th, Tm, U, **V**, Yb
1 – 10	**Al**, **Ba**, **Cd**, Ce, Cr, **Hg**, Kr, Gd, Ge, Mo, Na, Nd, Ni, Os, Pd, Rb, Rh, Ru, Sr, Te, Zn, Zr
10 – 100	Bi, Ca, K, Mg, P, Pt, Si, Sn, Ti, Tl, Xe, **Y**
100 - 1000	F, Fe, Nb, Ne
10000	Pb, S

What's next

- n-generator
  - 'real' online flux monitor (FC, α det.)
  - Irradiation setup at flux maximum
  - Optimal 'fast beam' setups
  - Optimal thermal flux position
- Detection stand
  - Anti-Compton active shield
  - Dedicated passive shielding
- Samples
  - Stoichiometrically favorable samples
  - Various enrichments
  - Common 'friendly' geometry

Summary

- Durable and friendly setup (most of the time)
- 13 (n,n') and 18 (n,γ) cross sections were evaluated for ~2.5 MeV neutrons
- Current Setup capabilities:
  - cross sections in the tens mb region
  - half lives as low as 5 seconds
  - LLD's in the few mg range
- Performance may be largly improved