A METHOD OF SPALLATION NEUTRON ENERGY SPECTRUM RECONSTRUCTION WITH YTTRIUM SAMPLE ACTIVATION

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Purpose of the work

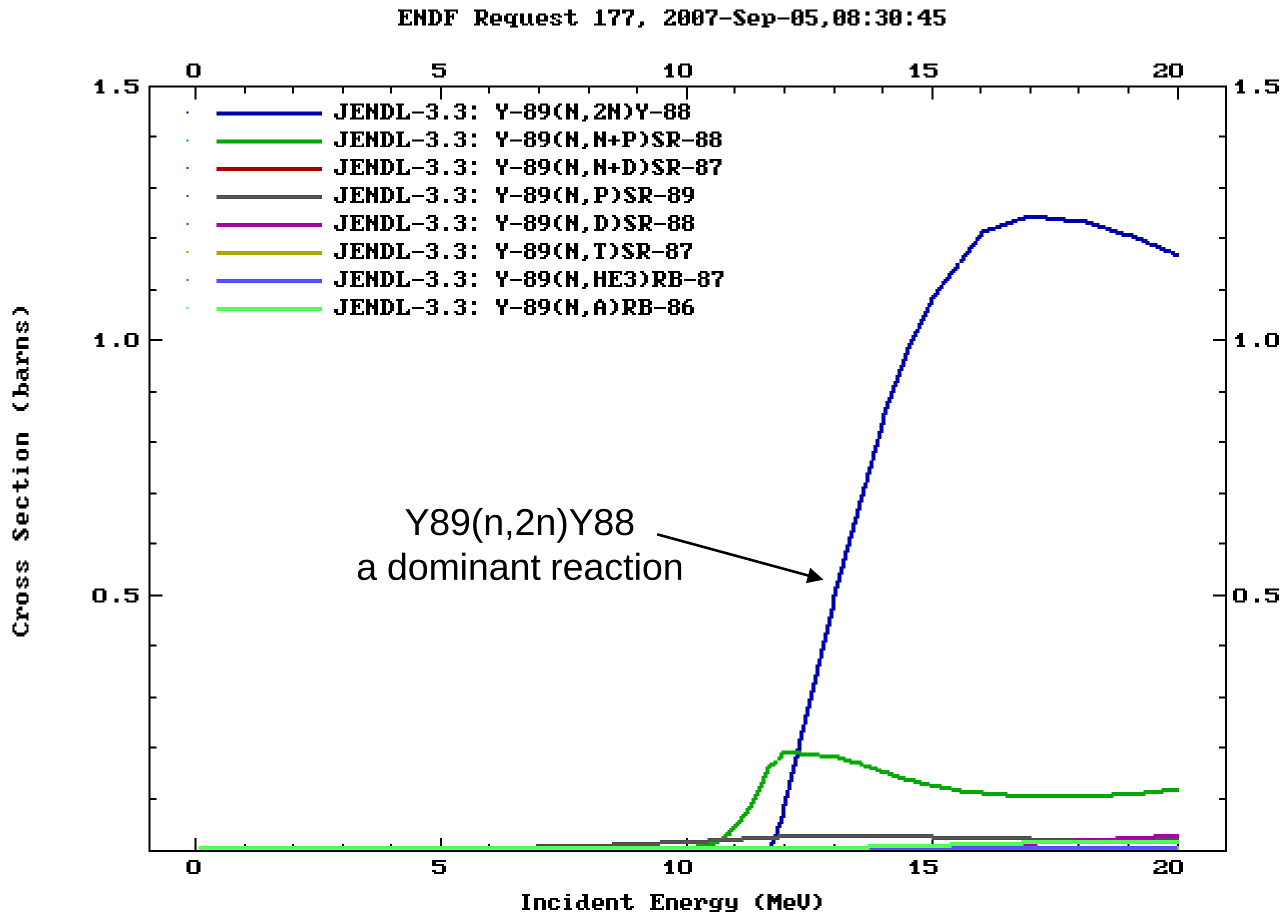
Looking for possibility to determine spallation neutron energy spectrum with **Y89** activation detector

Why yttrium?

Y89 the only one naturally occurring isotope
– no overlapping reactions
– easy to trace
- several residual nuclei

Resulting isotopes relatively easy to identify

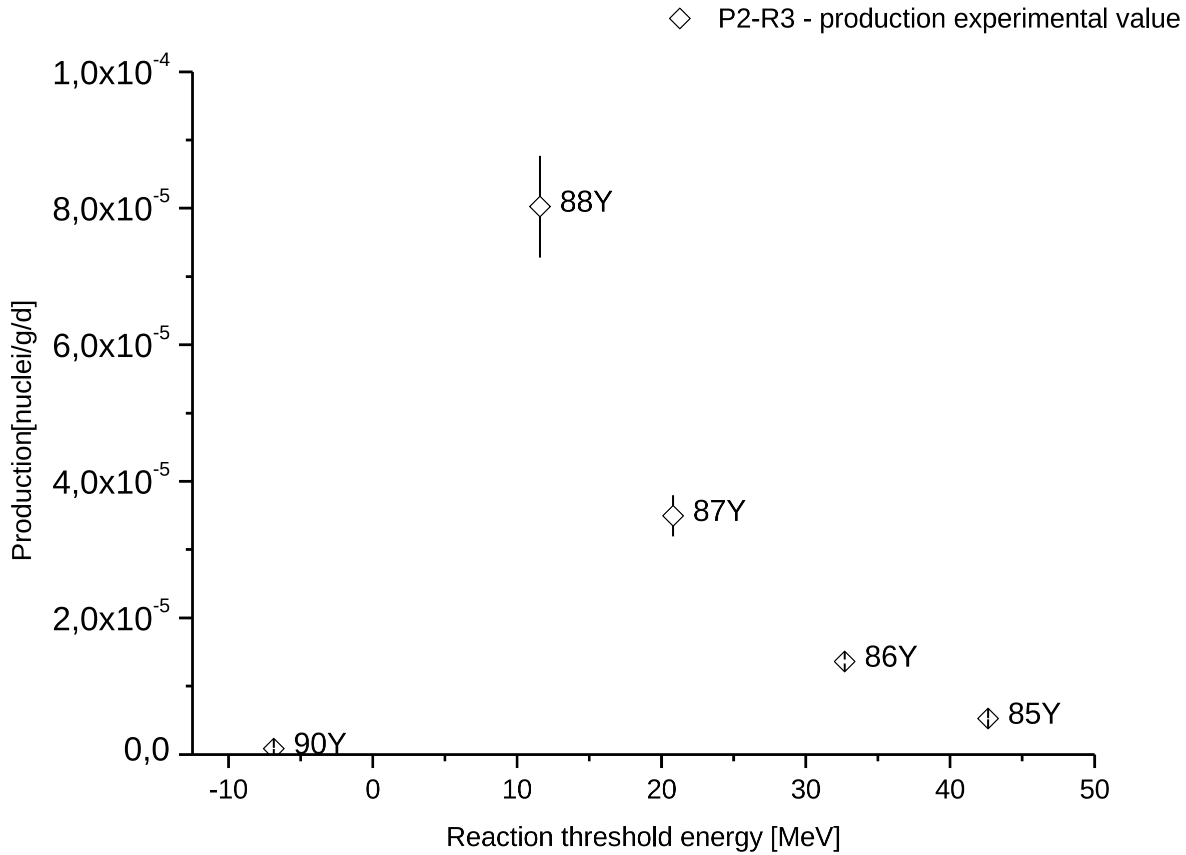
Y-89(n,?) reaction cross sections available in ENDFs



Yttrium-89 activation reactions taken into account

Reaction	Produced Isotope	T1/2	Reaction Threshold [MeV]	γ-line Energy [keV]	γ-line Intensity [%]
Y89(n,g)	Y90	3.19h	-6.8570*	202.51	97.3
				479.17	90.74
Y89(n,2n)	Y88	106,65d	11,5	898.042	93.7
				1836.063	99.2
Y89(n,3n)	Y87	79,8h	20,8	388.53	82.00
				484.805	89.7
Y89(n,4n)	Y86	14,74h	32,7	1076.64	82.00
Y89(n,5n)	Y85	2.68h	42.633	231.67	84.00
		4.86h	42.633	231.67	22.8

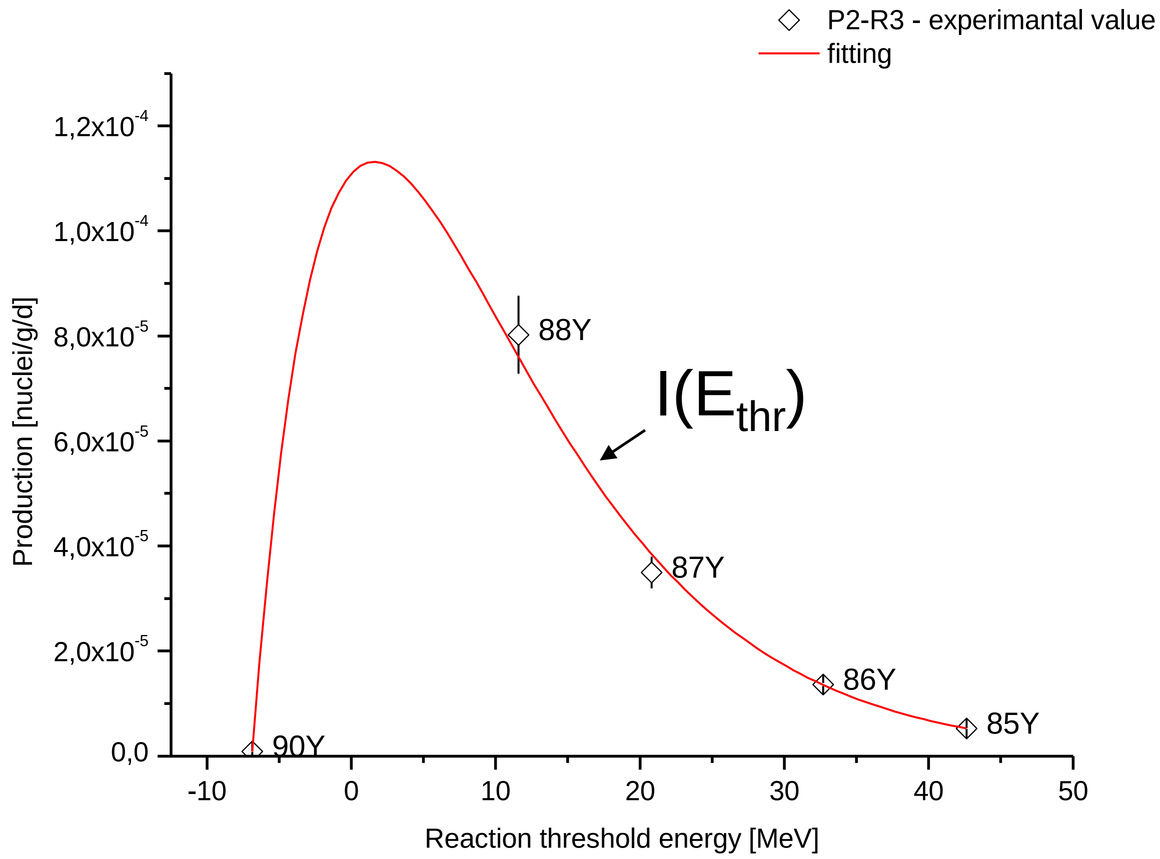
Y89(n,xn) reaction residual nuclei example production. Here is, where the method idea comes from.



$$I_k = Nt_{ir} \int_{E_{thr,k}}^{\infty} \varphi(E)\sigma_k\left(E, E_{thr,k}\right)dE$$

k = Y90, Y88, Y87, Y86, Y85

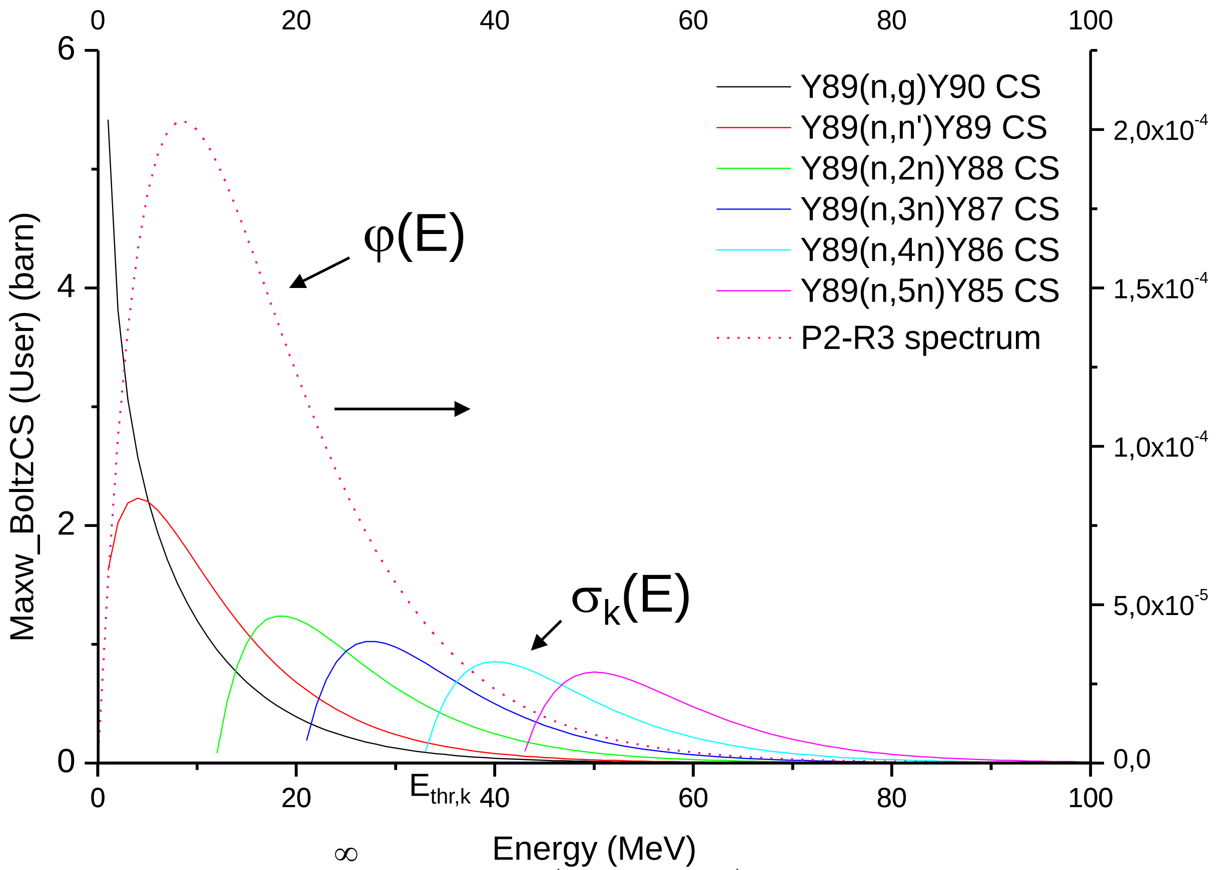
Y89(n,xn) reaction residual nuclei example production – remarks



1. The experimental values make a curve $I(E_{thr})$
2. For the $I(E_{thr})$ curve the E_{thr} is a variable

The method idea

Each isotope production is a result of convoluted influence of spallation neutron spectrum and reaction cross section



$$I_k = Nt_{ir} \int_{E_{thr,k}}^{\infty} \varphi(E)\sigma_k\left(E, E_{thr,k}\right)dE$$

Here the $E_{thr,k}$ is a value

The method idea

The idea is to transform one isotope production formula

$$I_k = N t_{ir} \int_{E_{thr,k}}^{\infty} \varphi(E) \sigma_k (E, E_{thr,k}) dE$$

$[I_k]$ - nuclei/g/d
$[N]$ - Y89 nuclei/g
$[t_{ir}]$ – s
$[\varphi(E)]$ – n/cm^2/s/MeV/d
$[\sigma_k]$ – barn $= 10^{-24}$ cm^2

or

$$I_k \xrightarrow{\varphi(E) t_{ir} \to \phi(E)} N \int_{E_{thr,k}}^{\infty} \phi(E) \sigma_k (E, E_{thr}) dE$$

$[\phi(E)]$ – neutron fluence/cm^2/MeV/d

into a general one valid for any isotope

$$I(E_{thr}) = N \int_{E_{thr}}^{\infty} \phi(E) \sigma(E, E_{thr}) dE$$

Here the E_{thr} is a variable

and solve it.

The generalized equation

$$I(E_{thr}) = N \int_{E_{thr}}^{\infty} \phi(E)\sigma(E, E_{thr})dE$$

$\phi(E)$ looked for, neutron fluence/cm^2/MeV/g/d = n/cm^2/MeV/g/d

$\phi(E)$ is in fact $\phi(\vec{r}, E)$ but dependence on r is not a subject

E_{thr} – a variable parameter

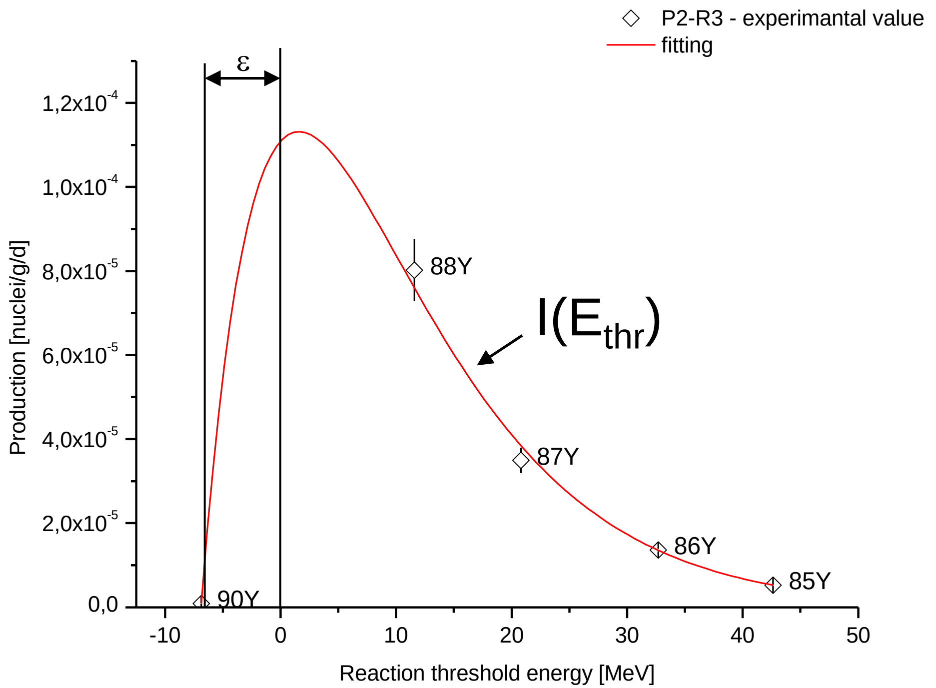
$I(E_{thr})$ and $\sigma(E,E_{thr})$ to be known and continuous functions.

$\phi(E) \rightarrow 0$ while $E \rightarrow \infty$

The equation creates two issues

- How to find an analytical form of $I(E_{thr})$
- Need to assume something about the $\sigma(E,E_{thr})$

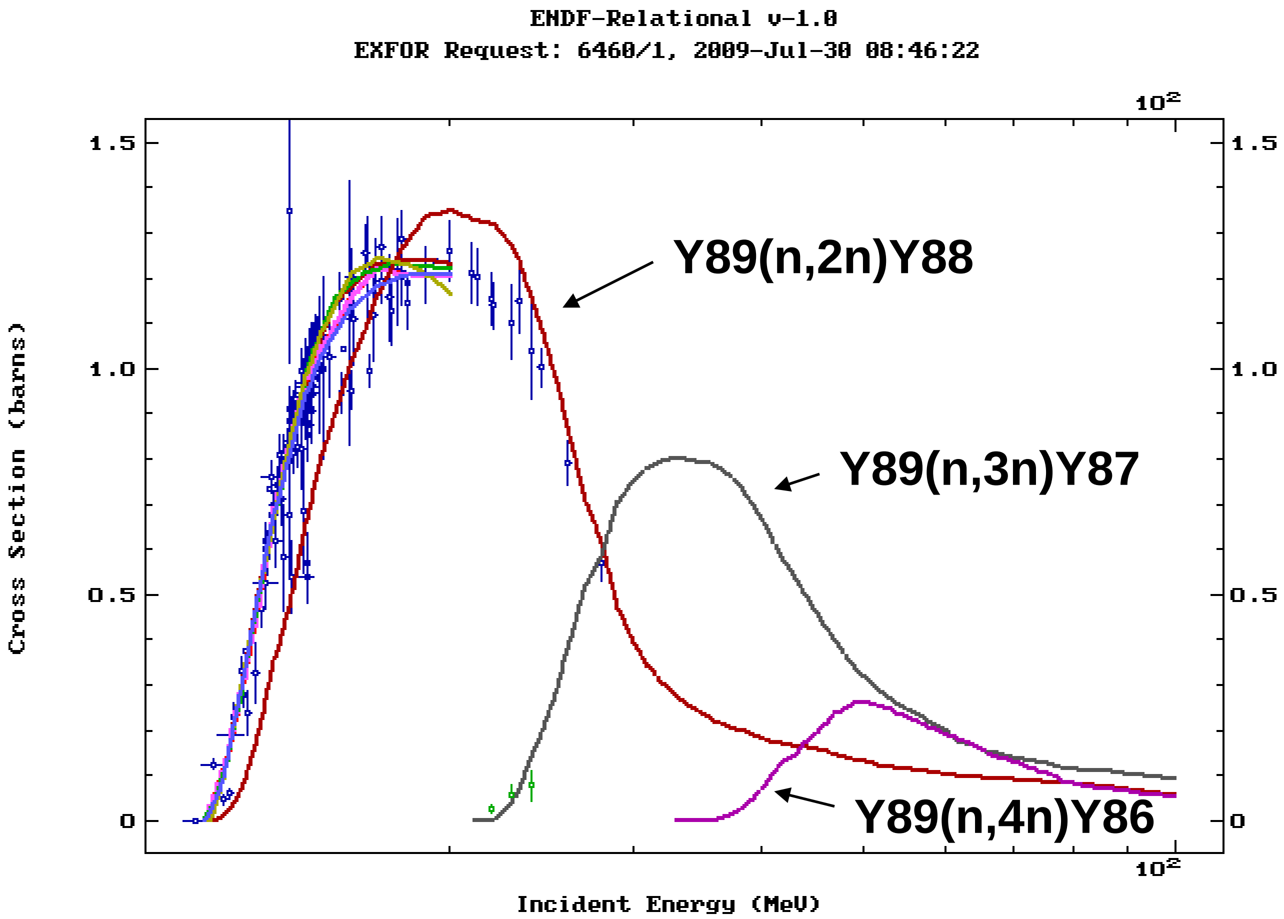
$I(E_{thr})$ case on example sample
(Ed = 4.0 GeV, position P2-R3)



$$I(E_{thr}) = \kappa (E_{thr} + \varepsilon) e^{-\frac{(E_{thr}+\varepsilon)}{\eta}}$$

with **κ, η** and **ε** as fitting parameters

Y89(n,xn) reaction cross section case available data – experimental and evaluated



Y89(n,xn) reaction cross section case - basic assumptions and consequences

$$\sigma(E, E_{thr}) = \begin{cases} \frac{\alpha}{(kT)^{\frac{3}{2}}} \frac{(E - E_{thr})}{\sqrt{E}} \exp\left(-\frac{(E - E_{thr})}{kT}\right) & \mathrm{E} > \mathrm{E}_{\mathrm{thr}} \\ 0 & \mathrm{E} \leq \mathrm{E}_{\mathrm{thr}} \end{cases}$$

because it fits the best EXFOR database for Y89(n,2n)Y88 reaction cross section experimental data.

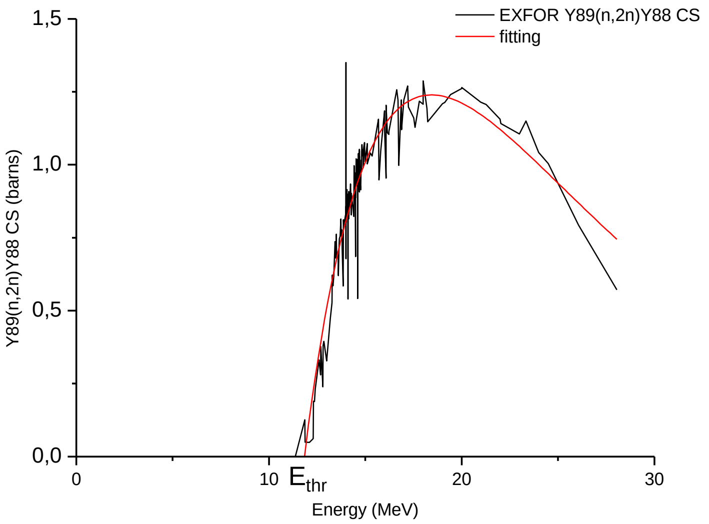
The same formula applies to all Y89(n,xn) reactions. The main difference between Y89(n,2n), (n,3n) and (n,4n) reactions is their threshold energy - E_{thr}.

Notes on Y89(n,xn) reaction cross section analytical formula

- **It resembles Maxwell-Boltzmann energy distribution function for ideal gas particles in equilibrium f(E) ~ sqrt(E)*exp(-E/kT).**
- **Statistical model of Y89 nucleus applies, i.e. nucleons behave like an ideal gas particles. The only interactions between them are elastic collisions. In an equilibrium stage their energy is dispersed around mean energy kT.**
- **Reaction CS proportional to f(E), σ(E)~f(E).**
- **N. Bohr reaction model applies to Y89(n,xn) one, where the reaction has two stages – compound nucleus formation (10^{-22} s) and compound nucleus evaporation (10^{-18} – 10^{-16} s).**
- **Y89(n,xn) reaction is caused by neutrons with energy larger than threshold energy – E_{thr}.**
- **Y89(n,xn) reaction cross section is directly proportional to a time of flight through yttrium nucleus, i.e. inversely proportional to neutron speed, i.e. inversely proportional to the square root of neutron energy**
- **Neutrons and protons are the two separated, not interacting gases**

Y89(n,2n)Y88 reaction cross section fitting

$$\sigma(E, E_{thr}) = \frac{\alpha}{(kT)^{\frac{3}{2}}} \frac{(E - E_{thr})}{\sqrt{E}} e^{-\frac{(E - E_{thr})}{kT}}$$

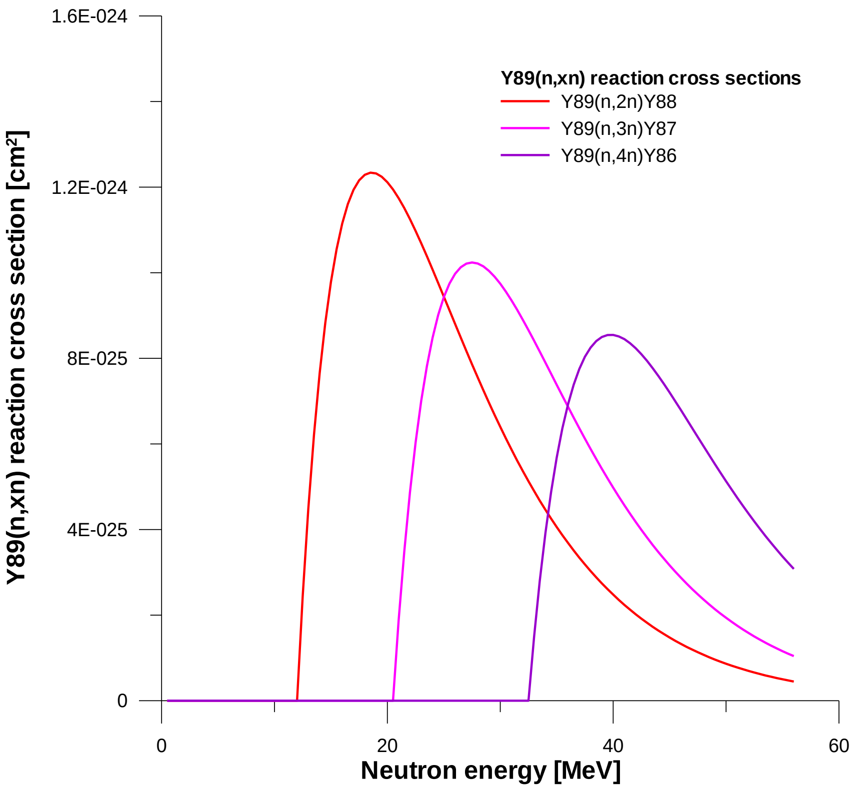


The fitting gives

- **α,kT - coefficients**
- **E_{thr} – reaction threshold energy**

Y89(n,xn) reaction cross section looks like

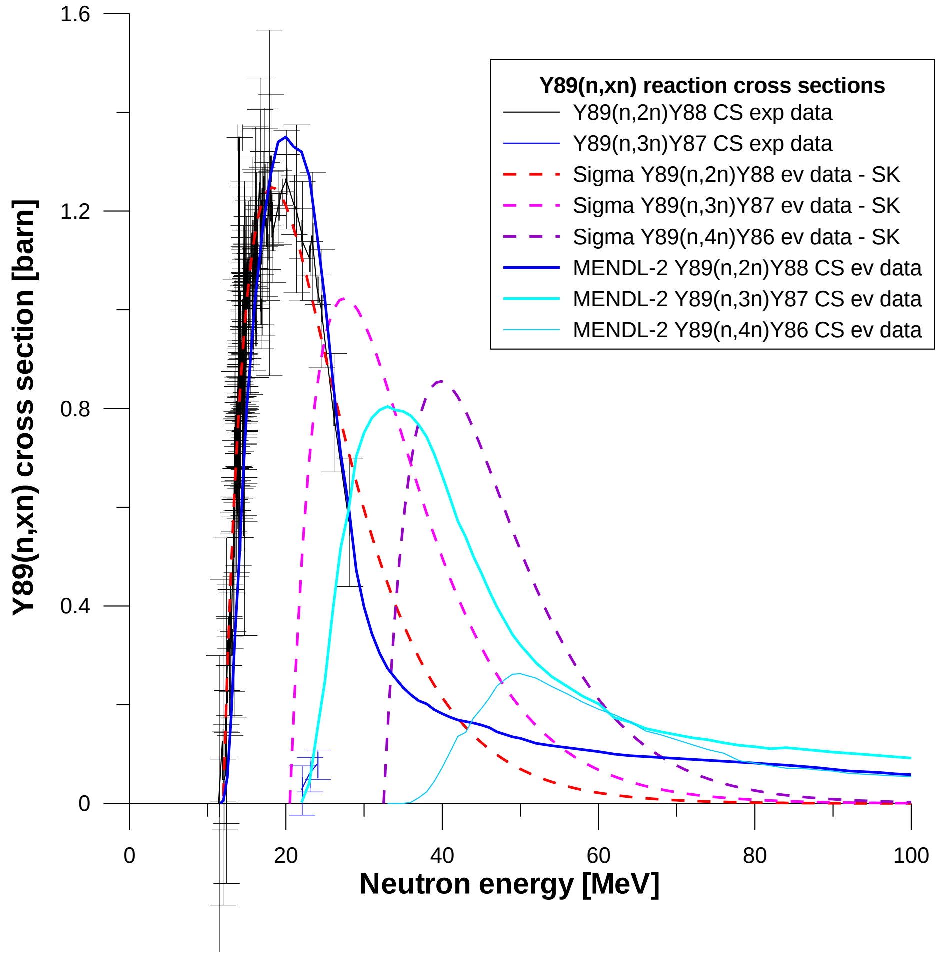
$$\sigma\left(E,E_{thr}\right)=\frac{\alpha}{\left(kT\right)^{\frac{3}{2}}}\frac{\left(E-E_{thr}\right)}{\sqrt{E}}e^{-\frac{\left(E-E_{thr}\right)}{kT}}$$



Y89(n,2n)Y88 CS fitting coefficients α,kT applied to any Y89(n,xn) reaction

E_{thr} – from elsewhere

Y89(n,xn) reaction cross section comparison: experimental data, MENDL-2 data base, and this work



Spallation neutron energy spectrum $\phi(E)$ determination basing on experimental data $I(E_{thr})$ and $\sigma(E,E_{thr})$

$$I(E_{thr}) = N \int_{E_{thr}}^{\infty} \phi(E)\sigma(E,E_{thr})dE$$

Solving an integral equation for yttrium isotope production

Using the mentioned earlier functions for $\sigma(E,E_{thr})$ and $I(E_{thr})$ the equation

$$I(E_{thr}) = N \int_{E_{thr}}^{\infty} \phi(E)\sigma(E,E_{thr})dE$$

becomes

$$\frac{\alpha N}{(kT)^{\frac{3}{2}}} \int_{\infty}^{E_{thr}} (E_{thr}-E)e^{-\frac{E}{kT}} \frac{\phi(E)}{\sqrt{E}} dE = \kappa(E_{thr}+\varepsilon)e^{-\frac{E_{thr}}{kT}}$$

Differentiating twice both sides on E_{thr} one gets:

$$\phi(E_{thr}) = \frac{\kappa(\eta+kT)^2}{\alpha N \eta^2 (kT)^{\frac{1}{2}}} \left[E_{thr} + \varepsilon - \frac{2\eta kT}{(\eta+\beta)} \right] e^{-\frac{(E_{thr}+\varepsilon)}{\eta}} \sqrt{E_{thr}}$$

Solving a Volterra's integral equation for Yttrium isotope production - continuation

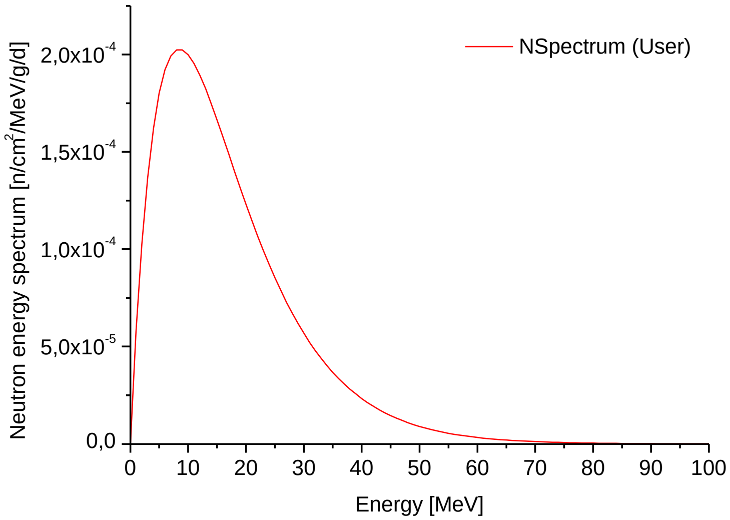
To fulfill request $\phi(E \leq 0) = 0$ and $\phi(E \to \infty) \to 0$ must be

$$\varepsilon = \frac{2\eta kT}{\eta + kT}$$

and $\phi(E)$ becomes

$$\phi(E) = \frac{\kappa(\eta + kT)^2}{\alpha N \eta^2 (kT)^{\frac{1}{2}}} E^{\frac{3}{2}} e^{-\frac{1}{\eta}\left(E + \frac{2\eta kT}{\eta + kT}\right)}$$

ϕ(E) example view
(Ed = 4 GeV, P2-R3)



$$\phi(E) = \frac{\kappa(\eta + kT)^2}{\alpha N \eta^2 (kT)^{\frac{1}{2}}} E^{\frac{3}{2}} e^{-\frac{1}{\eta}\left(E + \frac{2\eta kT}{\eta + kT}\right)}$$

$$\phi_{\max}(E) \text{ at } E = \frac{3}{2}\eta$$

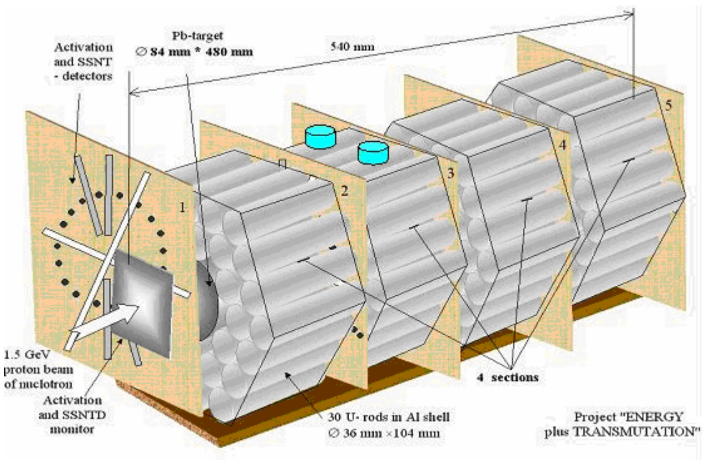
The method application example

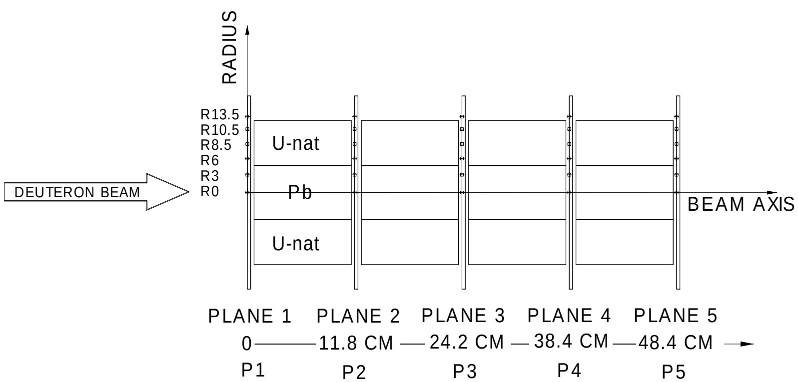
The method has been applied
to „Energy plus Transmutation” experiment
results elaboration

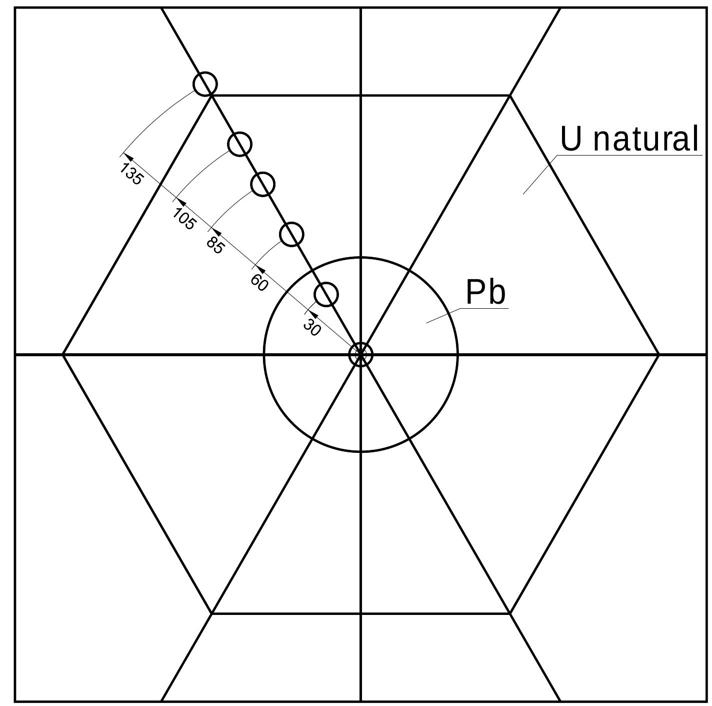
„Energy plus Transmutation” project

- **International research project realised in JINR Dubna.**
  - **12 states take part in**
  - **Started 1999**
- **Purpose of the project is to study transmutation on U/Pb-assembly driven by accelerator NUKLOTRON.**
- **Transmutation samples – ^{129}I, ^{237}Np, ^{238}Pu, ^{239}Pu**
- **Activation detectors – Al, Ti, V, Mn, Fe, Co, Ni, Cu, Y, Nb, In, Dy, Lu, W, Au, Bi**
- **^{3}He counter**
- **SSNTD**

Yttrium samples location during irradiation







Experiments with „Energy plus Transmutation” set-up

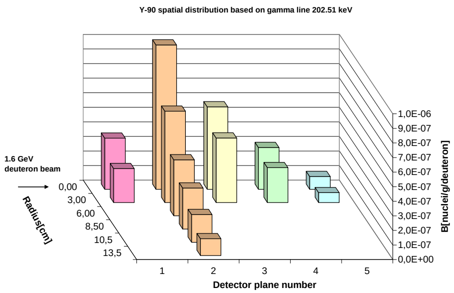
- **Proton beam:**
- **Ep = 0.7 GeV**
- **Ep = 1.0 GeV**
- **Ep = 1.5 GeV**
- **Ep = 2.0 GeV**

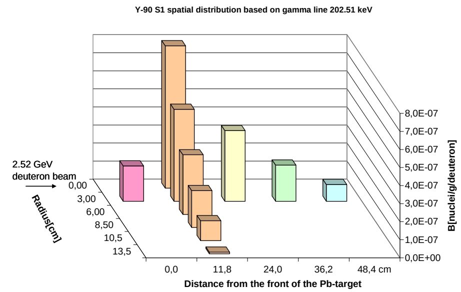
- **Deuteron beam:**
- **Ed = 1.6 GeV**
  = 0.8 GeV/nucleon
- **Ed = 2.52 GeV**
  = 1.26 GeV/nucleon
- **Ed = 4.0 GeV**
  = 2.0 GeV/nucleon

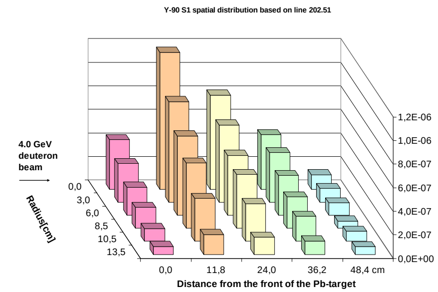
30 Y89 samples irradiated in each experiment

Y-90 and Y-88 production spatial distribution comparison

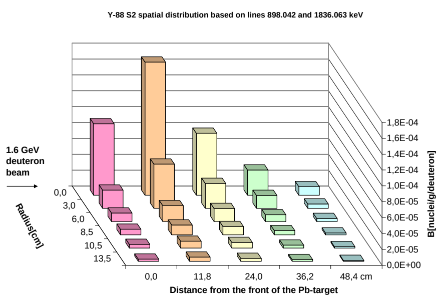
Y90

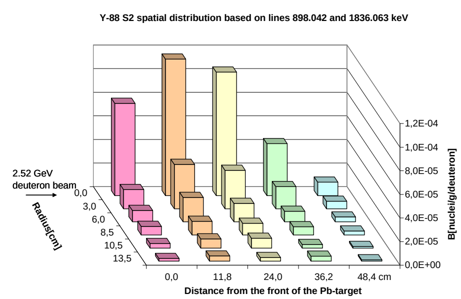


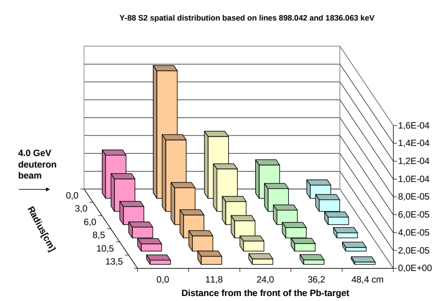




Y88



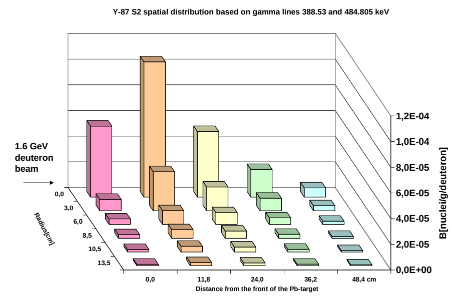


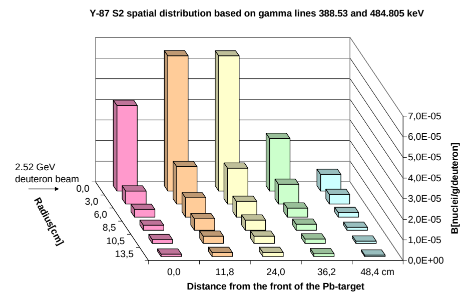


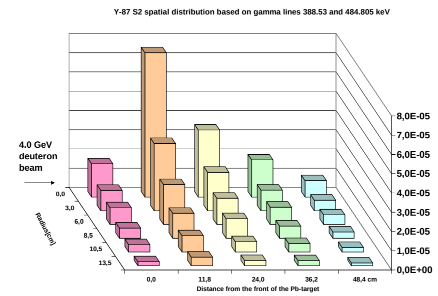
Ed = 1.6 GeV Ed = 2.52 GeV Ed = 4.0 GeV

Y-87 and Y-86 production spatial distribution comparison

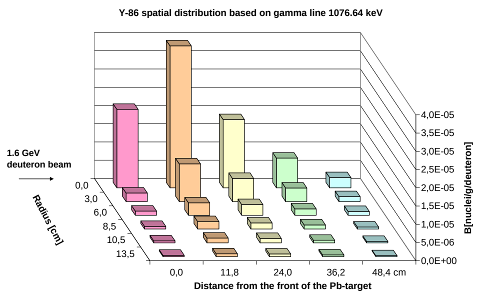
Y87

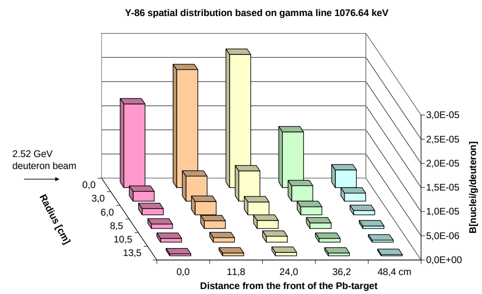


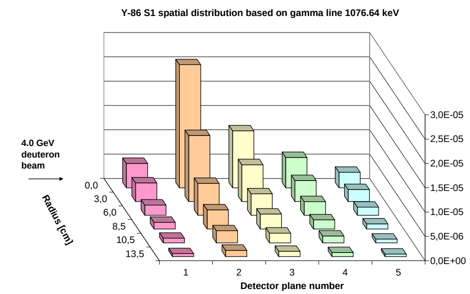




Y86







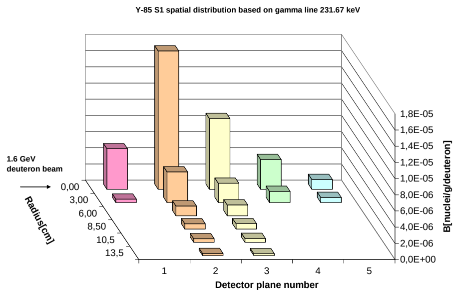
Ed = 1.6 GeV

Ed = 2.52 GeV

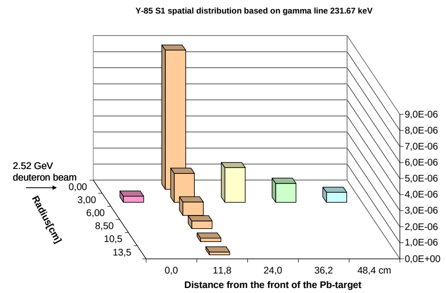
Ed = 4.0 GeV

Y-85 production spatial distribution comparison

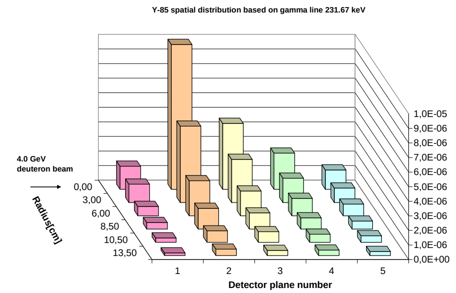
Y85



Ed = 1.6 GeV

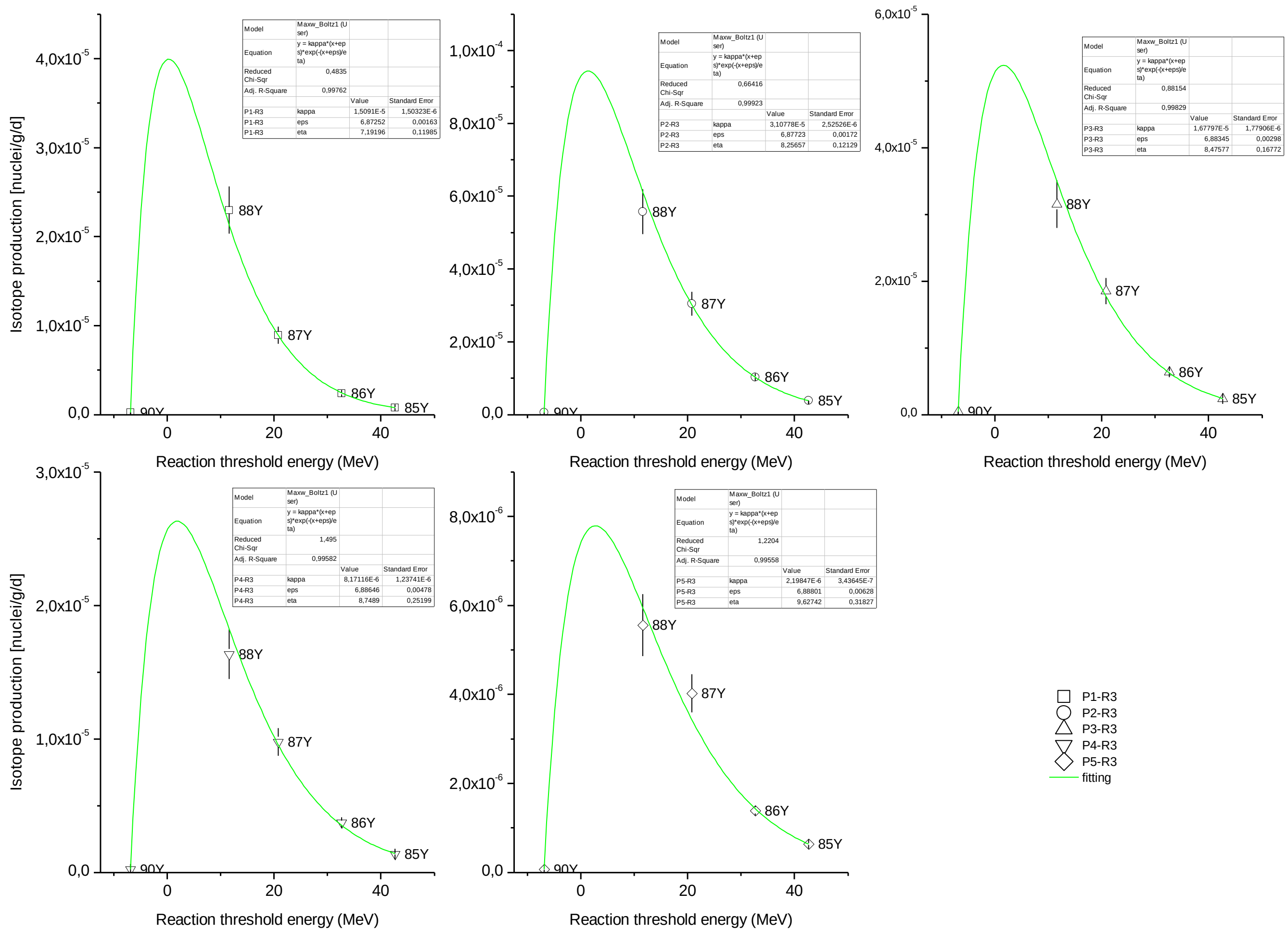


Ed = 2.52 GeV



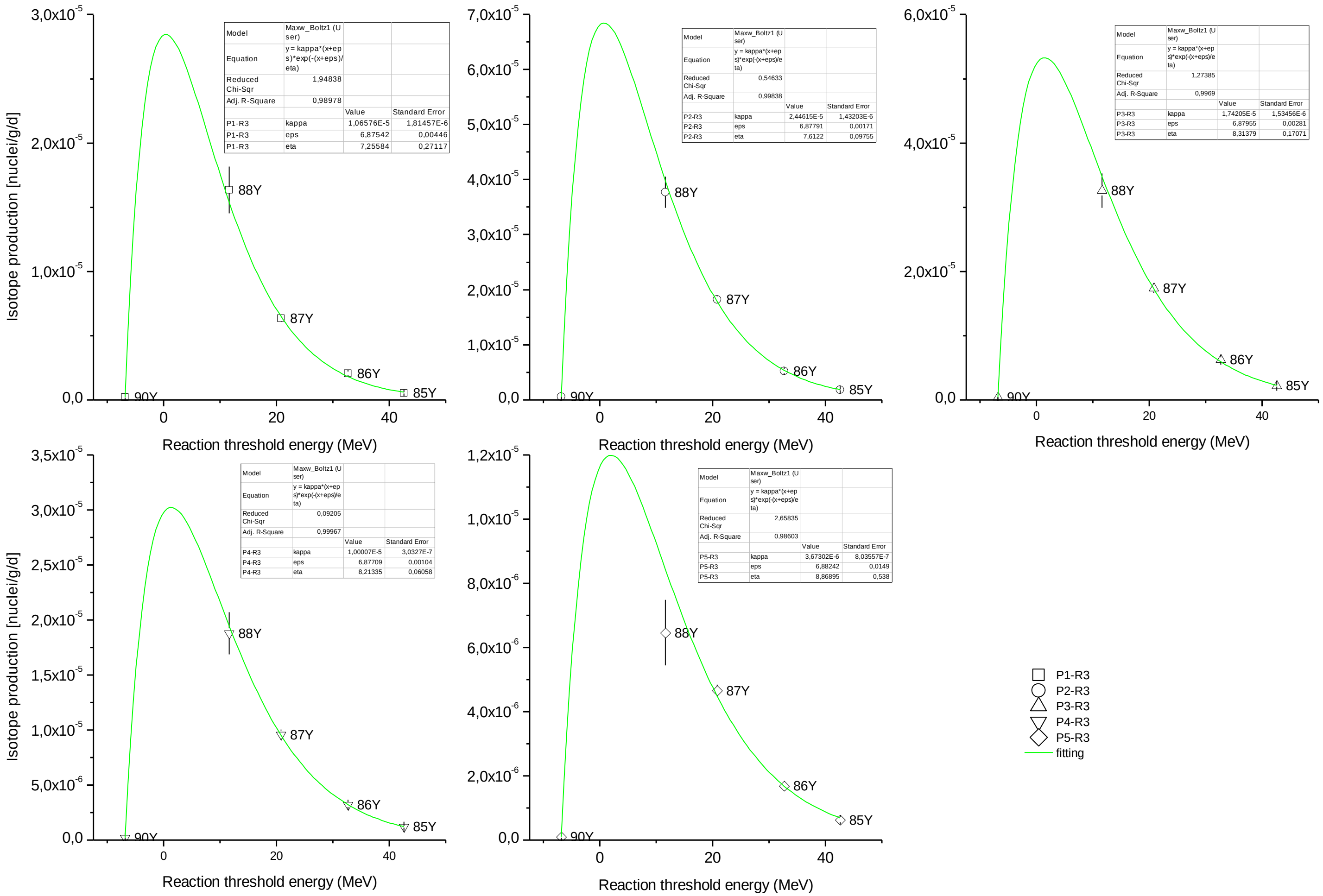
Ed = 4.0 GeV

Function I(E_{thr}) in various points of R = 3 cm axis for Ed = 1.6 GeV



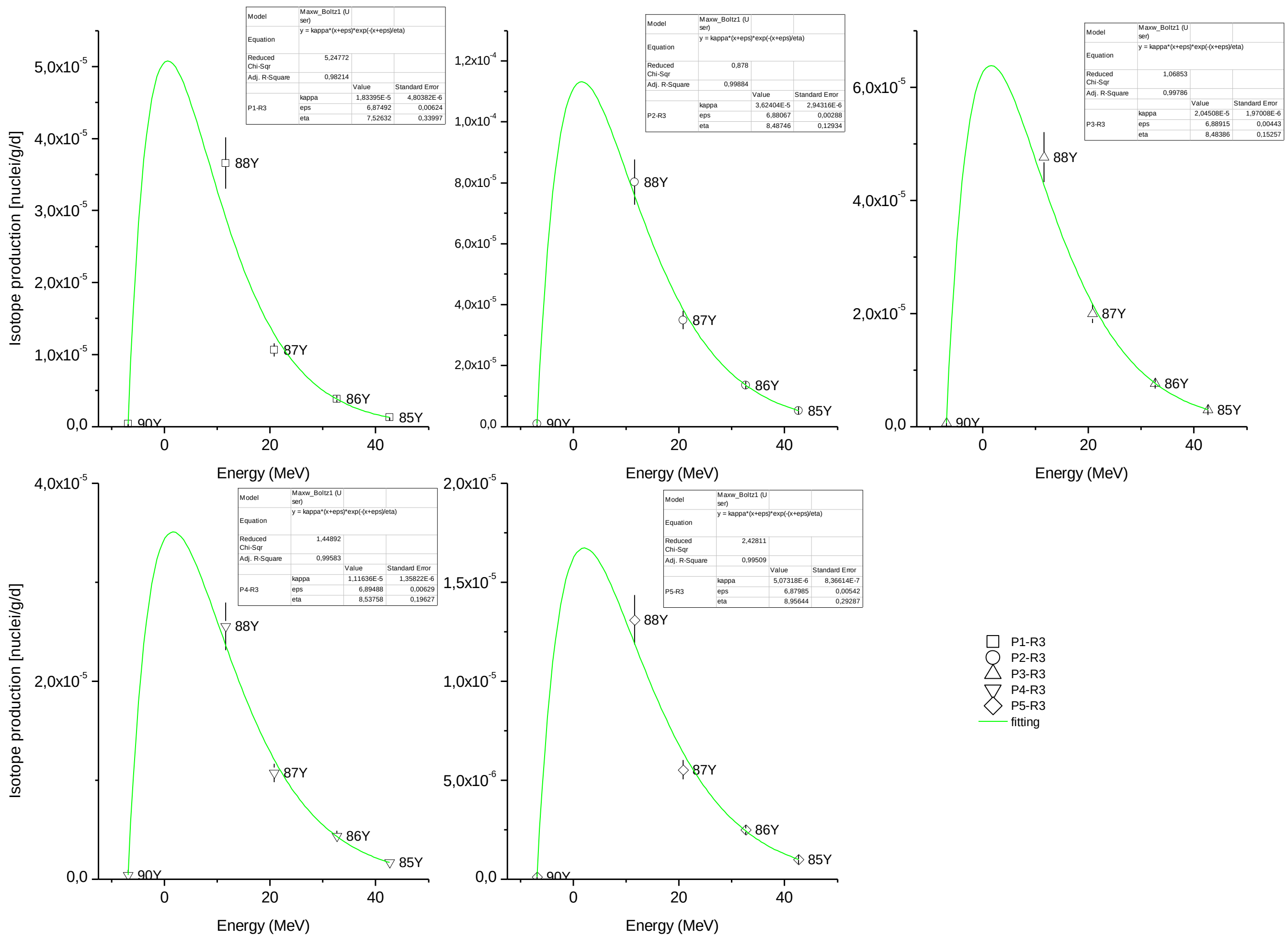
Ed = 1.6 GeV, axis R3 - yttrium isotope production dependence on reaction threshold energy at various axial positions

Function I(E_{thr}) in various points of R = 3 cm axis for Ed = 2.52 GeV



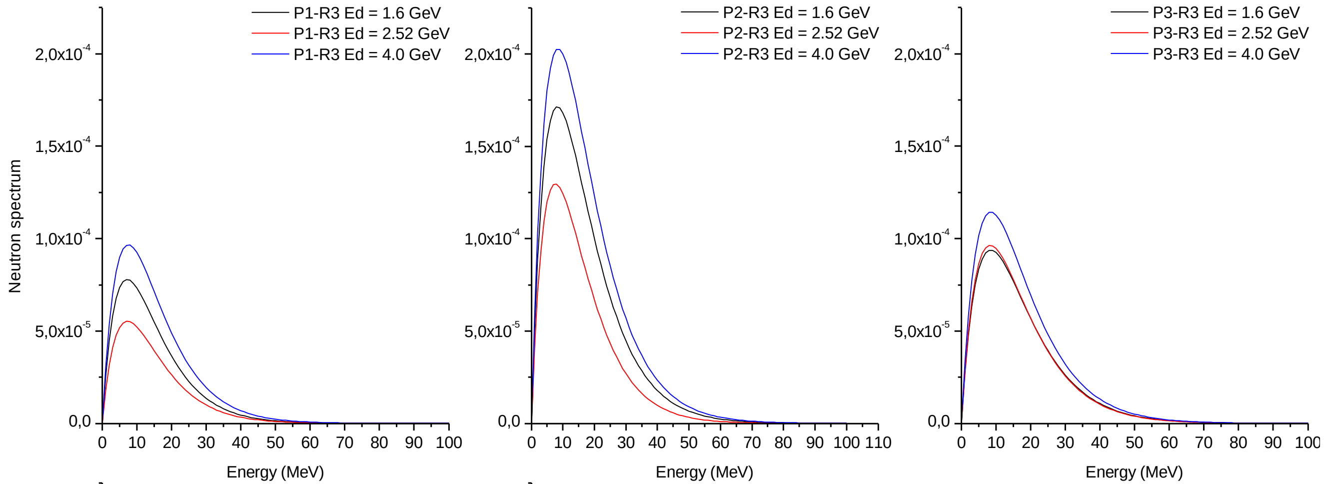
Ed = 2.52 GeV, axis R3 - yttrium isotope production dependence on reaction threshold energy at various axial positions

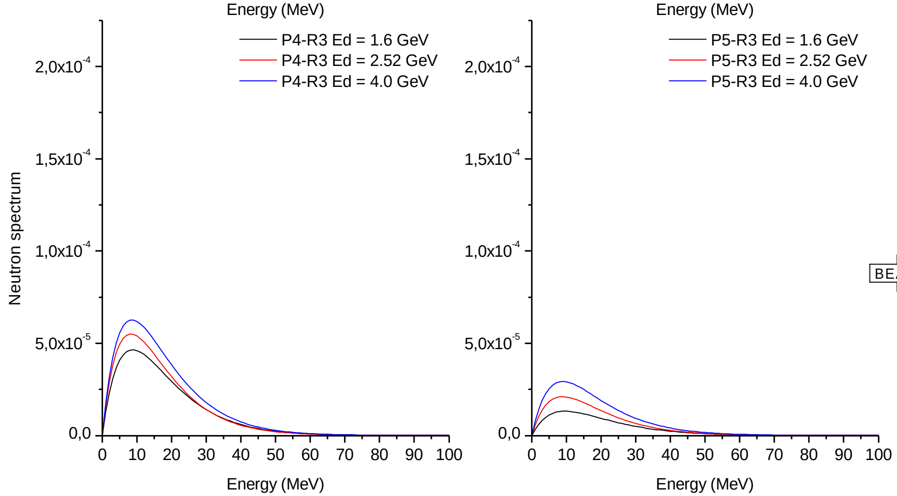
Function I(E_{thr}) in various points of R = 3 cm axis for Ed = 4.0 GeV

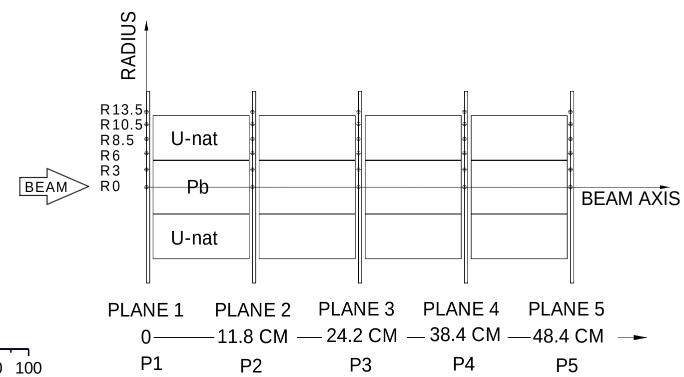


Ed = 4 GeV, axis R3 - yttrium isotope production dependence on reaction threshold energy at various axial positions.

Ed = 1.6; 2.52 and 4.0 GeV - Neutron flux energy spectra comparison at various R3-axis positions - common Y-scale.







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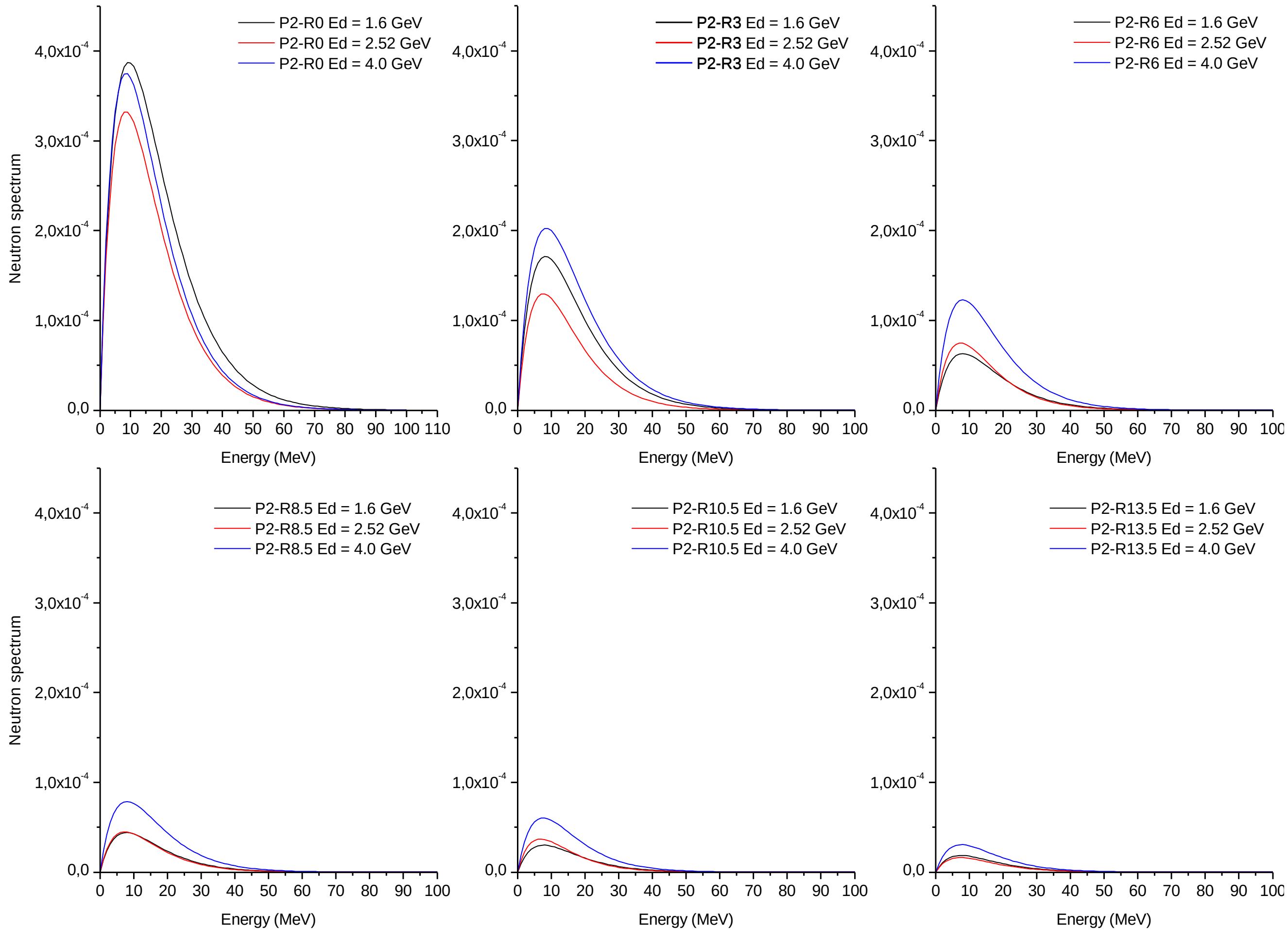
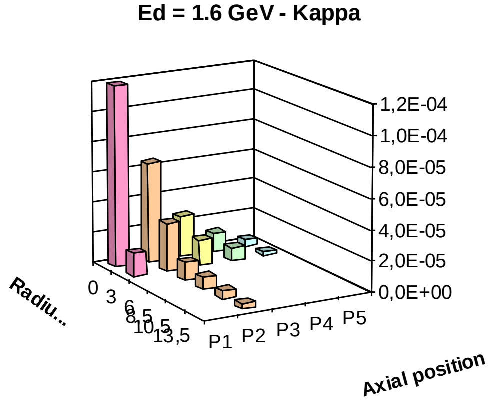
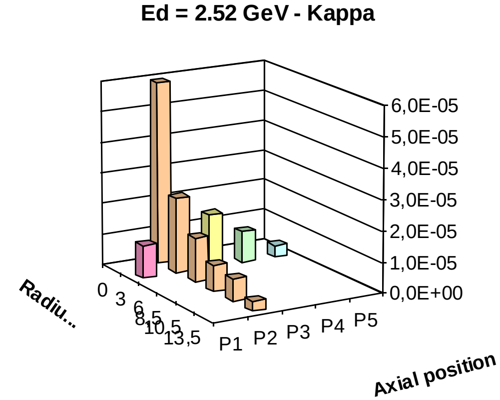
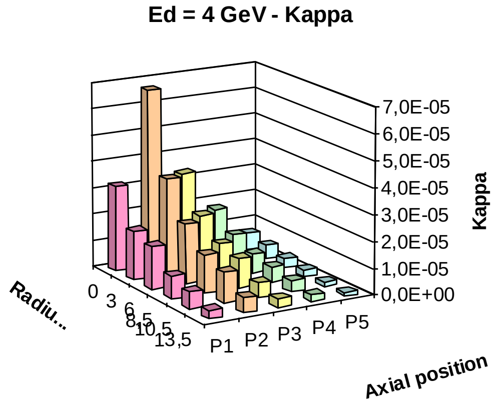
Ed = 1.6; 2.52 and 4.0 GeV - Neutron flux spectra comparison at various P2 radial positions - common Y-scale

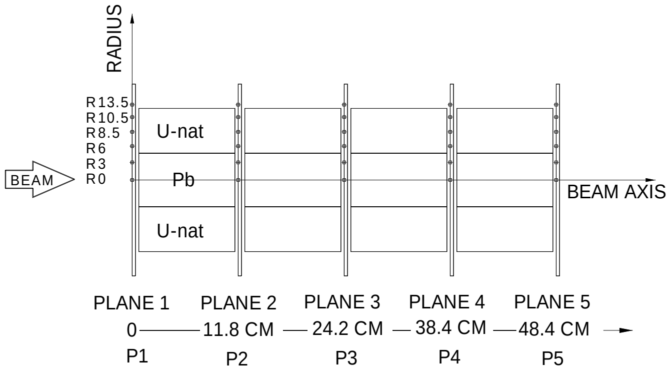


Ed = 1.6, 2.52 and 4.0 GeV parameter kappa comparison

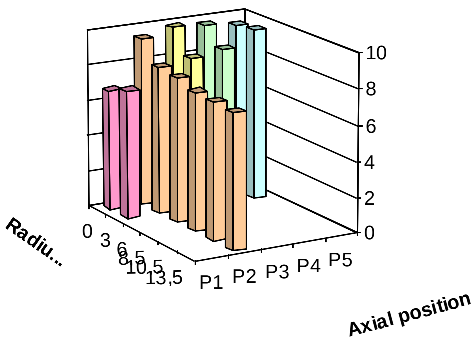


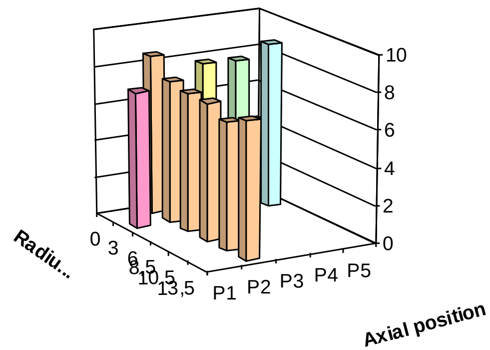


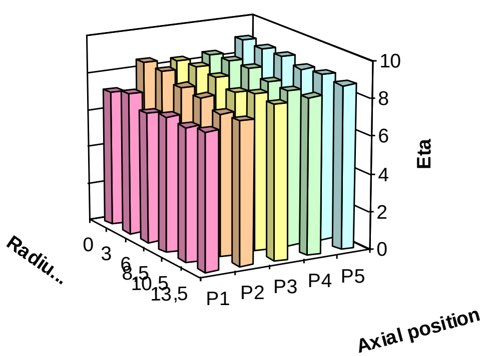


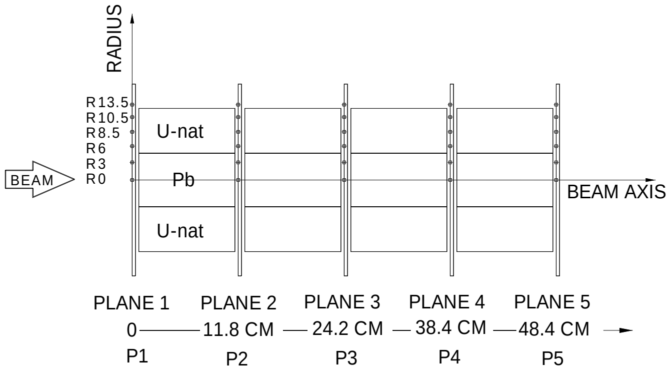


Ed = 1.6, 2.52 and 4.0 GeV parameter eta comparison

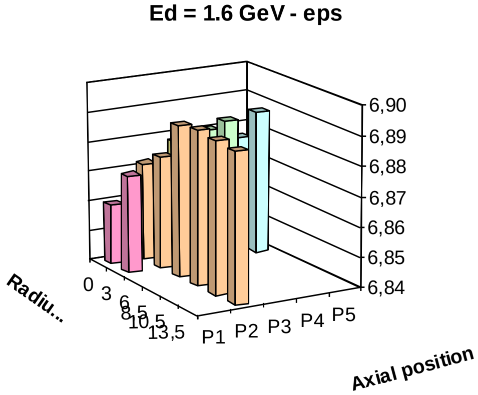


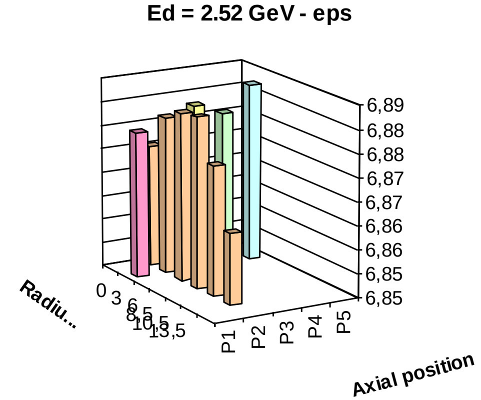


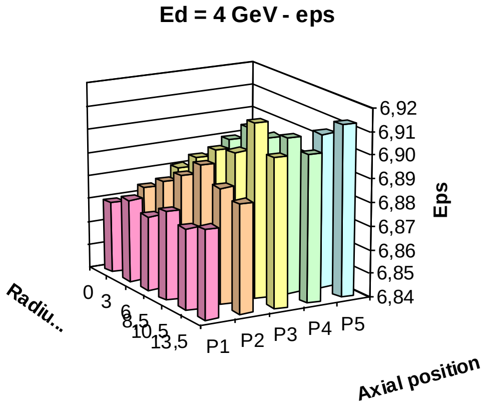


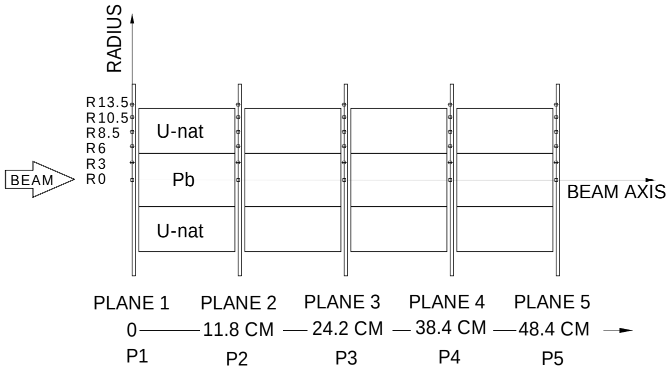


Ed = 1.6, 2.52 and 4.0 GeV parameter eps comparison

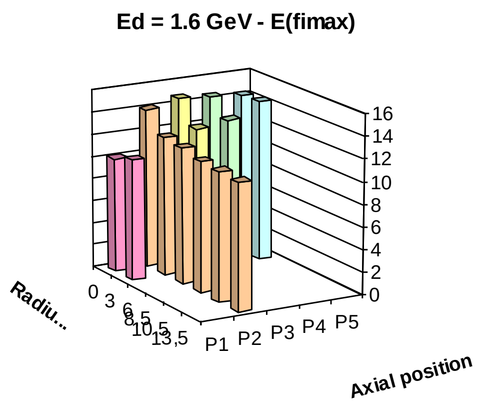


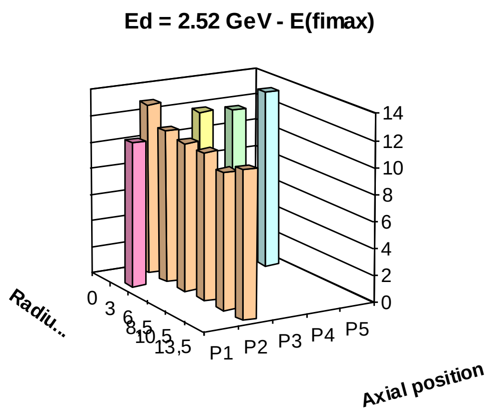


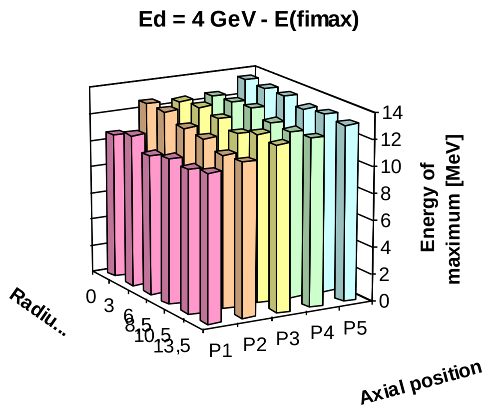


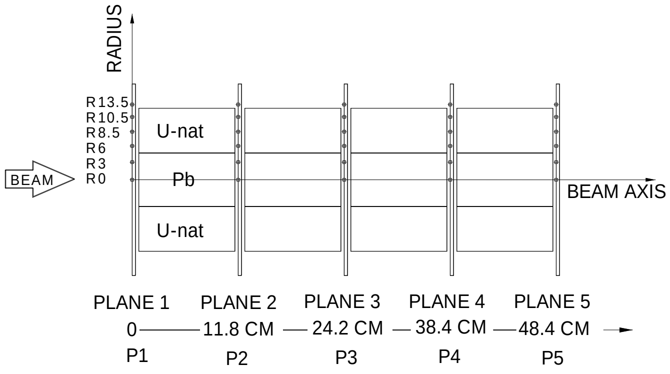


Ed = 1.6, 2.52 and 4.0 GeV E(ϕ_{max}) comparison

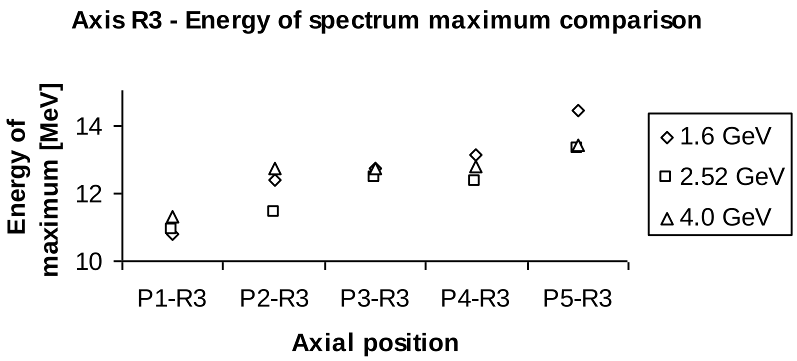


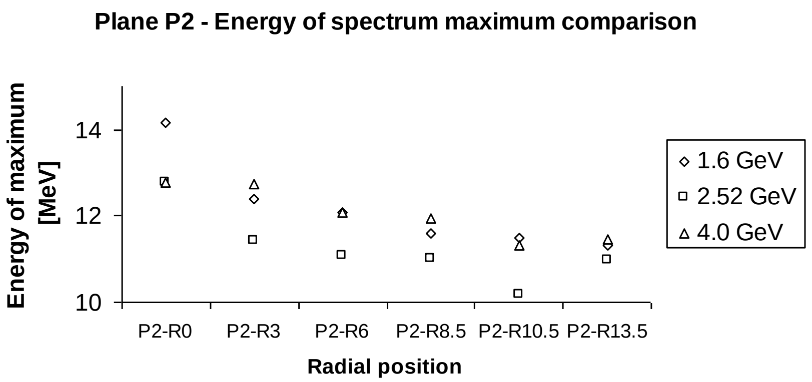


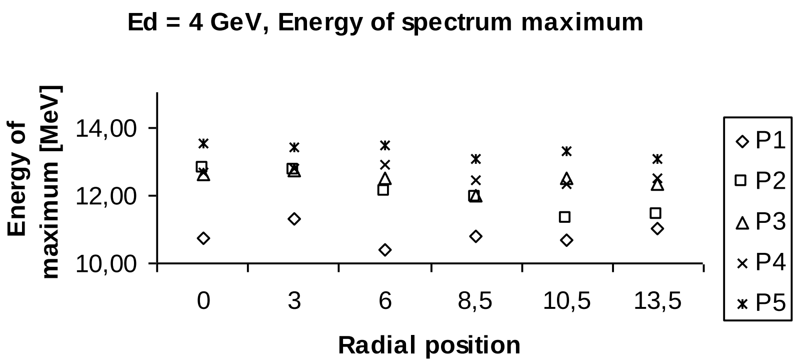


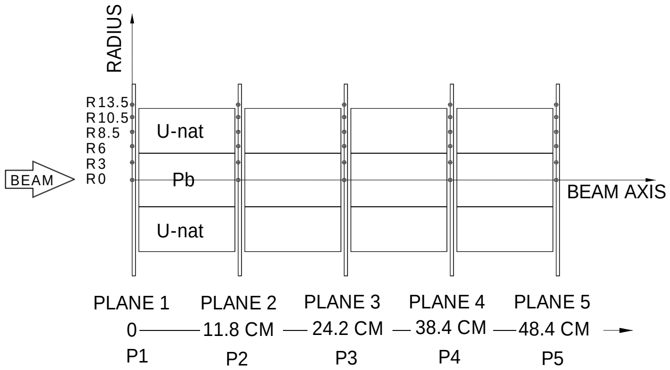


Ed = 1.6, 2.52 and 4.0 GeV E(ϕ_{max}) comparison









Energy of spectrum maximum

	Ed [GeV]	P1		P2		P3		P4		P5	
		Efmax	Efmax Error	Efmax	Efmax Error	Efmax	Efmax Error	Efmax	Efmax Error	Efmax	Efmax Error
R0	1.6	10,13	2,33	14,17	0,16	14,75	0,36	14,58	0,26	14,27	0,36
	2.52			12,78	0,27						
	4.0	10,74	0,17	12,78	0,07	12,59	0,06	12,67	0,17	13,55	0,11
R3	1.6	10,79	0,18	12,38	0,18	12,71	0,25	13,12	0,38	14,44	0,48
	2.52	10,88	0,41	11,42	0,15	12,47	0,26	12,32	0,09	13,30	0,81
	4.0	11,29	0,51	12,73	0,19	12,73	0,23	12,81	0,29	13,43	0,44
R6	1.6			12,10	0,19						
	2.52			11,08	0,15						
	4.0	10,41	0,11	12,10	0,07	12,48	0,11	12,90	0,47	13,49	0,65
R8.5	1.6			11,60	0,35						
	2.52			11,01	0,25						
	4.0	10,81	0,13	11,93	0,11	11,97	0,16	12,42	0,39	13,09	0,74
R10.5	1.6			11,48	0,21						
	2.52			10,18	0,34						
	4.0	10,68	0,19	11,33	0,23	12,51	0,20	12,36	0,26	13,30	1,17
R13.5	1.6			11,30	0,21						
	2.52			10,99	0,08						
	4.0	11,00	0,15	11,45	0,16	12,34	0,51	12,49	0,51	13,07	0,68

The method error discussion

- Typical errors like count statistics, deuteron fluence error, sample mass error and so on are of less importance;
- Y89(n,2n)Y88 reaction cross section approximation (fitting) error of much more importance, but difficult to assess;
- Y89(n,xn) reaction cross section approximation by Y89(n,2n)Y88 parameters makes additional error;
- Approximation of infinite number of ideal gas particles with 50 neutrons (statistical model of nucleus);
- Relativistic effects?

In some sense the method tests itself saying that where both η, kT and ε are fitting parameters.

$$\varepsilon = \frac{2\eta kT}{\eta + kT}$$

Discrepancy between the direct fitting ε value and calculated using η, kT should be an error measure of the method.

While direct fitting ε value ranges from 6.87 to 6.91 MeV the η, kT calculated from ranges from 7.44 to 8.5.
This suggests the method error to be of order of 10-20%.

This still doesn't explain the high energy of the spectrum maximum!

Are there any other errors of the method? Note: These are the experimental values.

Yurevich's measurement of spallation neutron spectrum (but induced by protons) gave high average neutron energy too.

What else could be done

Done

Reaction	Threshold energy [MeV]
Y89(n,g)Y90	-6,857
Y89(n,2n)Y88	11,6048
Y89(n,3n)Y87	21,0645
Y89(n,4n)Y86	33,008
Y89(n,5n)Y85	42,633

To be done

Reaction	Threshold energy [MeV]
Y89(n,He4)Rb86	-0,6916
Y89(n,p)Sr89	0,7183
Y89(n,d)Sr88	4,8995
Y89(n,2a)Br82	7,0633
Y89(n,np)Sr88	7,1494
Y89(n,T)Sr87	9,8104
Y89(n,He3)Rb87	10,0774
Y89(n,2p)Rb88	11,7317
Y89(n,2d)Rb86	23,4207
Y89(n,dT)Rb85	25,8424

But how?

Conclusions

- The method is simple, the results are coherent.

- Surprising is that using threshold detector with few threshold energies one can say so much about the spectrum in the entire energy range.

- According to the method there’s no big difference between Ed = 1.6, 2.52 and 4.0 GeV spectra. The spectrum seems to have a kind of saturation at these energies of deuterons.

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3. ANL/NDM-94 "Evaluated Neutronic Data File for Yttrium", A.B. Smith, D.L. Smith, P. Rousset, R.D. Lawson, and R.J. Howerton, January 1986
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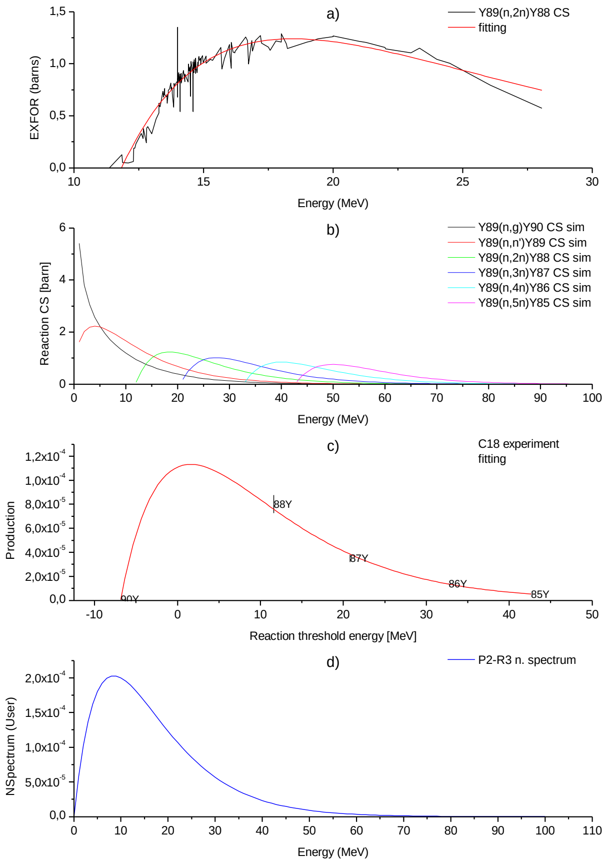
Thank You for attention

Background slides

Neutron spectrum determination stages



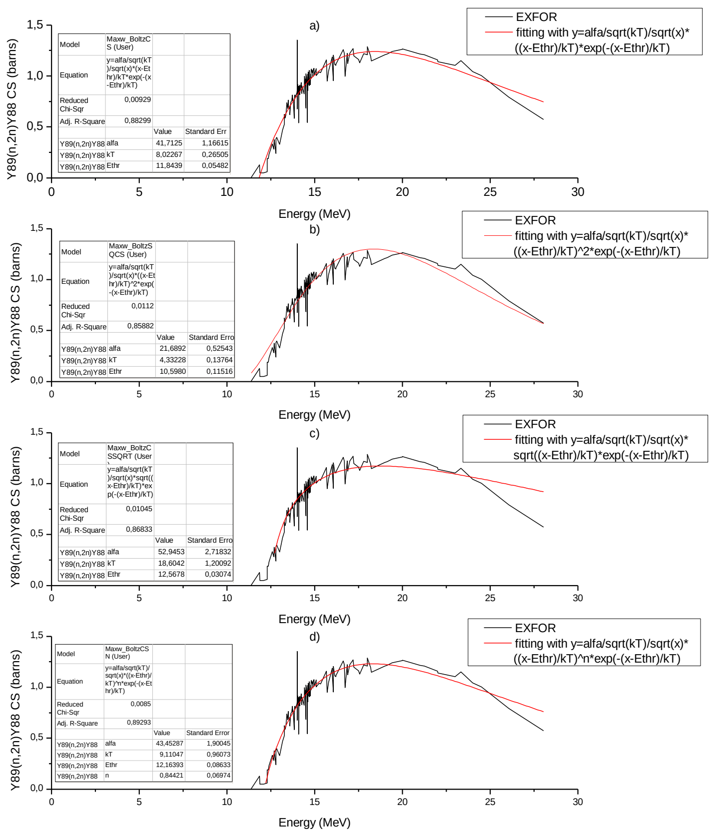
Y89(n,2n)Y88 reaction CS EXFOR data fitting to get parameters α and kT

Any Y89(n,xn) reaction CS determination using α and kT parameters and E_{thr}

Yttrium isotope production fitting to get parameters κ, η and ε

Spallation neutron spectrum determination as A function of α, kT, κ and η.

Y89(n,2n)Y88 reaction CS EXFOR data various fittings



Y89(n,2n)Y88 reaction CS EXFOR data various fittiings

$$\frac{\alpha}{(kT)^{\frac{3}{2}}}\frac{(E-E_{88})}{\sqrt{E}}\exp\left(-\frac{E-E_{thr,k}}{kT}\right)$$

Statistics	Statistics
Reduced Chi-Sqr	Adj. R-Square
0,00929	0,88299

$$\frac{\alpha}{(kT)^{\frac{3}{2}}}\frac{(E-E_{88})^2}{\sqrt{E}}\exp\left(-\frac{E-E_{thr,k}}{kT}\right)$$

Statistics	Statistics
Reduced Chi-Sqr	Adj. R-Square
0,0112	0,85882

$$\frac{\alpha}{(kT)^{\frac{3}{2}}}\frac{\sqrt{E-E_{88}}}{\sqrt{E}}\exp\left(-\frac{E-E_{thr,k}}{kT}\right)$$

Statistics	Statistics
Reduced Chi-Sqr	Adj. R-Square
0,01045	0,86833

$$\frac{\alpha}{(kT)^{\frac{3}{2}}}\frac{(E-E_{88})^n}{\sqrt{E}}\exp\left(-\frac{E-E_{thr,k}}{kT}\right)$$

n	Statistics	Statistics
Value	Reduced Chi-Sqr	Adj. R-Square
0,84421	0,0085	0,89293

Y89(n,2n)Y88 CS EXFOR data various fittings – cont.

	alfa	alfa	kT	kT	Ethr	Ethr	Statistics	Statistics
	Value	Standard Error	Value	Standard Error	Value	Standard Error	Reduced Chi-Sqr	Adj. R-Square
Y89(n,2n)Y88 CS	41,71254	1,16615	8,02267	0,26505	11,84393	0,05482	0,00929	0,88299

	alfa	alfa	kT	kT	Ethr	Ethr	Statistics	Statistics
	Value	Standard Error	Value	Standard Error	Value	Standard Error	Reduced Chi-Sqr	Adj. R-Square
Y89(n,2n)Y88 CS	21,68922	0,52543	4,33228	0,13764	10,59808	0,11516	0,0112	0,85882

	alfa	alfa	kT	kT	Ethr	Ethr	Statistics	Statistics
	Value	Standard Error	Value	Standard Error	Value	Standard Error	Reduced Chi-Sqr	Adj. R-Square
Y89(n,2n)Y88 CS	52,94533	2,71832	18,60426	1,20092	12,5678	0,03074	0,01045	0,86833

	alfa	alfa	kT	kT	Ethr	Ethr	n	n	Statistics	Statistics
	Value	Standard Error	Value	Standard Error	Value	Standard Error	Value	Standard Error	Reduced Chi-Sqr	Adj. R-Square
Y89(n,2n)Y88 CS	43,45287	1,90045	9,11047	0,96073	12,16393	0,08633	0,84421	0,06974	0,0085	0,89293

Evaporation mechanism of Y89(n,xn) reaction