Spatial distributions of $^{238}U(n,\gamma)$ and $^{238}U(n,f)$ reactions in uranium target of assembly "QUINTA-M" irradiated with 2, 4 and 6 GeV deuterons

V.A. Voronko[1], V.V. Sotnikov[1], M.Y. Artiushenko[1], Y.T. Petrusenko[1], M.G. Kadykov[2], S.I. Tyutyunnikov[2], V.V. Chilap[3], A.V. Chinenov[3]

[1]National Science Center Kharkov Institute of Physics and Technology, NAS Kharkov, Ukraine 61108

[2]Joint Institute for Nuclear Research, Dubna, Russia 141980

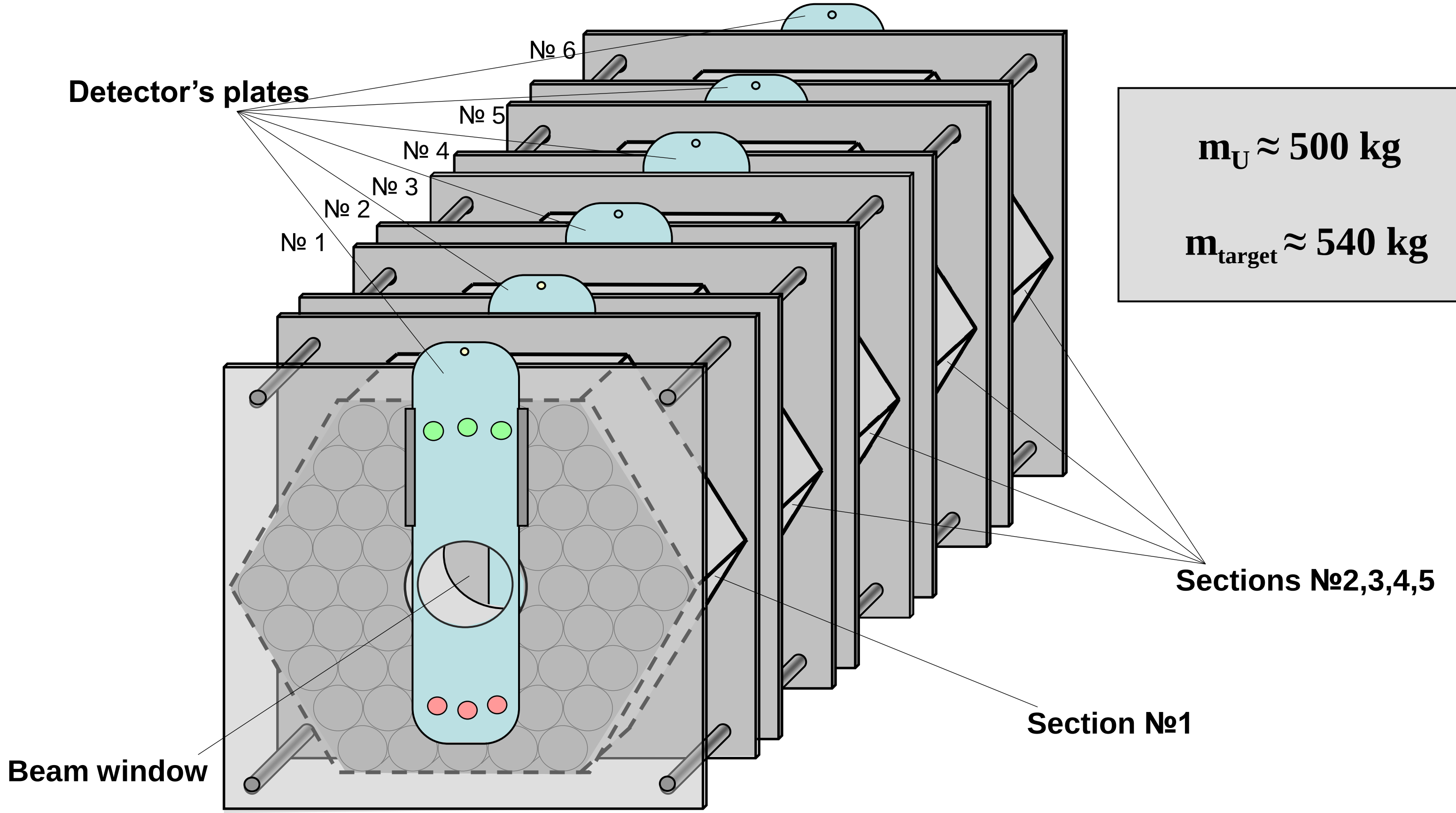
[3]Center of Physic and Technical Projects "Atomenergomash", Moscow, Russia

ISINN-20 Alushta, Ukraine, May 21-26, 2012

Main purposes of this work

- to obtain spatial distributions of density of radiative capture reactions (the number of accumulating ^{239}Pu nuclei) and density of ^{238}U fissions in the volume of uranium target of assembly QUINTA-M;
- to obtain spatial distribution of spectral indices;
- to determine the total number of ^{238}U fissions and total amount of ^{239}Pu, accumulated at the volume of uranium target of assembly QUINTA-M;
- to compare of the obtained experimental results in dependence on the energy of the deuteron beam (per unit of beam power).

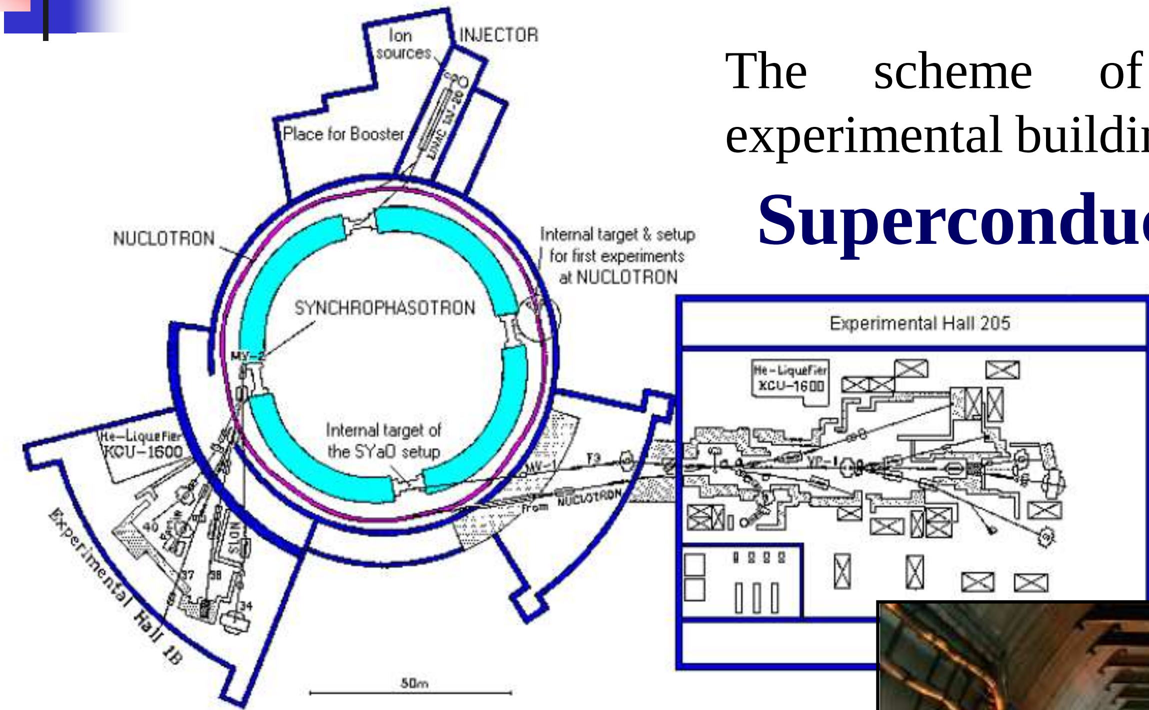
Experimental assembly "QUINTA-M"



$m_U \approx 500$ kg

$m_{target} \approx 540$ kg

Nuclotron accelerator at LHEP, JINR, Dubna



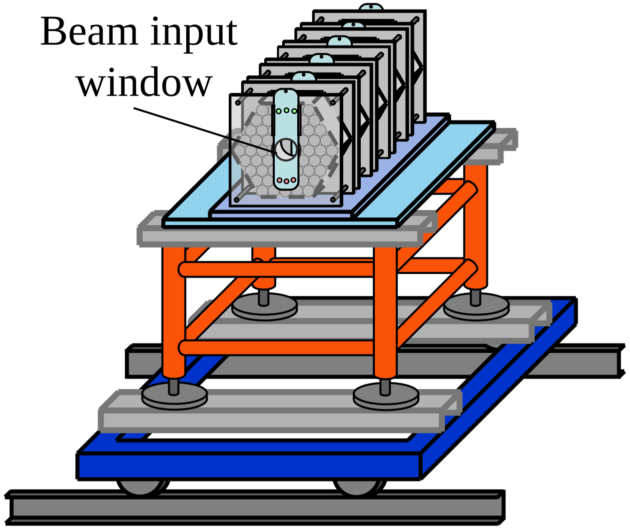
The scheme of the accelerator and experimental building 205

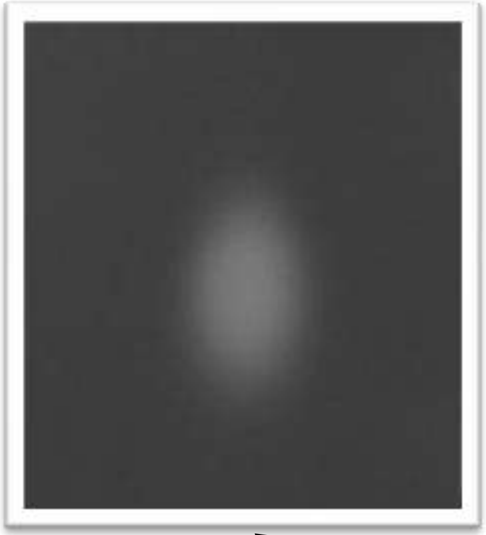
Superconductive accelerator –

- up to 12.8 GeV for protons or 6 GeV on nucleon (possibility of acceleration up to U)

- Extraction time ~ 10 s,
- Beam intensity $10^8 – 10^{11}$,
- Perimeter 251.5 m, weight of cooled magnets over 80 tons

Experimental assembly “QUINTA-M” at the irradiation position





Before the irradiation the target was carefully adjusted to the direction of the Nuclotron beam using polaroid films, i.e. the longitudinal axis of a target was combined with a direction of Nuclotron beam. The traces of one bunch of beam particles on polaroid film placed in front of the target

Determination of total beam intensity

basic relations

$$\Phi = \frac{A_M}{\sigma \cdot N_A \cdot D_{foil}} \cdot \left[\frac{S_p}{\varepsilon_p(E) \cdot I_\gamma} \cdot \frac{e^{\lambda t_c}}{1 - e^{-\lambda t_{live}}} \cdot \frac{e^{-\lambda \tau} - 1}{\lambda \tau} \cdot K_B \cdot K_{COI} \cdot K_{ABS} \cdot K_G \cdot K_{NP} \right]$$

$\boldsymbol{K_B}$ is factor determining the course of irradiation: $K_B = \sum_i f_i \Big/ \sum_i f_i e^{-\lambda t_{ci}}$

where $\boldsymbol{t_{ci}}$ - time from the end of each beam pulse until the end of irradiation ,
$\boldsymbol{f_i}$ - the intensity of the pulse (according to the values of monitoring of ionization chambers);

$\boldsymbol{K_{ABS}}$ is correction on self-absorption : $K_{ABS} = \frac{\mu D_{sum}}{1 - \exp(-\mu D_{sum})}$

$\boldsymbol{K_G}$ is correction on changed detector efficiency due to sample dimensions; effective center of the bulk sample is closer to the effective center of the detector (R_1) in comparison with a point calibration source (R_2):

$$K_G \approx (R_1/R_2)^2$$

$\boldsymbol{K_{NP}}$ is correction on non-point like emitters;
$\boldsymbol{K_{COI}}$ is correction for gamma-lines coincidences

The values of correction factors
(for example)

	Detector «C» (“ЯСНАПП”)		Detector “W” (room.216, 205 building)	
	Eγ 1368 кeV	Eγ 2754 кeV	Eγ 1368 кeV	Eγ 2754 кeV
K_G	0.945	0.945	0.982	0.982
K_{NP}	1.008	1.008	1.014	1.014
K_{ABS}	1.014	1.010	1.014	1.010
K_{COI}	1.010	1.014	1.02	1.03
$K_G K_{NP} K_{ABS} K_{COI}$	**0.976**	**0.976**	**1.030**	**1.036**

The choice of monitor $^{27}Al(d,x)^{24}Na$ reaction cross sections

In the energy range of deuterons above 300 MeV the cross section for ^{24}Na is known only for deuterons with energy:

2.33 GeV : **15.25 1.5 mb**

(*Banaigs J. et al.: Nuclear Instruments and Methods in Physics Research 95 (1971) 307-311.*)

6 GeV : **14.1 1.3 mb**

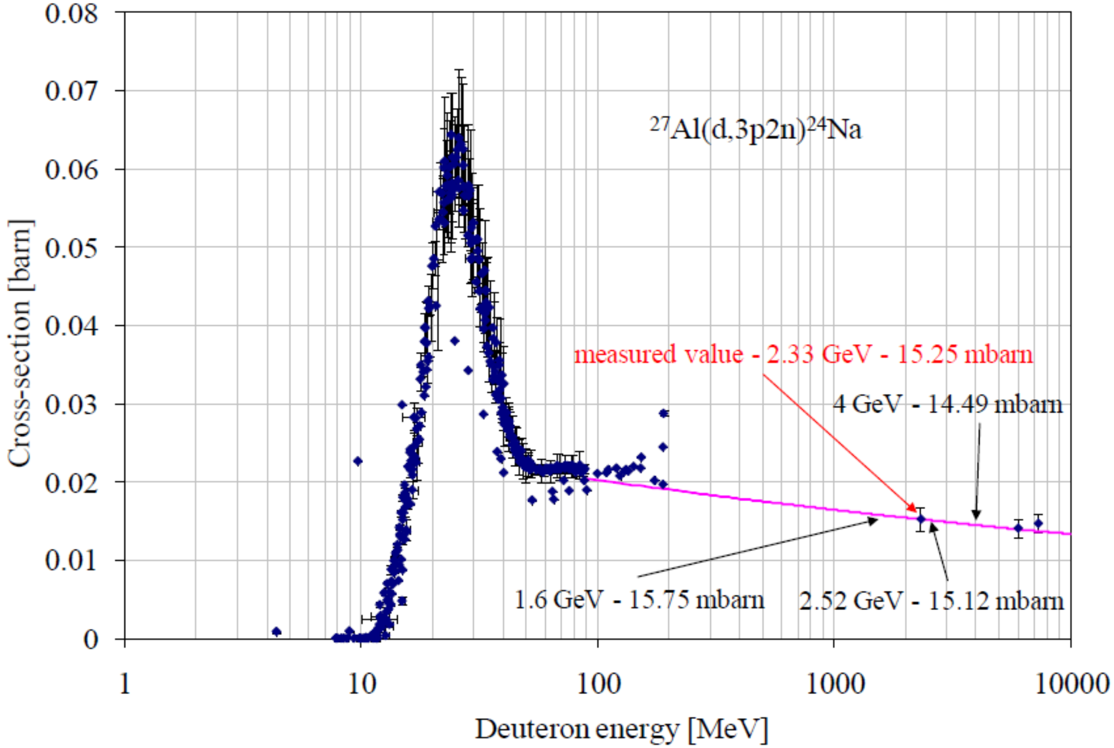
7.3 GeV : **14.7 1.2 mb**

(*Kozma P. and Yanovski V. V.: Application of BaF_2 scintillator to off-line gamma ray spectroscopy, Czech Journal of Physics, 40 (1990) 393-397*)

The choice of monitor $^{27}Al(d,x)^{24}Na$ reaction cross sections

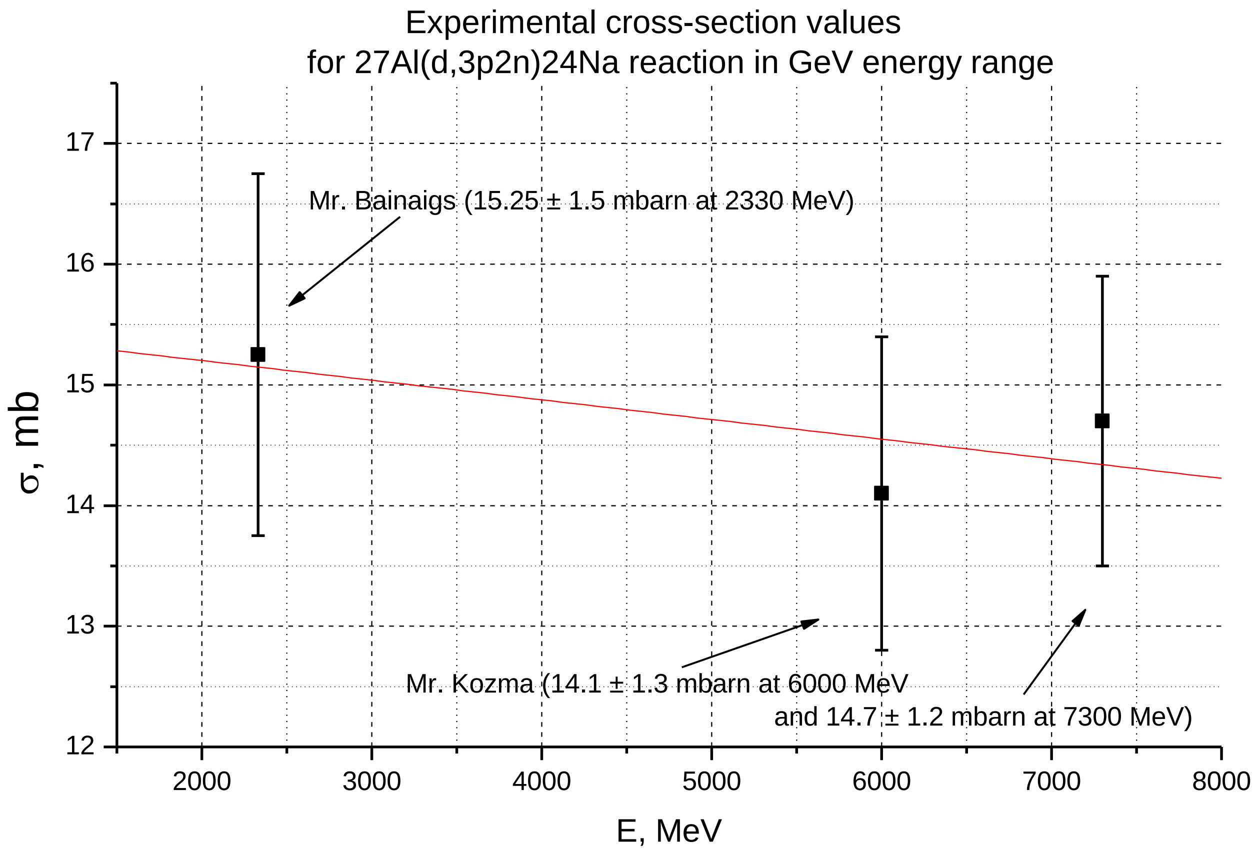
Two methods were proposed:

1) approximation of the available data
2) recalculation of the $^{27}Al(p,x)^{24}Na$ reaction cross sections on protons (it was proposed by W. Westmier).



Doctor Wagner group determined the cross-sections by **approximation** as shown in the figure. (from **Ondřej Svoboda** dissertation**)**

The choice of monitor $^{27}Al(d,x)^{24}Na$ reaction cross sections



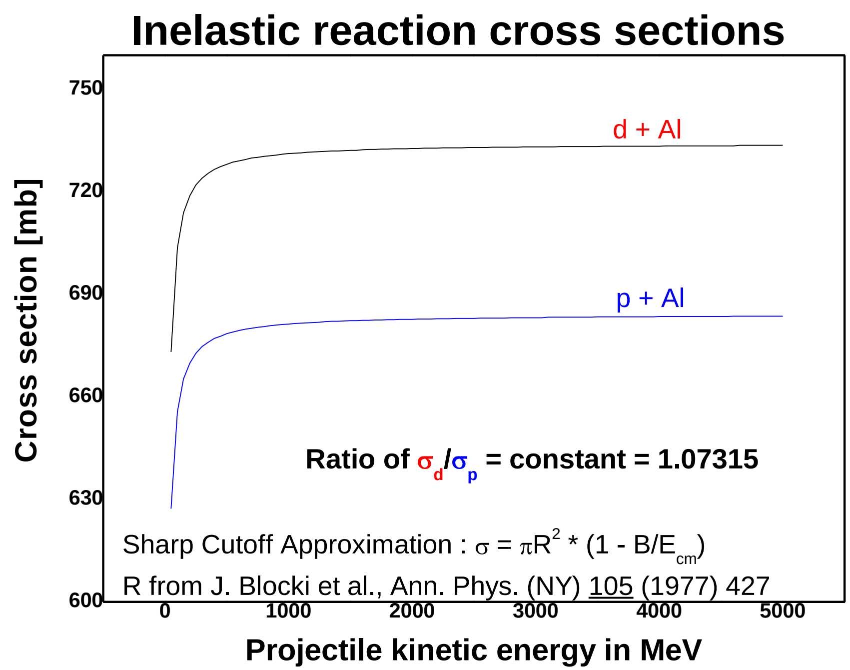
Moreover, it is possible in the range of ~ 2 GeV up to ~ 8 GeV interpolate the three available experimental points (weighted least squares method). Then cross-section are equal

CS(mb)=15.25-0.163*E(GeV)

The choice of monitor $^{27}Al(d,x)^{24}Na$ reaction cross sections

The suggestion of **W.Westmier** :

"The excitation functions for the inelastic reaction cross section of protons and deuterons on aluminium are shown in Figure 5, yielding a constant ratio between the inelastic cross sections at any energy. It is therefore assumed that cross sections for the individual reaction channels leading from ^{27}Al to ^{24}Na in proton and deuteron induced reactions will also run parallel. With this reasonable assumption we can calculate cross sections for the deuteron- induced reaction (σ_d) for any deuteron kinetic energy E_d as :



$$\sigma_{d,Ed} = \sigma_{d,2.33} \bullet (\sigma_{p,Ed} / \sigma_{p,2.33})$$

In this way the cross section for 4 GeV deuterons producing ^{24}Na from aluminium is calculated as **14.628   1.132 mb**."

The choice of monitor $^{27}Al(d,x)^{24}Na$ reaction cross sections

Cross-sections of $^{27}Al(p,x)^{24}Na$ reaction

E, Gev	CS, mb	Error (+-)	Error (%)	Ref.
0.4	10.7	0.21	2.0	[3]
0.591	11	0.5	4.5	[6]
0.6	11.3	0.3	2.7	[5]
0.8	11.2	0.8	7.1	[1]
0.81	10.7	0.13	1.2	[3]
1.2	10.8	0.8	7.4	[1]
1.6	12.3	1	8.1	[1]
1.6	10.18	0.11	1.1	[3]
2.0	9.75	0.11	1.1	[3]
2.6	10.6	0.8	7.5	[1]
7.0	8.94	0.14	1.6	[3]
12	8.1	0.9	11	[2]average
22.4	8.78	0.14	1.6	[3]
28	8.3	0.5	6	[4]

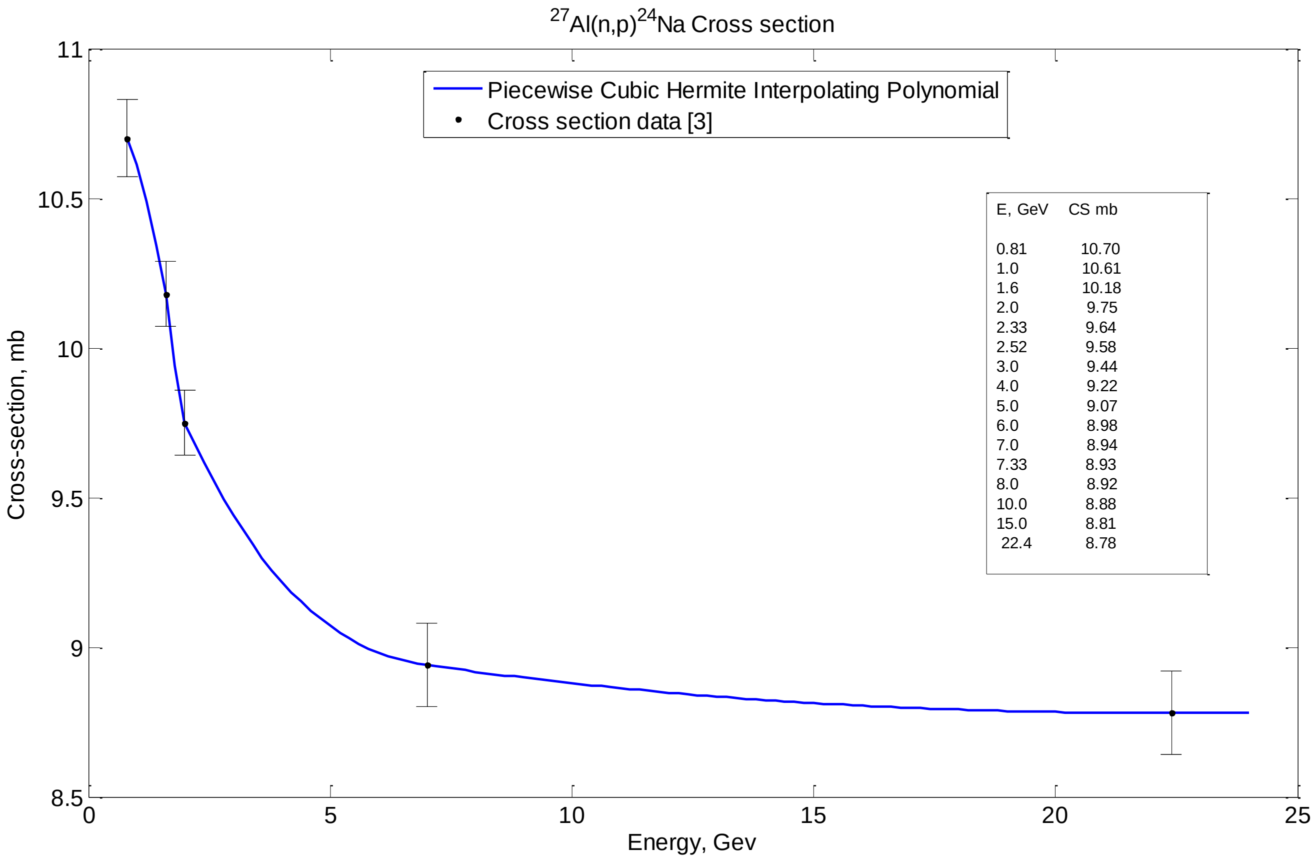
In contrast to the deuteron for the proton cross sections for ^{24}Na experimentally determined up to 28 GeV (see Table)

Note that in the publication

G.L.Morgan et al. Total cross sections for the production of ^{22}Na and ^{24}Na in proton-induced reactions on ^{27}Al from 0.40 to 22.4 GeV.// Nucl. Instrum. Methods in Physics Res., Sect.B; Vol.211 (2003), p.297.

cross-sections on the protons are measured with very high accuracy.

The choice of monitor $^{27}Al(d,x)^{24}Na$ reaction cross sections



The cross sections for the ^{27}Al (p,x)^{24}Na reaction from [3, Morgan], and piecewise cubic interpolation of Hermite polynomials

The choice of monitor $^{27}Al(d,x)^{24}Na$ reaction cross sections

E, GeV	$^{27}Al(p,x)^{24}Na$ CS, mb		$^{27}Al(d,x)^{24}Na$ CS, mb				
	CS experiment (bold) and estimation	Exp. Error	**Recalc. From proton CS**	Linear approx.	Svoboda (* - fig)	Exp	err.
0.81	**10.7**	**0.13**	**16.9**				
1	10.645		**16.8**	15.4	15.9 (V.Wagner)		
1.6	**10.18**	**0.11**	**16.1**	15.3	15.75		
2	**9.75**	**0.11**	**15.4**	*15.2*	15.43 (V.Wagner)		
2.33	9.67		**15.25**	*15.1*	15.25*	**15.25**	**1.5**
2.52	9.56		**15.1**	*15.1*	15.12		
3	9.49		**15.0**	*15.0*			
4	9.27		**14.6** 14.628 - W. Westmier	*14.9*	14.49 (V.Wagner)		
5	9.1		**14.4**	*14.7*			
6	9.01		**14.2**	*14.5*	14.1*	**14.1**	**1.3**
7	**8.94**	**0.14**	**14.1**	*14.4*			
7.33	8.93		**14.1**	*14.3*		**14.7**	**1.2**
8	8.9		**14.0**	14.2	14*		
10	8.85		**14.0**	13.9			
15	8.81		**13.9**	13.1			
22.4	**8.78**	**0.14**	**13.8**	11.9			

Total intensities

(Run March 2011, December 2011, March 2012)

Run	Energy of deuterons, GeV	Exposure time of activation detectors	Total deuteron intensity	γ-spectrometers
March 2011	**2**	start: 06.03.11 03^{28} finish: 06.03.11 20^{56}	**$1.69\cdot10^{13}$**	“ЯСНАПП”, detector ORTEC(B)
	4	start: 08.03.11 10^{39} finish: 09.03.11 05^{56}	**$1.41\cdot10^{13}$**	
	6	start: 20.03.11 21^{46} finish: 21.03.11 14^{14}	**$1.94\cdot10^{13}$**	
December 2011	**1**	start: 14.12.11 17^{39} finish: 15.12.11 08^{05}	**$1.47\cdot10^{13}$**	“ЯСНАПП”, detector ORTEC(C), дет. к.216
	4	start: 16.12.11 19^{57} finish: 17.12.11 08^{21}	**$1.96\cdot10^{13}$**	
	8	start: 19.12.11 00^{59} finish: 19.12.11 05^{10}	**$6.3\cdot10^{10}$** (defined by IC)	
March 2012 (the intensity of the beams will be corrected)	**1**	start: 10.03.12 16^{04} finish: 10.03.12 21^{00}	**$1.9\cdot10^{13}$**	Detector room 338
	4	start: 15.03.12 00^{18} finish: 15.03.12 09^{10}	**$2.7\cdot10^{13}$**	
	8	start: 19.03.12 06^{26} finish: 19.03.12 15^{27}	**$3.7\cdot10^{12}$**	

Determination of the accumulated plutonium

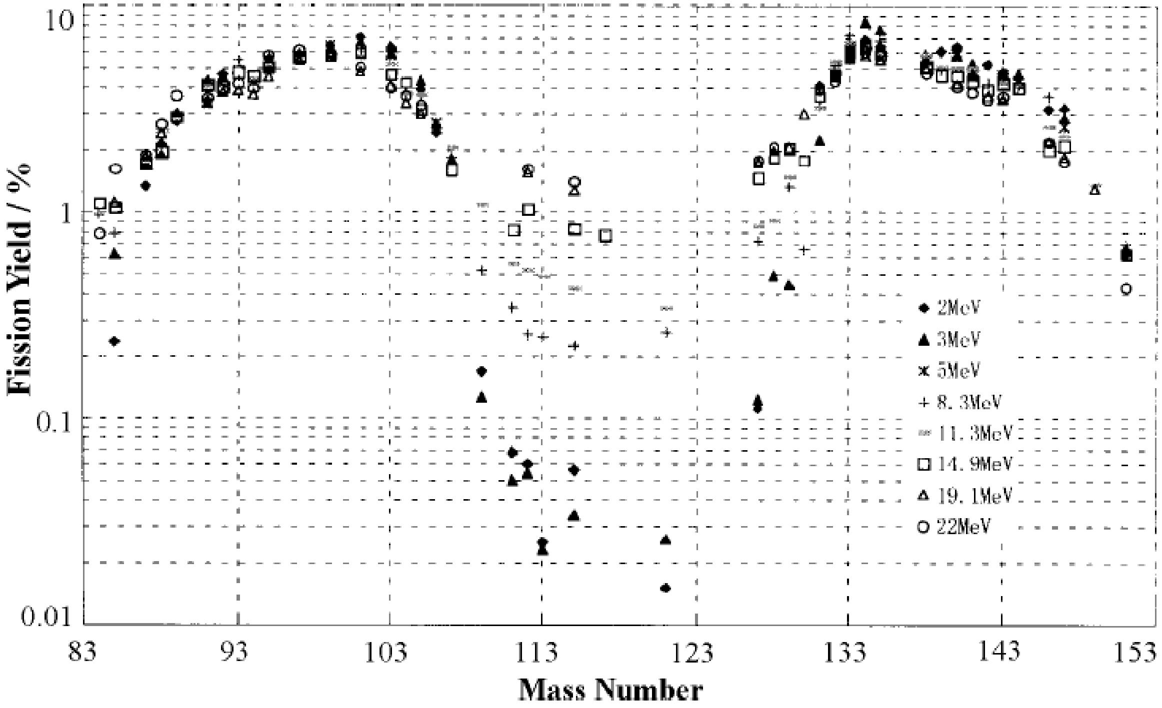
The number of ^{238}U neutron radiative capture reactions corresponds to the number of ^{239}Pu nuclei, which are formed by the chain of ^{239}U β-decay:

$$^{238}U(n,\gamma)^{239}U\ \beta^-\ (23{,}54\ min) \rightarrow {}^{239}Np\ \beta^-\ (2{,}36\ d) \rightarrow {}^{239}Pu$$

After the irradiation of uranium assembly γ-spectra of irradiated uranium foils were measured. Before measurement uranium foils were exposure for more than 4 hours to reach 99.9% of the decays of ^{239}U. The number of neutron capture reactions of uranium-238 was determined by measuring the activity of the ^{239}Np nuclide.

Mass distribution of fission products of ^{nat}U at different neutron energies

The number of nuclear fissions was determined by averaging the results for the following fragments :

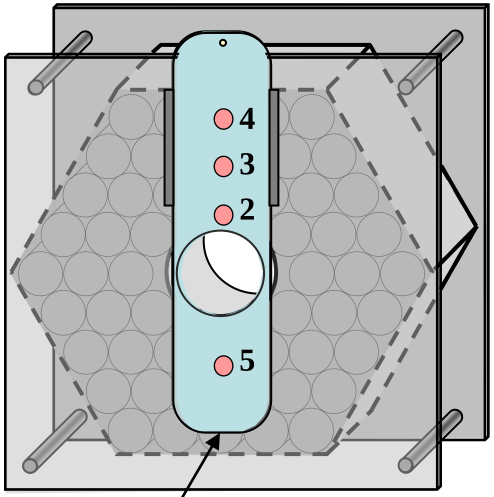


^{97}Zr (5.7%), ^{131}I (3.6%) ^{133}I (6.3%), ^{143}Ce(4.3%)

the cumulative yield, averaged over data for neutrons with energy 2-22 MeV is shown in brackets.

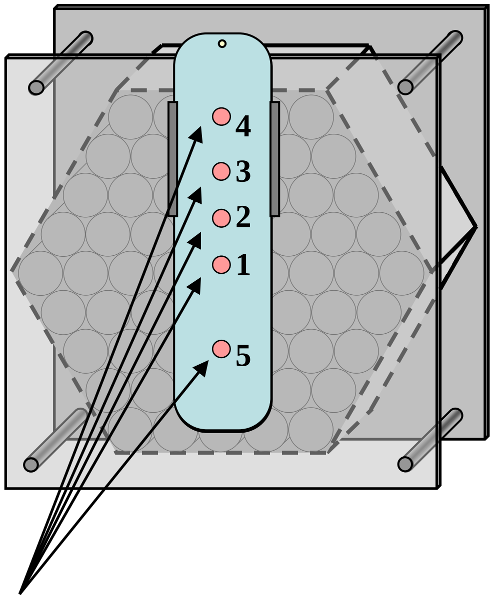
Location of detectors on the plate

Section № 1



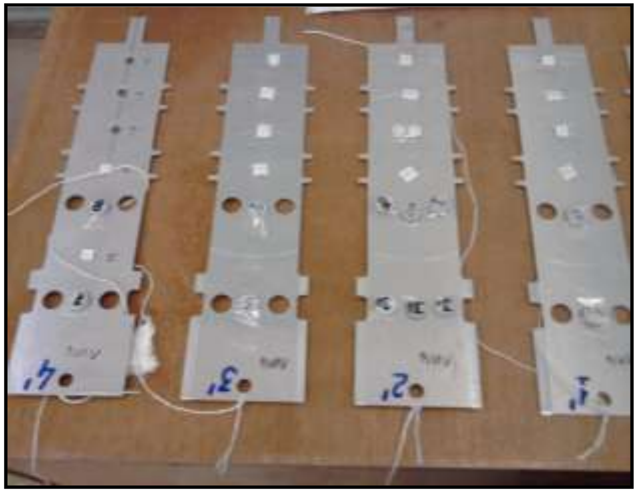
Location of the detector plate

Sections № 2,3,4,5



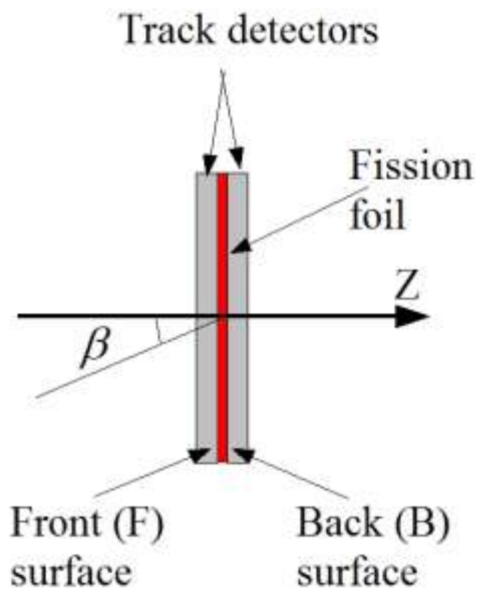
The scheme of ^{nat}U foils location on the detector plate, used in the experiment. Each plate had 5 positions at different distances from the radial symmetry axis of the target

Detector plates



- **4** R = 120 mm
- **3** R = 80 mm
- **2** R = 40 mm
- **1** R = 0
- **5** R = -80 mm

γ-track technique



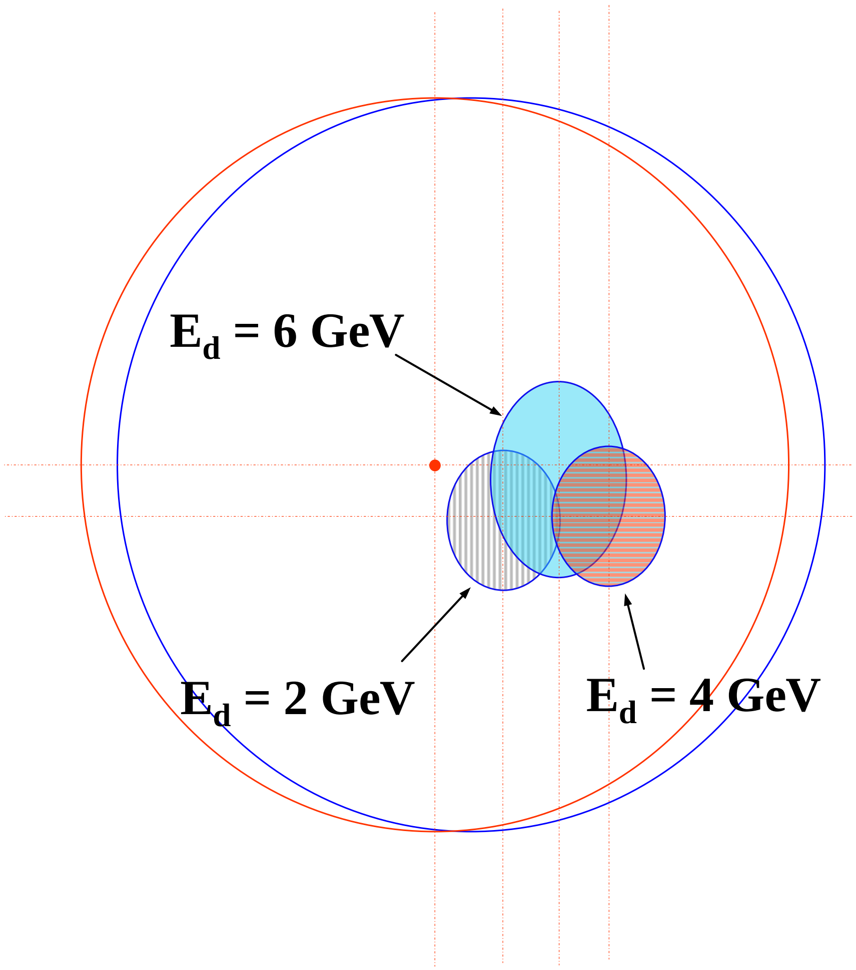
Solid nuclear track detector
(synthetic mica)
+
radiator (metal foil)

<u>Detector scheme</u>:
uranium metal foil (diameter 7 mm, thickness 1 mm) used both as an activation detector, and a radiator for SSNTD (synthetic mica - fluorinephlogopite).

<u>SSNTD</u>:
Track counting and fission rate determination

<u>Metal foil</u>:
Activation measurement and capture reaction rate determination

Deuteron beam parameters determination (2, 4 and 6 GeV deuterons)



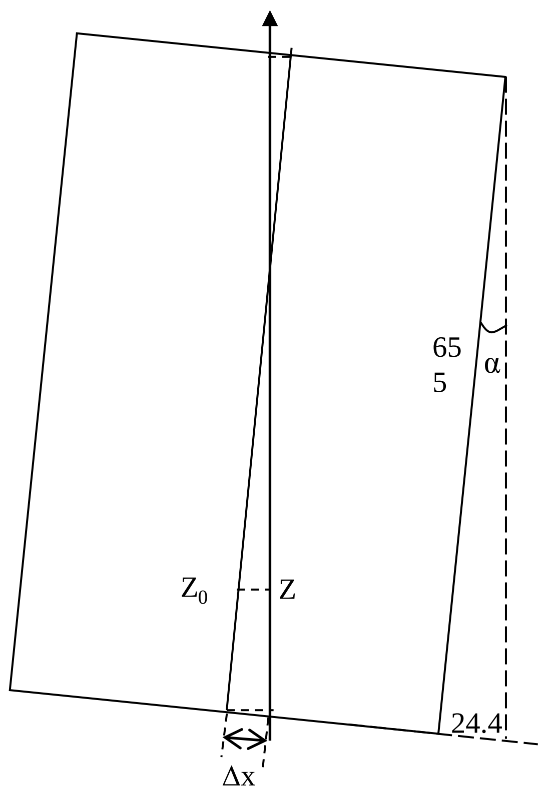
Results obtained by SSNTD method (Patapenka et al.)

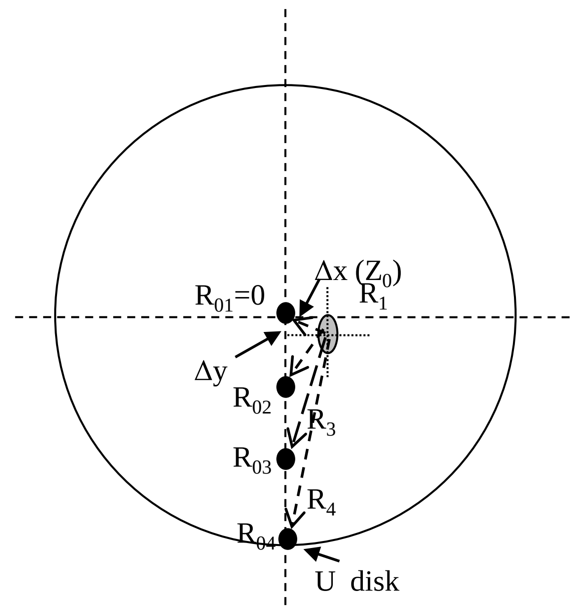
E, GeV	FWHM of distributions (mm)		Beam shift (mm)	
	X direction	Y direction	in X	in Y
2	13	16	7.8	6.4
4	13	16.1	20	5.9
6	15.6	22.4	14.2	1.8

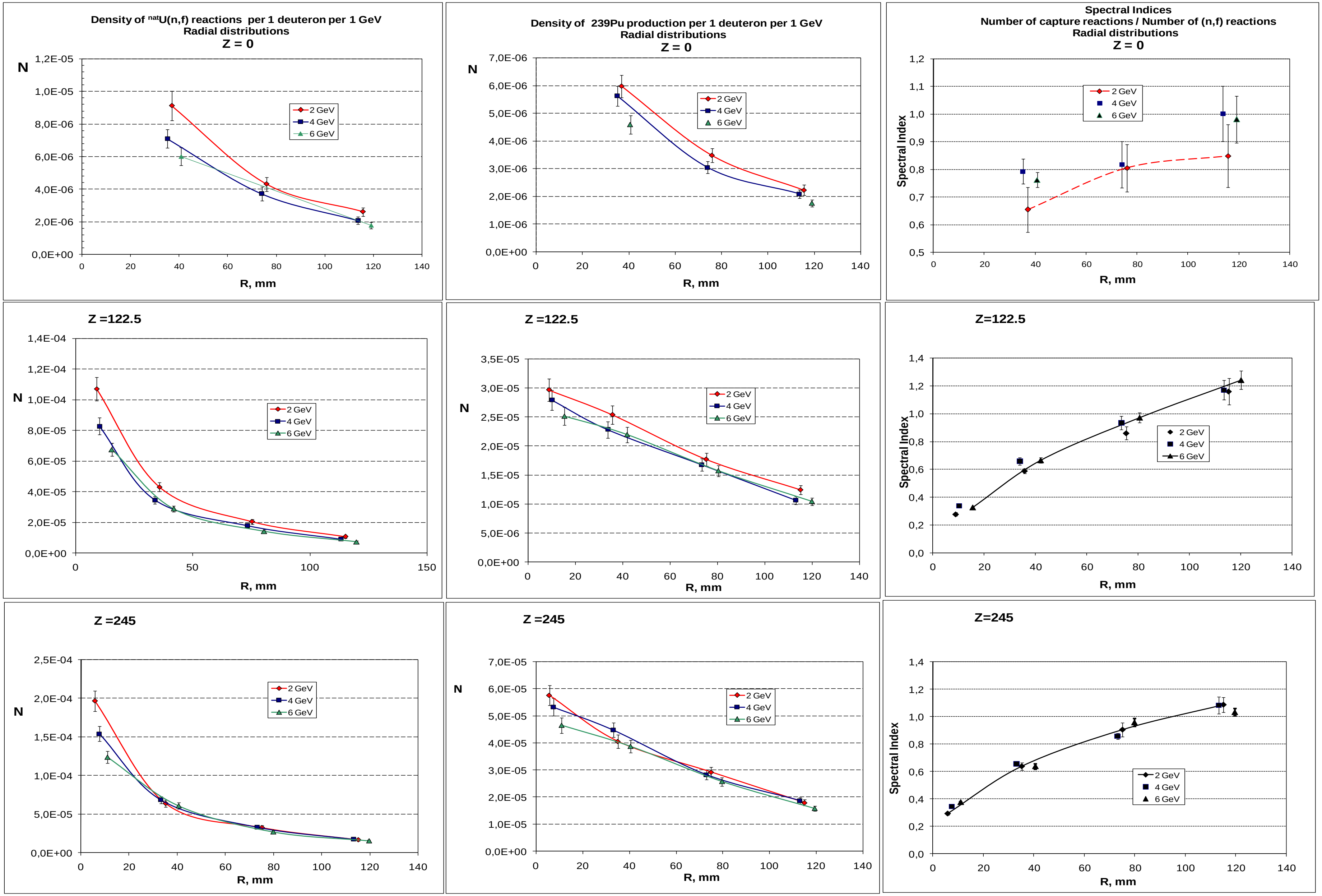
Correction on beam position

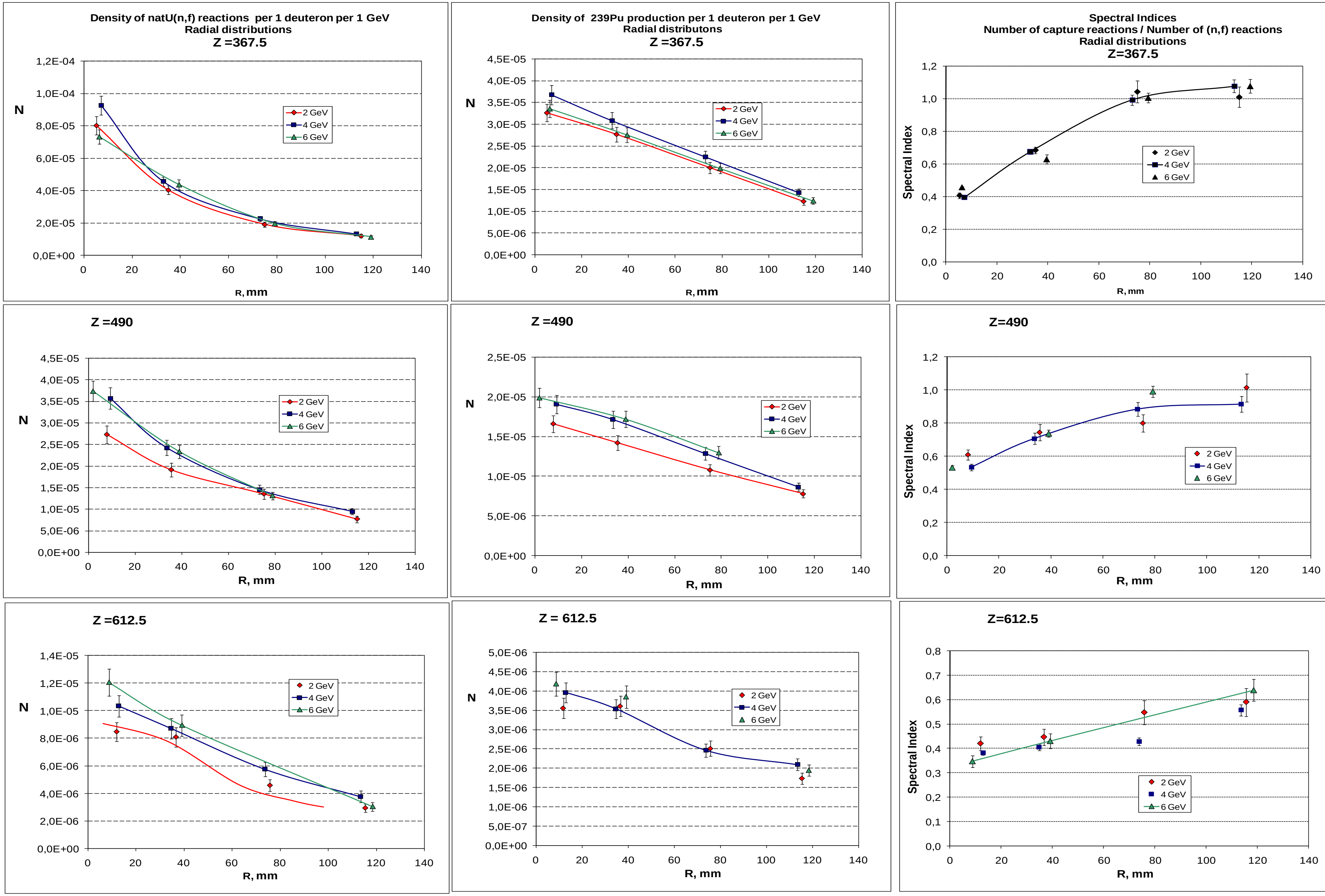
$$R = \sqrt{\left(R_0 - \Delta y\right)^2 + \left(\Delta x \cdot \cos\alpha - Z_0 \sin\alpha\right)^2}$$

$$Z = Z_0 \cdot \cos\alpha + \Delta x \cdot \sin\alpha \approx Z_0$$

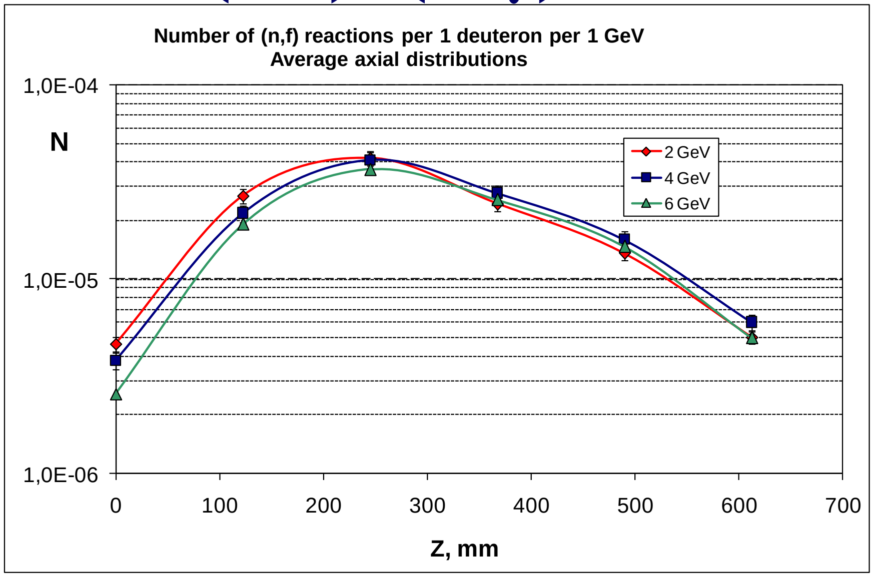


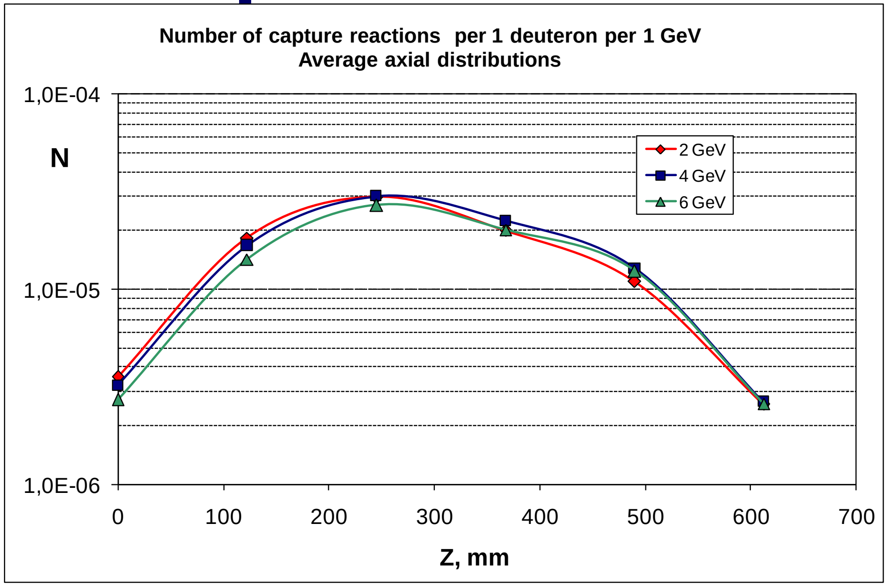



Density of natU(n,f) reactions per 1 deuteron per 1 GeV
Radial distributions
Z = 0
N
1,2E-05
1,0E-05
8,0E-06
6,0E-06
4,0E-06
2,0E-06
0,0E+00
0 20 40 60 80 100 120 140
R, mm
2 GeV
4 GeV
6 GeV
Density of 239Pu production per 1 deuteron per 1 GeV
Radial distributions
Z = 0
N
7,0E-06
6,0E-06
5,0E-06
4,0E-06
3,0E-06
2,0E-06
1,0E-06
0,0E+00
0 20 40 60 80 100 120 140
R, mm
2 GeV
4 GeV
6 GeV
Spectral Indices
Number of capture reactions / Number of (n,f) reactions
Radial distributions
Z = 0
Spectral Index
1,2
1,1
1,0
0,9
0,8
0,7
0,6
0,5
0 20 40 60 80 100 120 140
R, mm
2 GeV
4 GeV
6 GeV
Z =122.5
N
1,4E-04
1,2E-04
1,0E-04
8,0E-05
6,0E-05
4,0E-05
2,0E-05
0,0E+00
0 50 100 150
R, mm
2 GeV
4 GeV
6 GeV
Z =122.5
N
3,5E-05
3,0E-05
2,5E-05
2,0E-05
1,5E-05
1,0E-05
5,0E-06
0,0E+00
0 20 40 60 80 100 120 140
R, mm
2 GeV
4 GeV
6 GeV
Z=122.5
Spectral Index
1,4
1,2
1,0
0,8
0,6
0,4
0,2
0,0
0 20 40 60 80 100 120 140
R, mm
2 GeV
4 GeV
6 GeV
Z =245
N
2,5E-04
2,0E-04
1,5E-04
1,0E-04
5,0E-05
0,0E+00
0 20 40 60 80 100 120 140
R, mm
2 GeV
4 GeV
6 GeV
Z =245
N
7,0E-05
6,0E-05
5,0E-05
4,0E-05
3,0E-05
2,0E-05
1,0E-05
0,0E+00
0 20 40 60 80 100 120 140
R, mm
2 GeV
4 GeV
6 GeV
Z=245
Spectral Index
1,4
1,2
1,0
0,8
0,6
0,4
0,2
0,0
0 20 40 60 80 100 120 140
R, mm
2 GeV
4 GeV
6 GeV

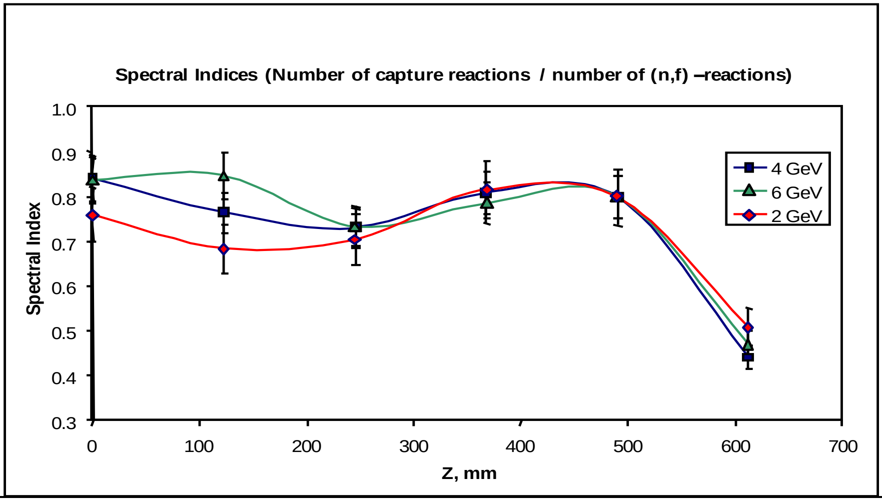

Density of natU(n,f) reactions per 1 deuteron per 1 GeV
Radial distributions
Z =367.5
Density of 239Pu production per 1 deuteron per 1 GeV
Radial distributons
Z =367.5
Spectral Indices
Number of capture reactions / Number of (n,f) reactions
Radial distributions
Z=367.5
Z =490
Z =490
Z=490
Z =612.5
Z = 612.5
Z=612.5
N
Spectral Index
R, mm
2 GeV
4 GeV
6 GeV

Average spatial distributions of (n,f), (n,γ) reactions and spectral index



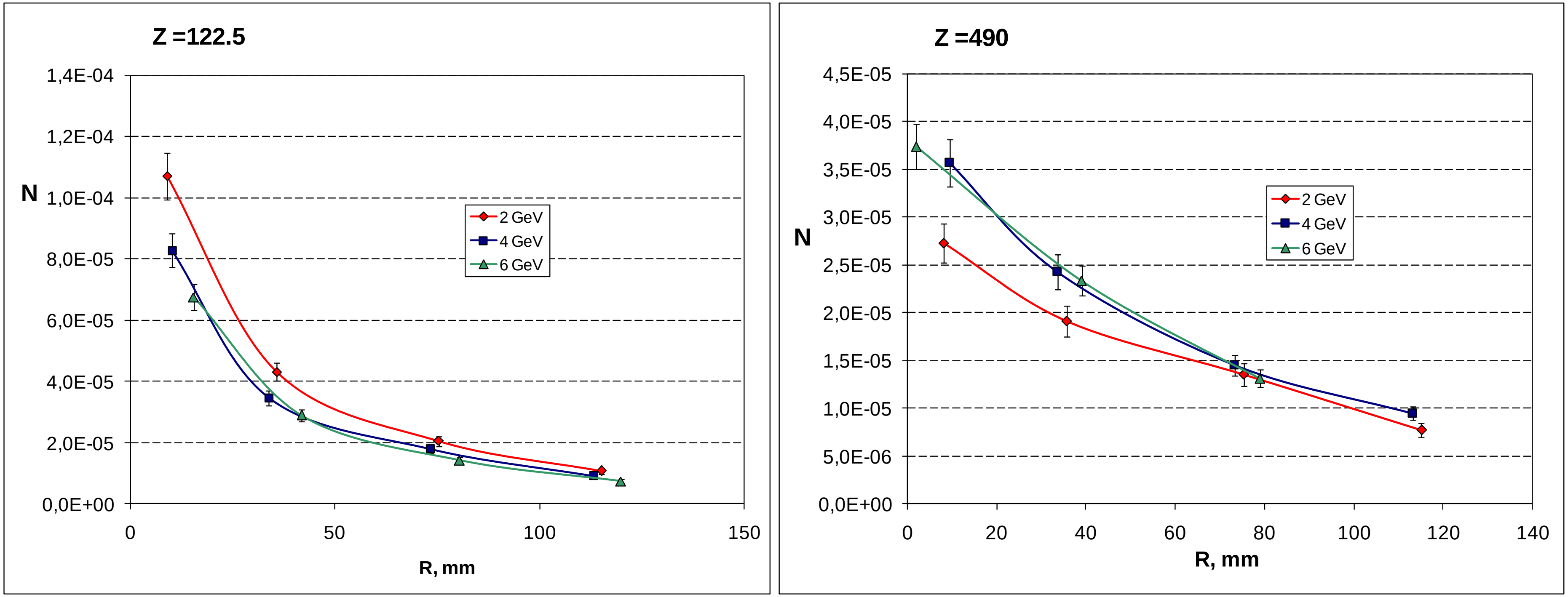


Densities of the number of fissions, the number of plutonium nuclei and spectral indices averaged over the radial cross sections of the uranium target. The maximum values of density of fission and plutonium accumulation is approximately at a distance of about 120 mm from the entrance of the beam at the target. This is observed for all similar axial distributions of deuterons energies from 1 to 8 GeV.

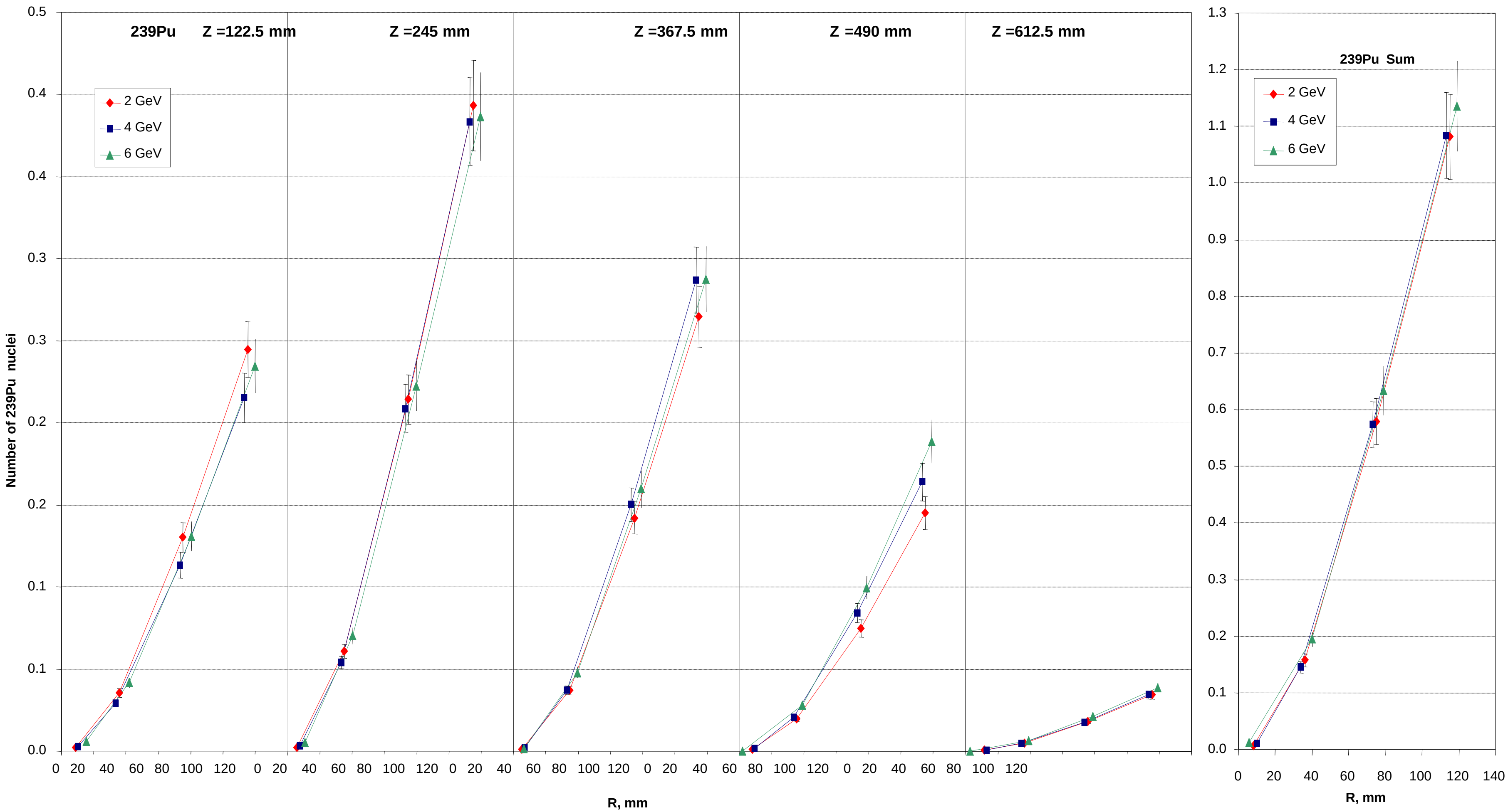


Comparison of results at the different deuteron energies

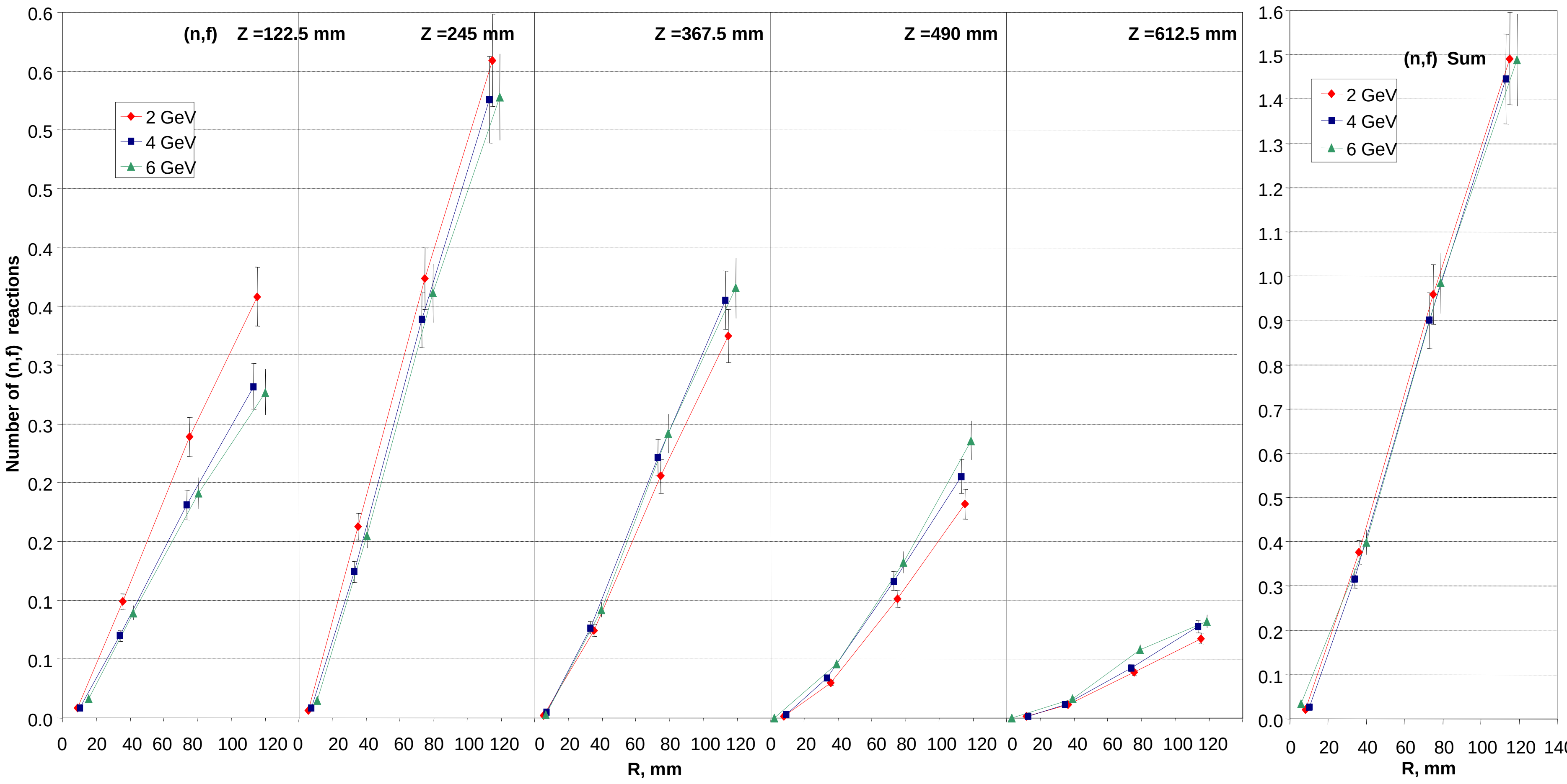
Density of $^{nat}U(n,f)$ reactions per 1 deuteron per 1 GeV
Radial distributions



Integral distribution of (n,γ) reaction
(dependencies on uranium target radius)



Integral distribution of (n,f) reaction
(dependencies on uranium target radius)



Integral numbers of ^{239}Pu accumulation and ^{nat}U fission in the volume of uranium target of assembly "QUINTA-M"

Run	E, GeV	^{239}Pu	^{nat}U fission
March 2011	2	(7.0 0.3) 0.8	(8.8 0.4) 1.0
	4	(7.2 0.4) 0.8	(8.8 0.4) 1.0
	6	(6.9 0.3) 0.7	(8.3 0.4) 0.9
Dec. 2011	1	(11.6 0.6) 1.2	(10.2 0.5) 1.1
	4	(10.9 0.5) 1.1	(8.7 0.4) 1.0
March 2012	1	(11.2 0.6) 1.2	(9.6 0.5) 1.1
	4	(10.8 0.5) 1.1	(8.8 0.4) 1.0
	8	(10.5 0.5) 1.1	(9.8 0.5) 1.1

CONCLUSIONS

- The spectral index changes from the deuteron beam axis to the periphery of the uranium target from about 0.3 to 1 for “QUINTA-M” and does not depend on the energy of the primary beam for 2, 4 and 6 GeV deuteron.

- The total number of fissions in the volume of uranium target of “QUINTA-M”, that was defined by activation method, remains approximately constant within our statistical errors for all energies of the deuteron beam in the range from 1 to 8 GeV (calculated per a deuteron and per 1 GeV primal energy of the deuteron).

- The total number of accumulating ^{239}Pu nuclei in the whole volume of the uranium target, calculated per a deuteron and per 1 GeV energy of the deuteron, does not depend on the primal energy of the deuteron beam, but increases by more than 50% in the presence of a lead blanket.

CONCLUSIONS

- Type of spatial distributions of the density of number of uranium fission and the number of accumulating ^{239}Pu per unit of power of the primary deuteron beam energy depends on the deuteron energy: the growth of primary energy deuterons decreases the number density of uranium fission and the number of ^{239}Pu nuclei accumulation in the near zone to the entrance of the deuteron beam at the target and at the same time, there is an increase in the density of uranium fission and ^{239}Pu accumulation to the periphery of the target.

- for a given sizes of uranium targets of “QUINTA-M” assembly, for a deuteron energies exceeding 1 GeV, we can’t experimentally estimate (at least, by the activation technique) required size of the uranium target, satisfying it kvasi-infinity and, therefore, it is impossible to estimate the total number of fissions and accumulated ^{239}Pu nuclei for kvasi-infinit target. This requires the measurement of uranium targets of larger mass.

Thank you for your attention!