RESULTS OF EXPERIMENTS 2012-2013 WITH MASSIVE URANIUM TARGET SETUP QUINTA AT NUCLOTRON AND PLANS FOR 2014 -2016.

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RESULTS OF EXPERIMENTS 2012-2013 WITH MASSIVE URANIUM TARGET SETUP QUINTA AT NUCLOTRON AND PLANS FOR 2014 -2016

Content

- ***Introduction***
- ***Results of experiments 2012-2013***
- ***Discussion of results***
- ***Plans for 2014 - 2016***
- ***Conclusion***

INTRODUCTION

- **This report discusses the results obtained after ISINN-20 in the framework of ADS project "Energy and Transmutation RAW" (E&T RAW).**

- **The project is aimed mainly at study of possibilities to use in ADS deep subcritical natural (depleted) uranium or thorium active core (AC) with very hard neutron spectrum inside for effective burning of the core material together with spent nuclear fuel (SNF).**
- **In principle such ADS permits to utilize SNF with simultaneous energy production.**

- **For the practical implementation of the ADS with deep subcritical (DS) AC recently there was proposed the new electro-nuclear scheme, so-called Relativistic Nuclear Technologies (RNT).**

- **It had been discussed at ISINN 19-20 in some detail including its main technical aspects.**

- **For present consideration it is important that RNT proposes to use quasi-infinite (QI) AC with negligible leakage of neutrons produced by incident high energy particles inside of AC serving as beam target.**

- **So to clear out a feasibility of RNT approach it is necessary to study the spatial and spectral characteristics of neutron fields within QI AC and basic nuclear reactions induced by these neutrons.**
- **First of all these are the spallation reactions, (n,f), (n,γ) and (n,xn) on nuclei of AC and (n,f), (n,γ) and (n,xn) on minor actinides and fission products.**
- **Above reactions provide a transmutation of long-lived components of SNF and determine the amount of energy produced in AC.**
- **All of these reactions will be considered in more detail in talks J. Adam and L. Zavorka during present session.**

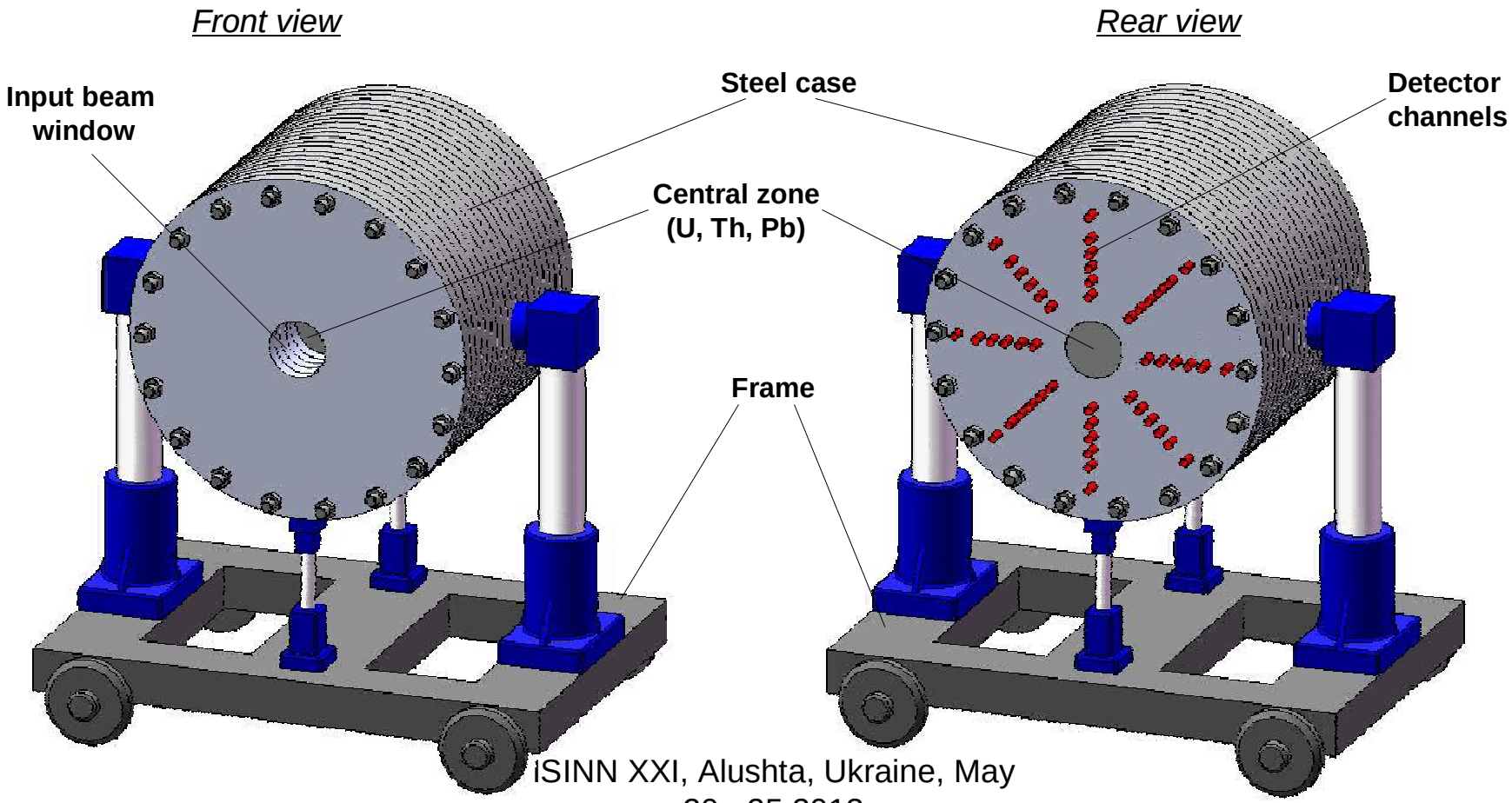
- **For full scale modeling of RNT scheme JINR has the quasi-infinite (~20 tones) AC –**

  Big depleted URANium target assemly (BURAN) and high energy beams of super conducting accelerator NUCLOTRON.
- **As a preparatory stage for main experiments with BURAN there was used an intermediate size massive (512 kg) target assembly (TA) QUINTA from natural metallic uranium.**
- **TA QUINTA models the central part of BURAN and allows to work out and to test the measurement and data analysis procedures for main experiments.**

Target assembly BURAN with replacement central zone

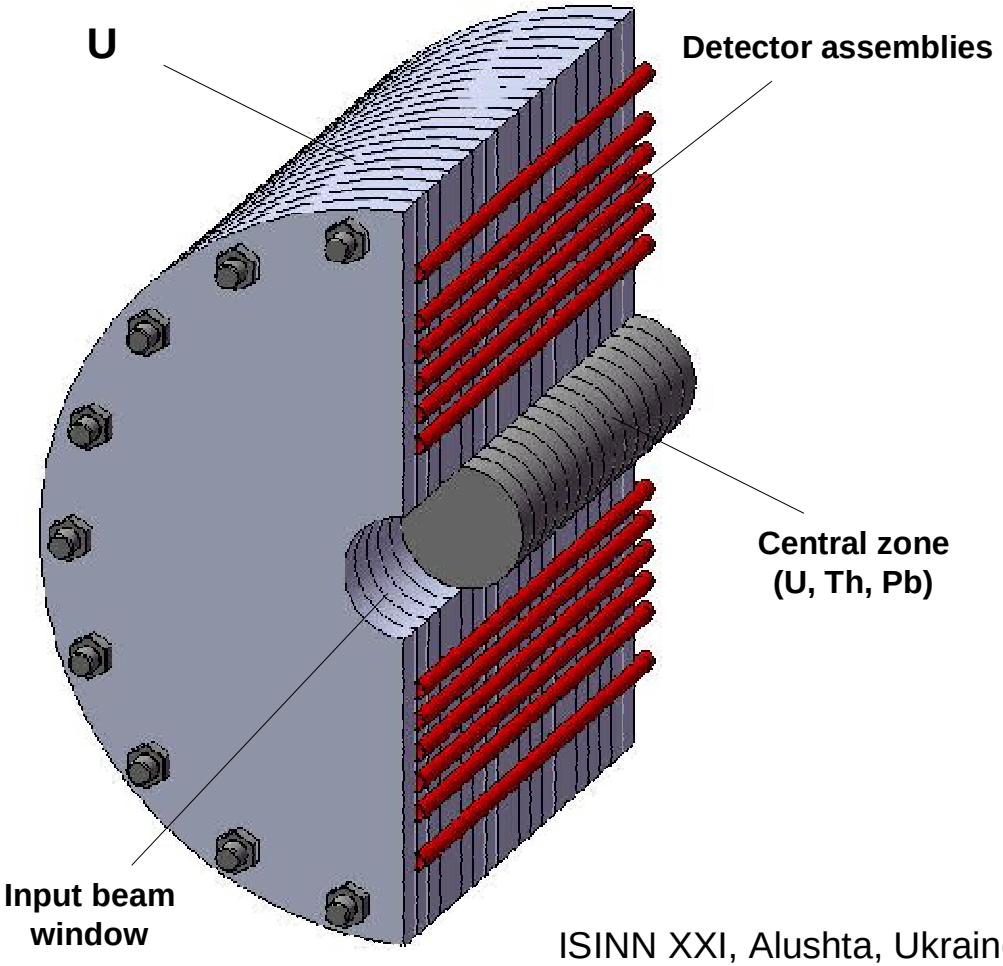
Mass of uranium – 19.5 т.
Diameter – 1,2 м.
Length – 1 м.

Materials of central zone – U, Th, Pb.
Diameter of central zone – 0,2 м.

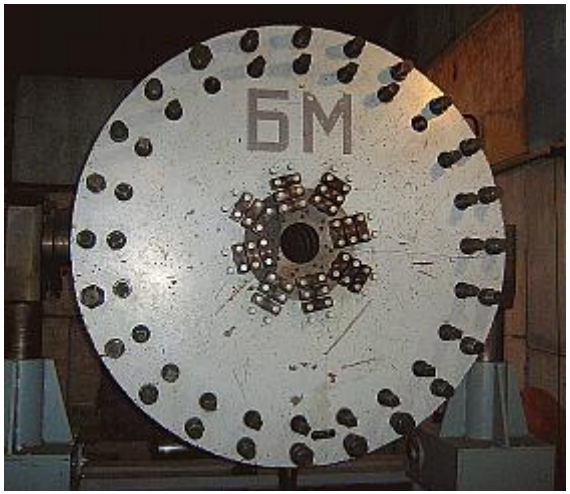


Target assembly BURAN with replacement central zone

Longitudinal section of the BURAN together with central zone and detector sets

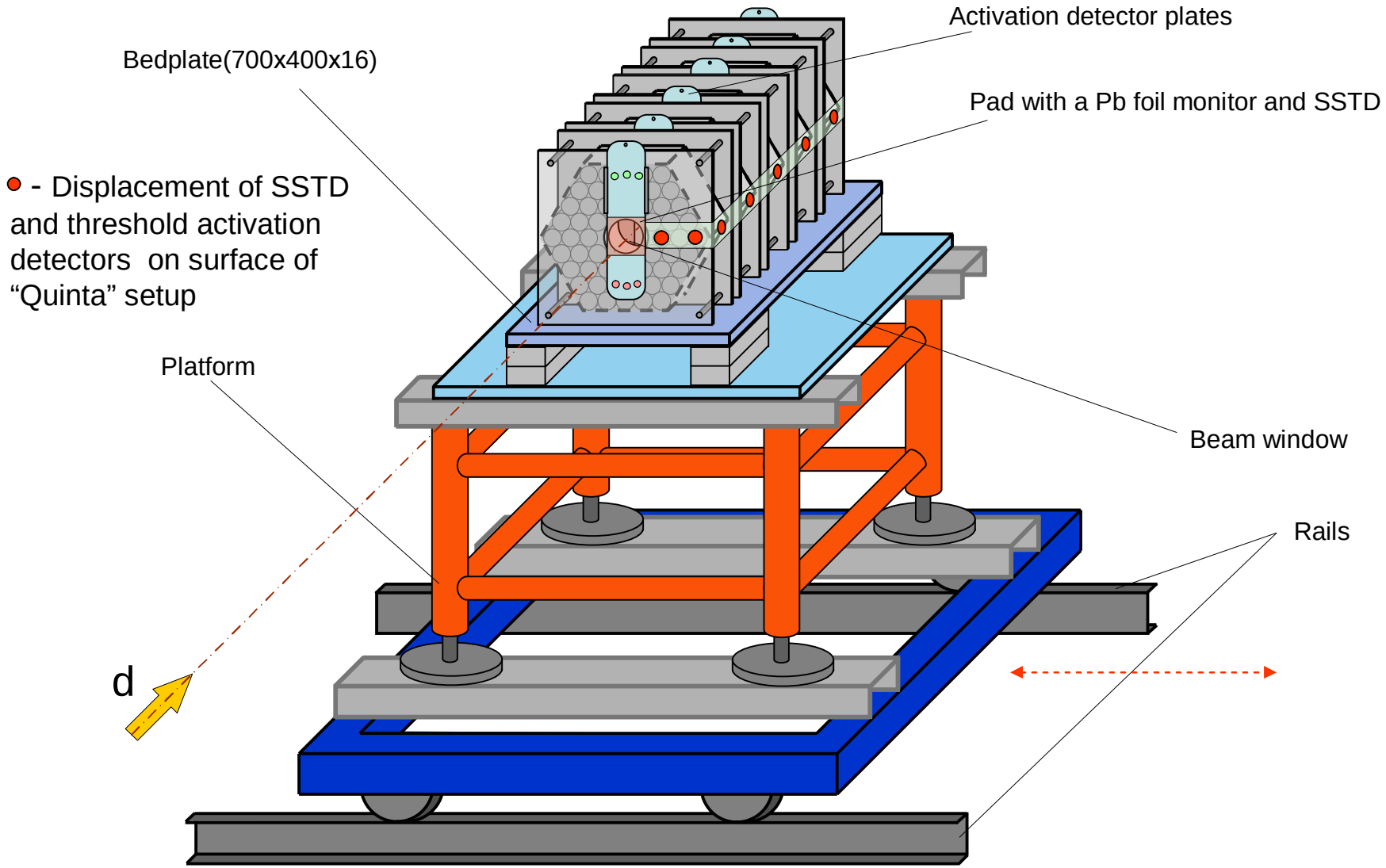


Front view photo

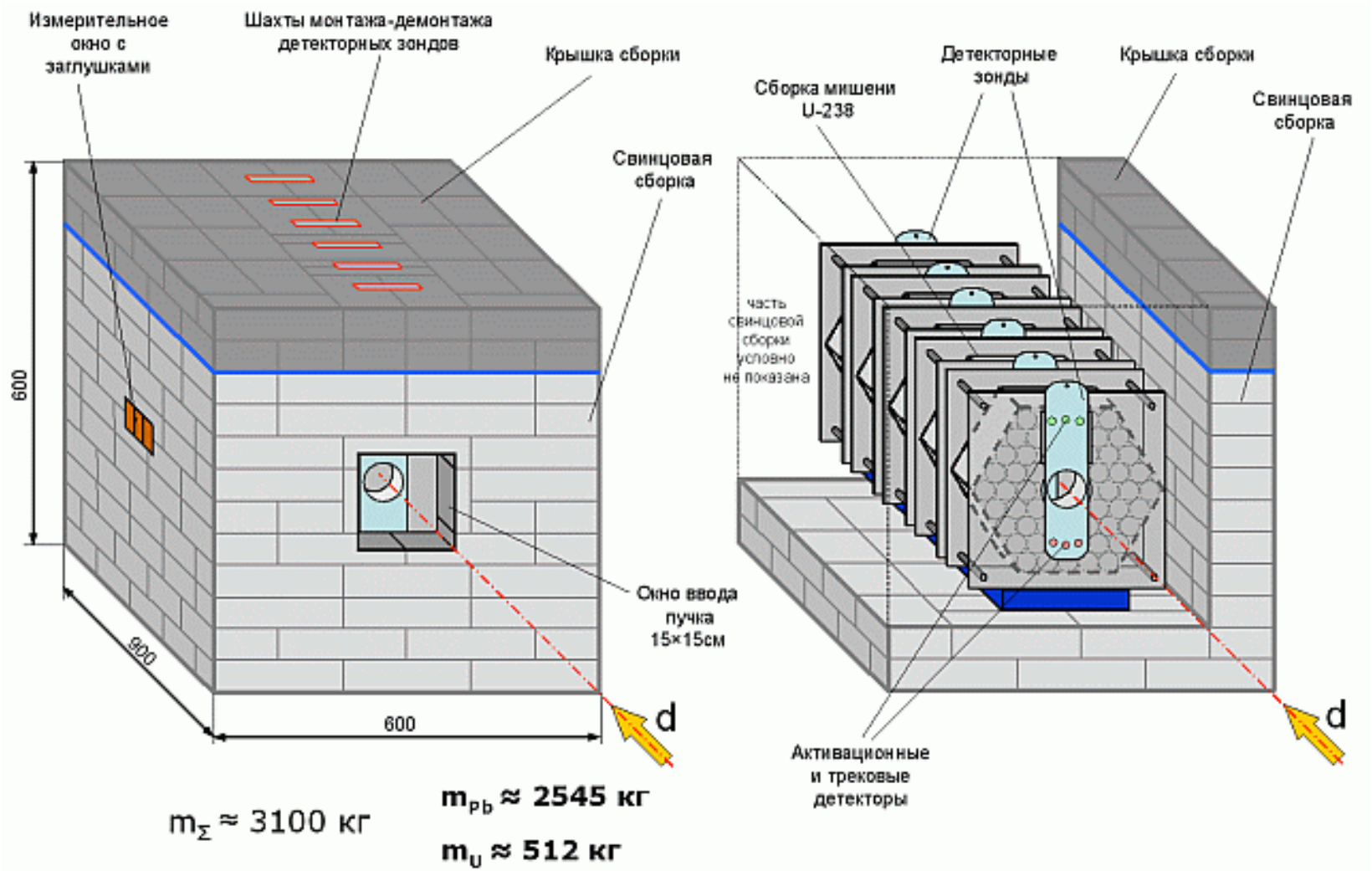


Rear view photo

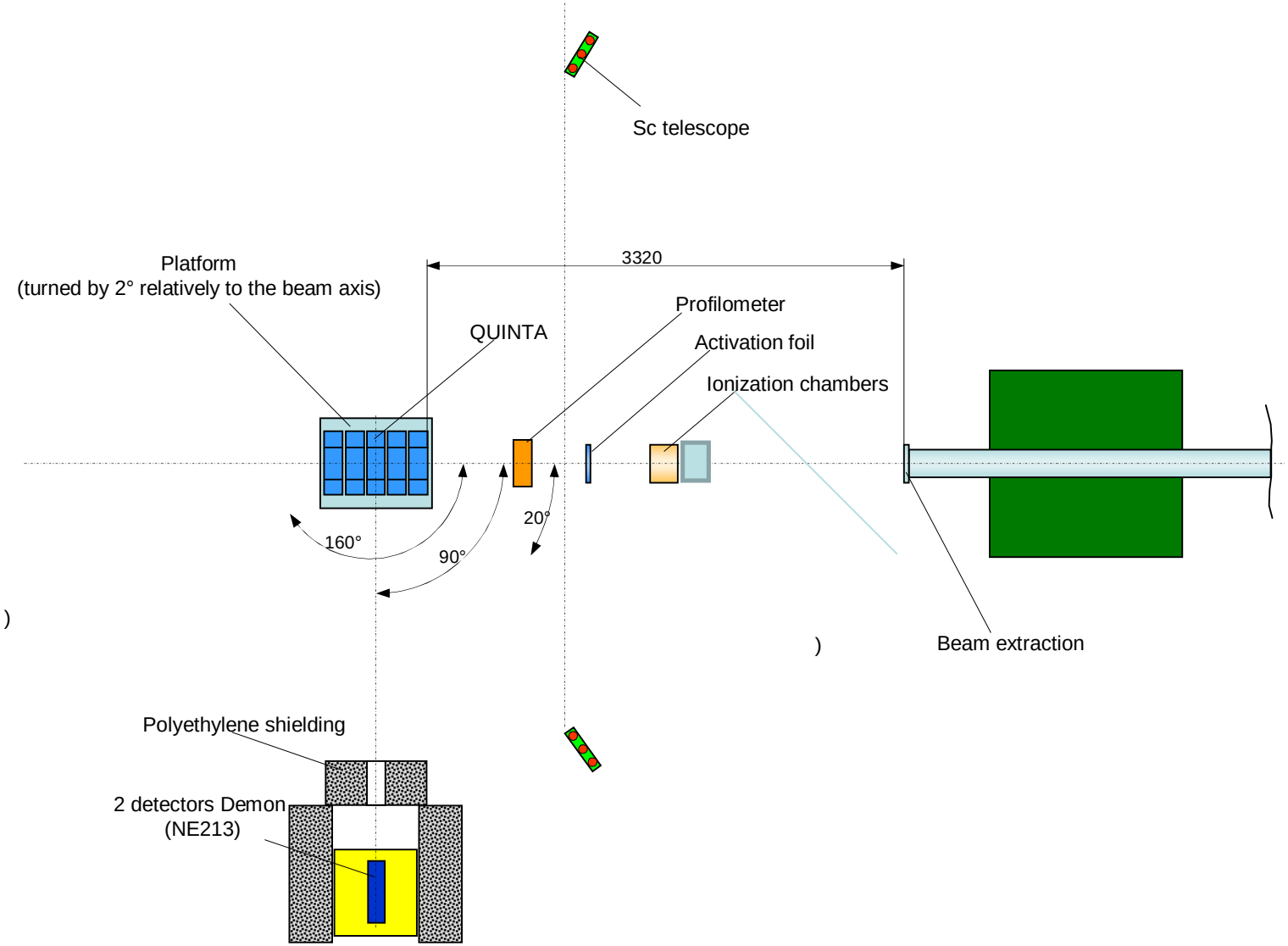
Target assembly QUINTA at the irradiation position (March 2011)



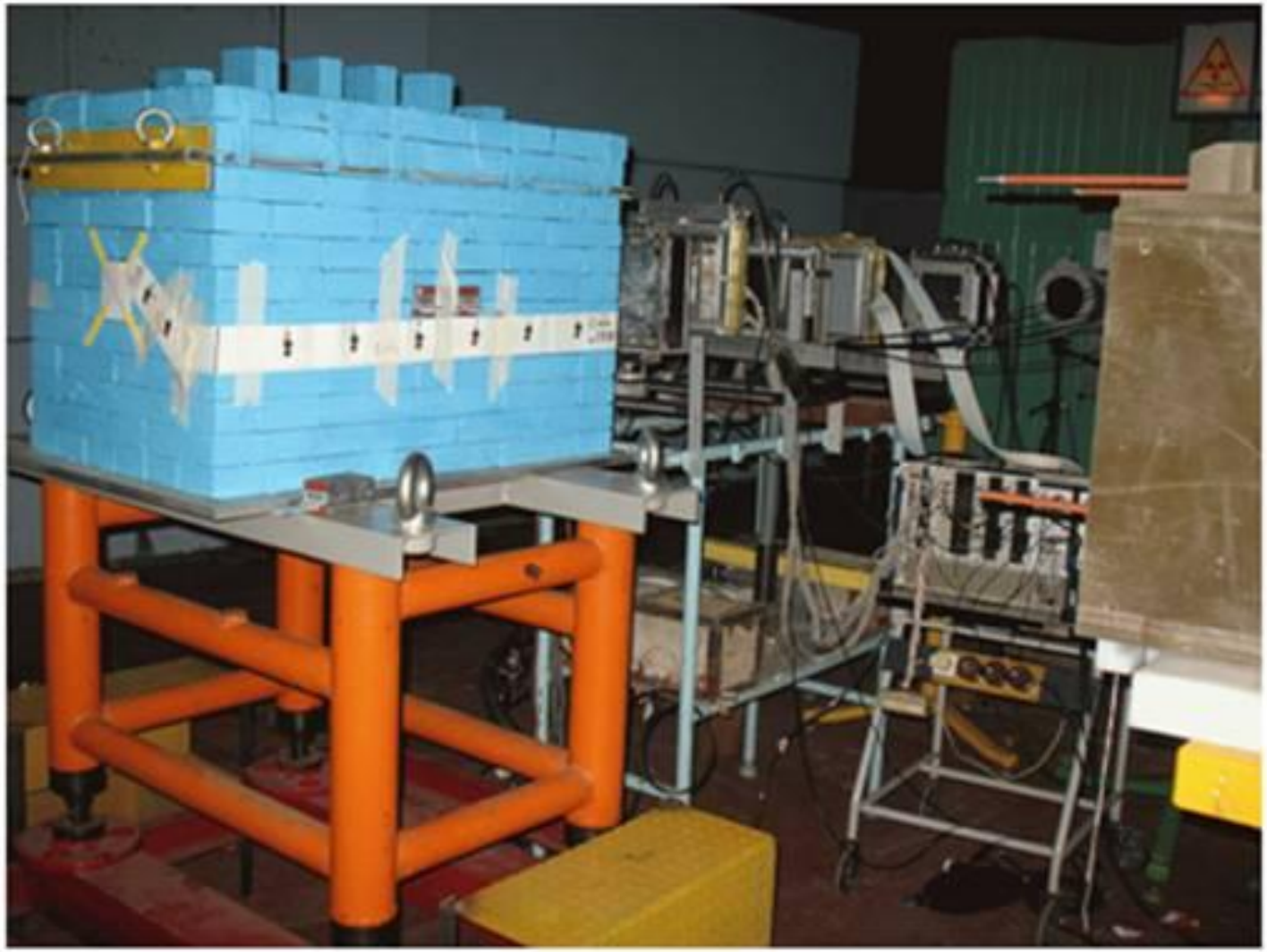
TA QUINTA with lead blanket



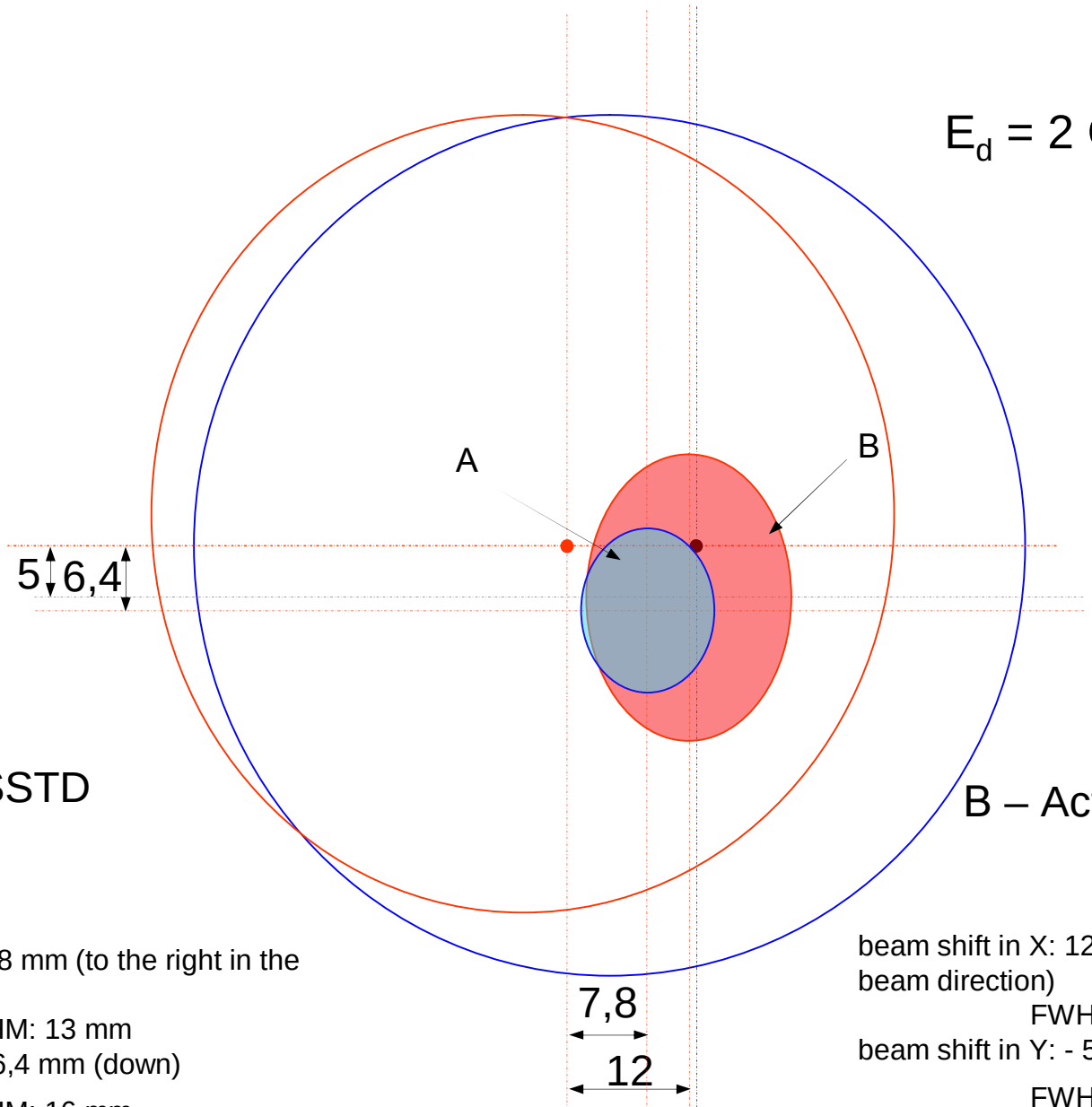
SCHEME OF EXPERIMENTS WITH TA QUINTA



TA QUINTA ON IRRADIATION POSITION (March 2013)



CONTROL OF BEAM POSITION WITHIN INPUT WINDOW (FRONT VIEW)



A – SSTD

B – Activation method

beam shift in X: 7,8 mm (to the right in the beam direction)

FWHM: 13 mm

beam shift in Y: - 6,4 mm (down)

FWHM: 16 mm

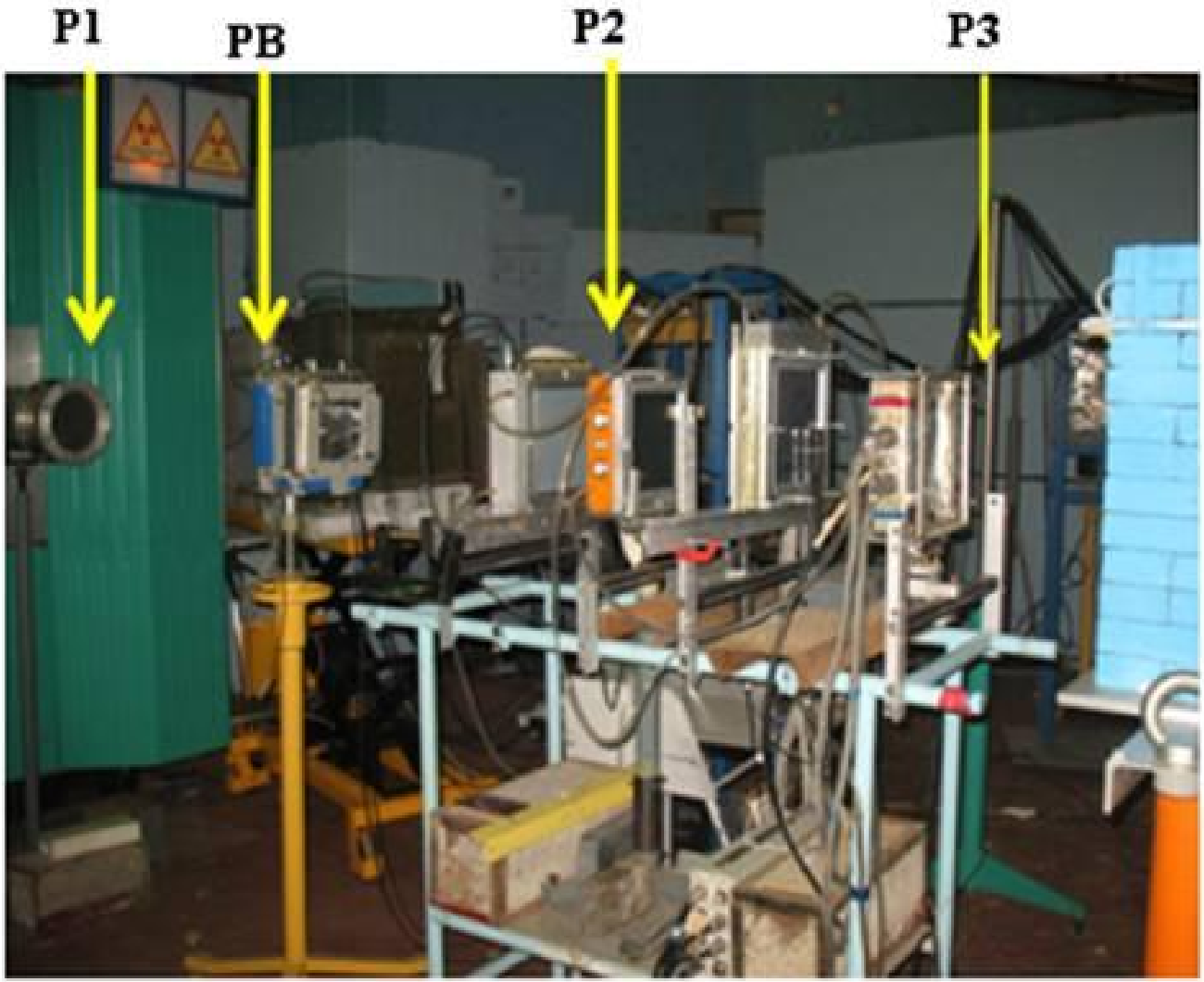
beam shift in X: 12 mm (to the right in the beam direction)

FWHM: 20 mm

beam shift in Y: - 5 mm (down)

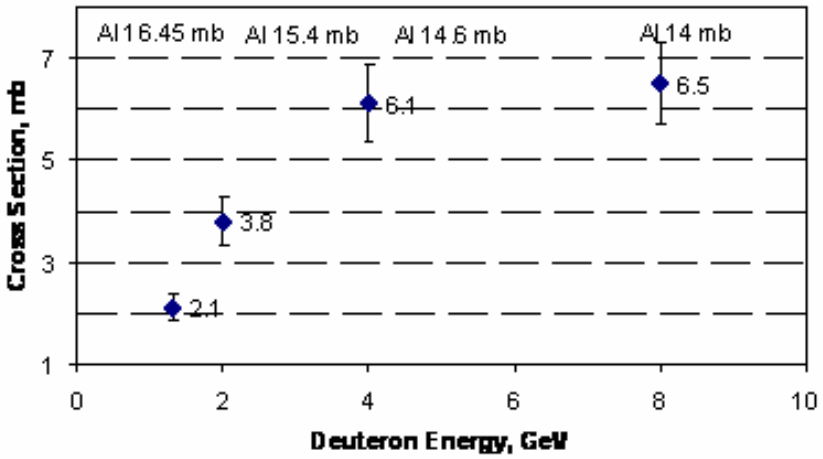
FWHM: 28 mm

GENERAL VIEW OF BEAM MONITORING SYSTEMS



UPGRADING OFF-LINE MONITORING TECHNIQUE

- Cu foils can provide more confident off-line monitoring of integral beam intensity in presence of high neutron background in comparison with previously used Al foils.
- To realize this method of beam control the special measurements were carried out in 2012.
- Its results allow one has monitoring only incident deuteron intensity due to insensitivity to low energy neutron albedo from TA.

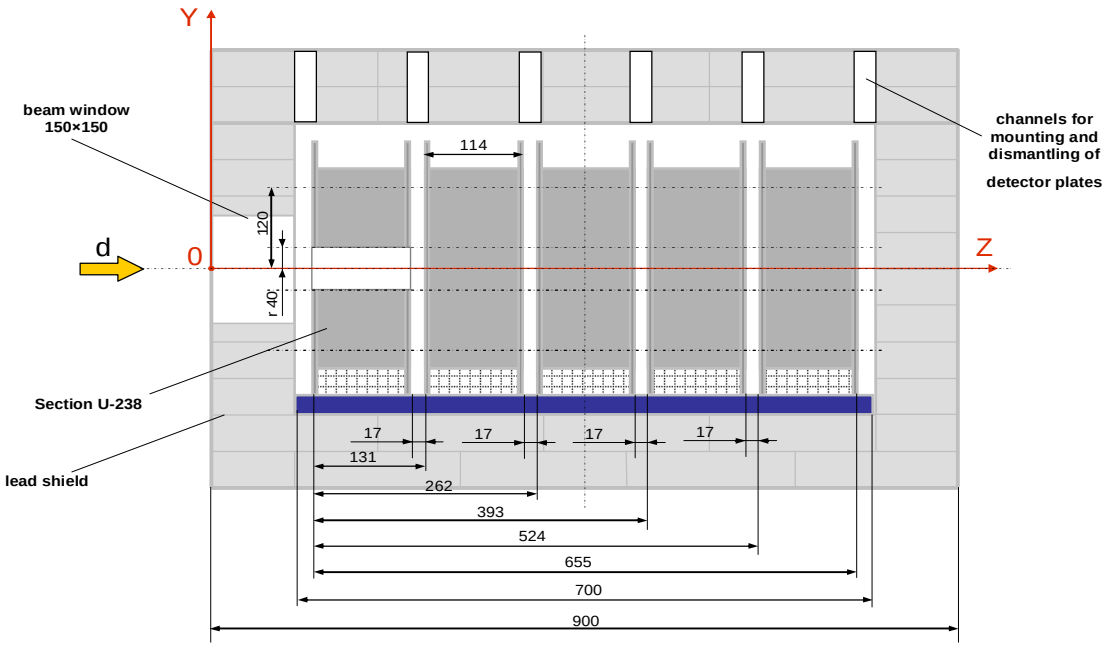


VERTICAL AXIAL CROSS-SECTIONAL VIEW OF THE TARGET

- Spatial distributions of neutron fluxes and reactions

  (n,f), (n,γ), (n,xn) studied with aid Al foils, ^{nat}U convertors (Ø 10 мм, thickness 1 мм), located on six detector plates in positions of R = 0; 4; 8 и 12 см from beam axis as well ^{nat}U and ^{209}Bi thin convertors on the surface of the lead blanket

- On some of these plates there were the samples of Th, minor actinides and long lived fission products.



Plutonium production

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

277,6 keV g-line from ^{239}Np

g- detector calibrated with ^{60}Co, ^{54}Mn, ^{57}Co, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{133}Ba, ^{137}Cs, ^{139}Ce, ^{152}Eu, ^{228}Th, ^{226}Ra standard sources.

Number of fissions in the place of measurements defines by averaging of following fission product yields:

^{97}Zr (5.42%), ^{131}I (3.64%), ^{133}I (6.39%), ^{143}Ce (4.26%)

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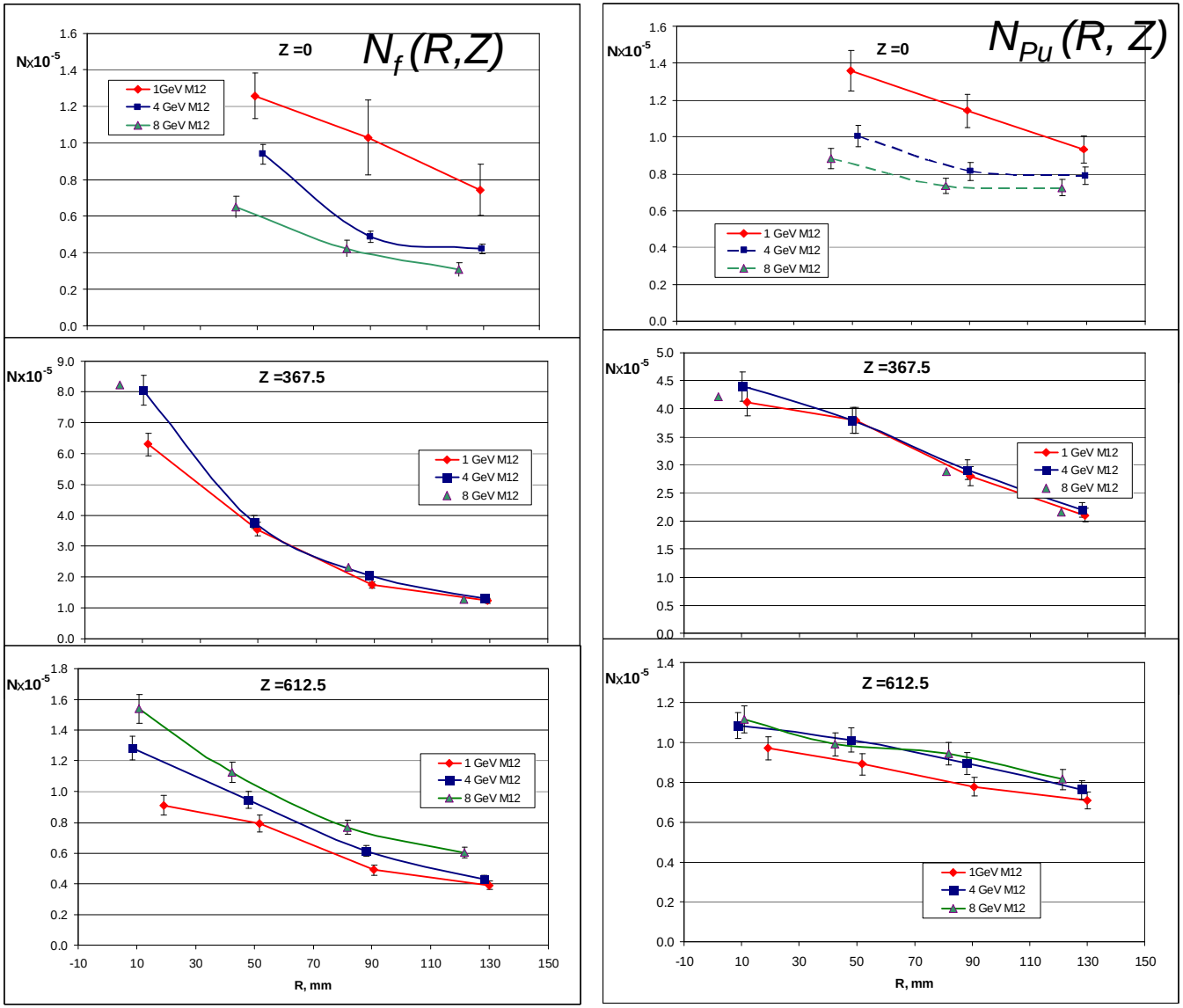
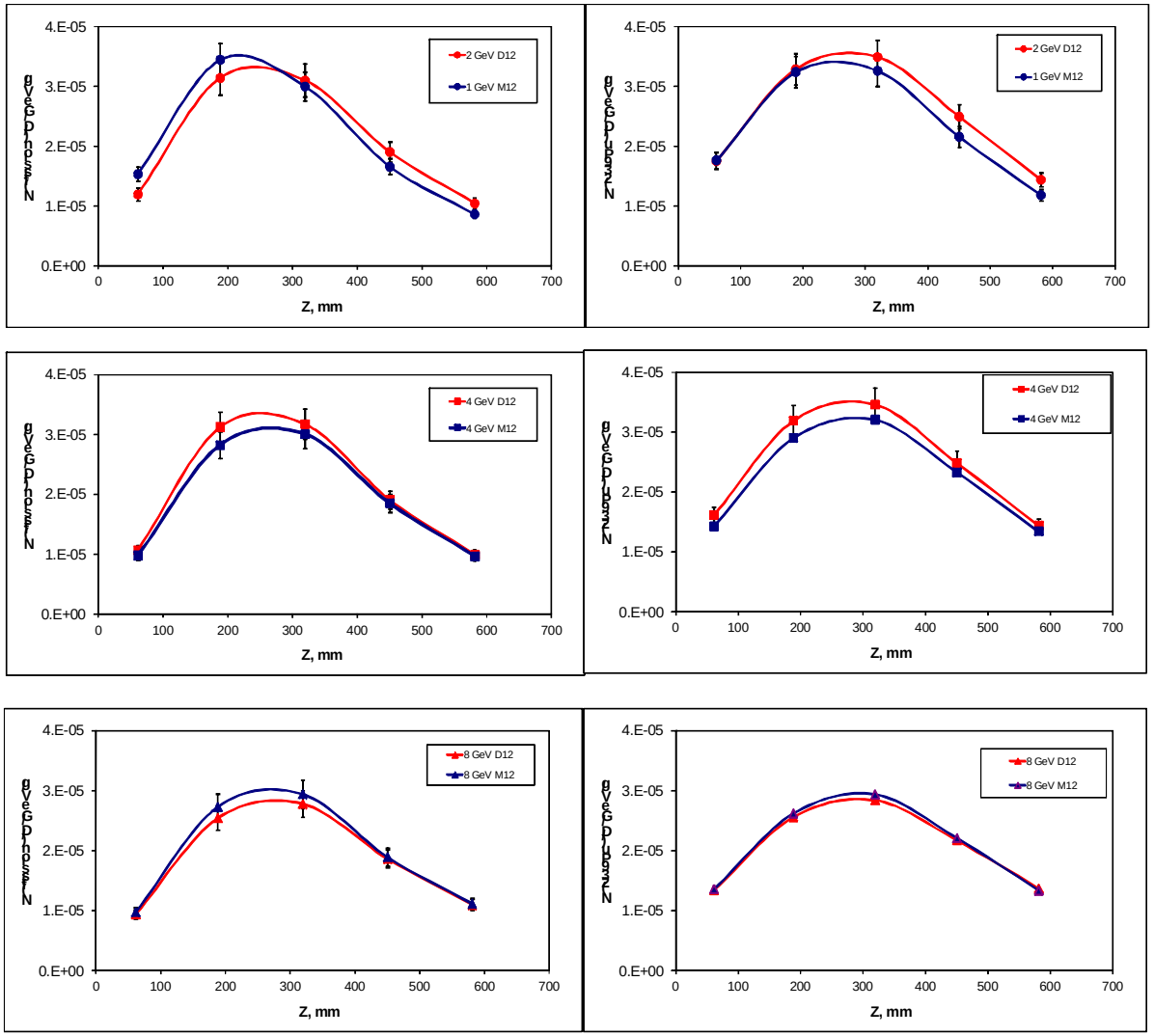
In brackets there are mean cumulative FP yields

Spatial distributions of of ^{239}Pu production and ^{nat}U fission in TA QUINTA



Distributions of $N_f(Z)$ (left) and $N_{Pu}(Z)$ (right) measured in March 2012 and December 2012

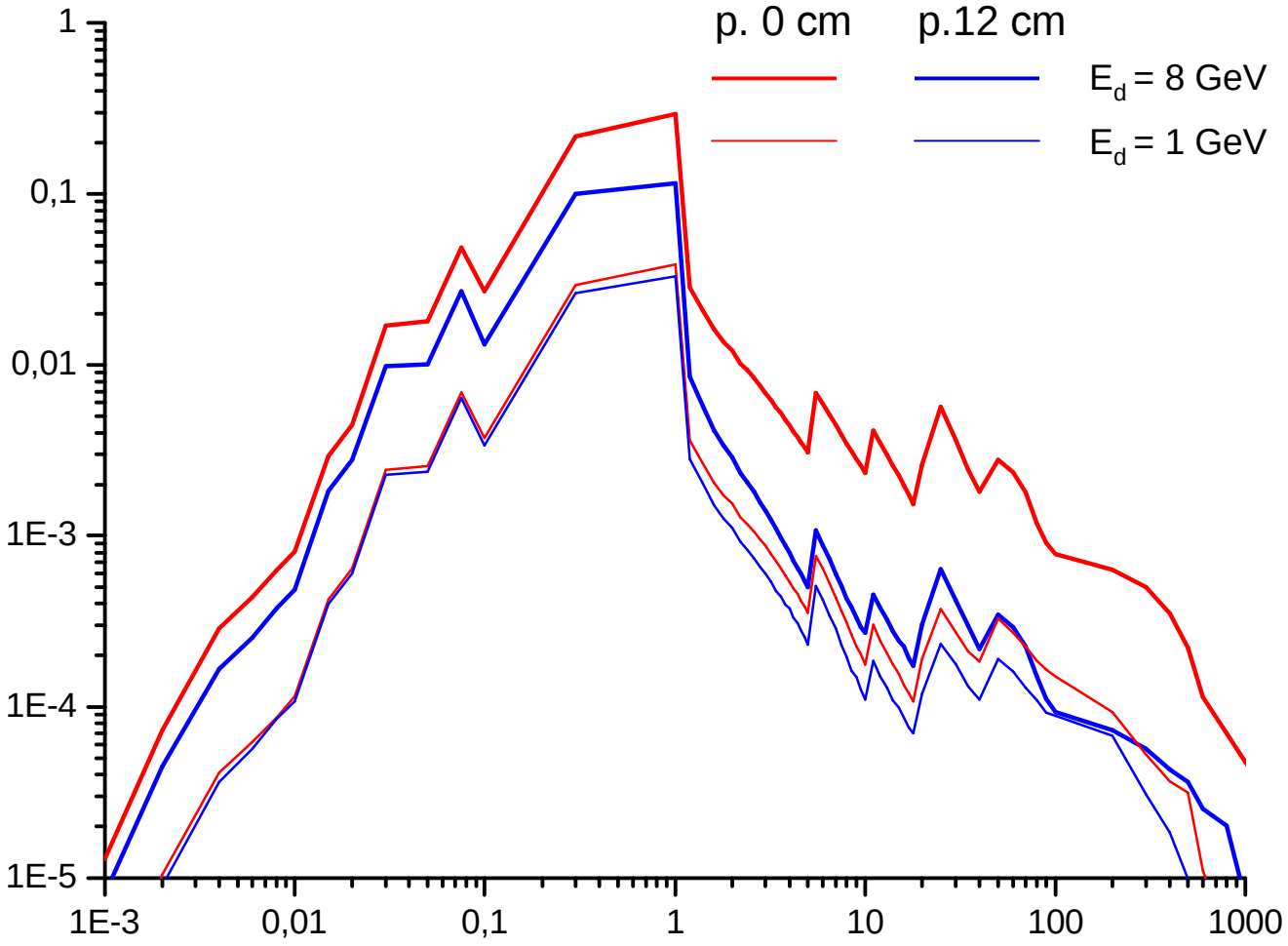


E_d	1 GeV	2 GeV	4 GeV	6 GeV	8 GeV
Date	*Total number of ^{nat}U fission in TA QUINTA $N_f(tot)$*				
03.11 No Pb		(8.8±0.4) ±1.0	(8.8± 0.4) ±1.0	(8.3±0.4) ±0.9	
12.11	(10.6±0.5) ±1.1		(8.5±0.4) ±1.0		7,3
03.12 (SSTD)	8.9 ±1.5		8.1 ±1.5		9.2 ±1.6
03.12	(10.2±0.5) ±1.1	9,1	(9.6± 0.4) ±1.0	7,7	(9.4±0.5) ±1.0
12.12	9,5	(10.5±0.5) ±1.1	(10.3±0.5) ±1.1		(9.3±0.5) ±1.0
	Total number of produced ^{239}Pu nuclei				
03.11 No Pb		(7.0±0.3) ±0.8	(7.2±0.4) ±0.8	(6.9±0.3) ±0.7	
12.11	(11.8±0.6) ±1.2		(10.8±0.5) ±1.1		9,2
03.12	(11.3±0.6) ±1.2	11,6	(11.0±0.5) ±1.1		(10.2±0.5) ±1.1
12.12	12	(12.5±0.7) ±1.3	(12.2±0.7) ±1.3		(10.3±0.5) ±1.1

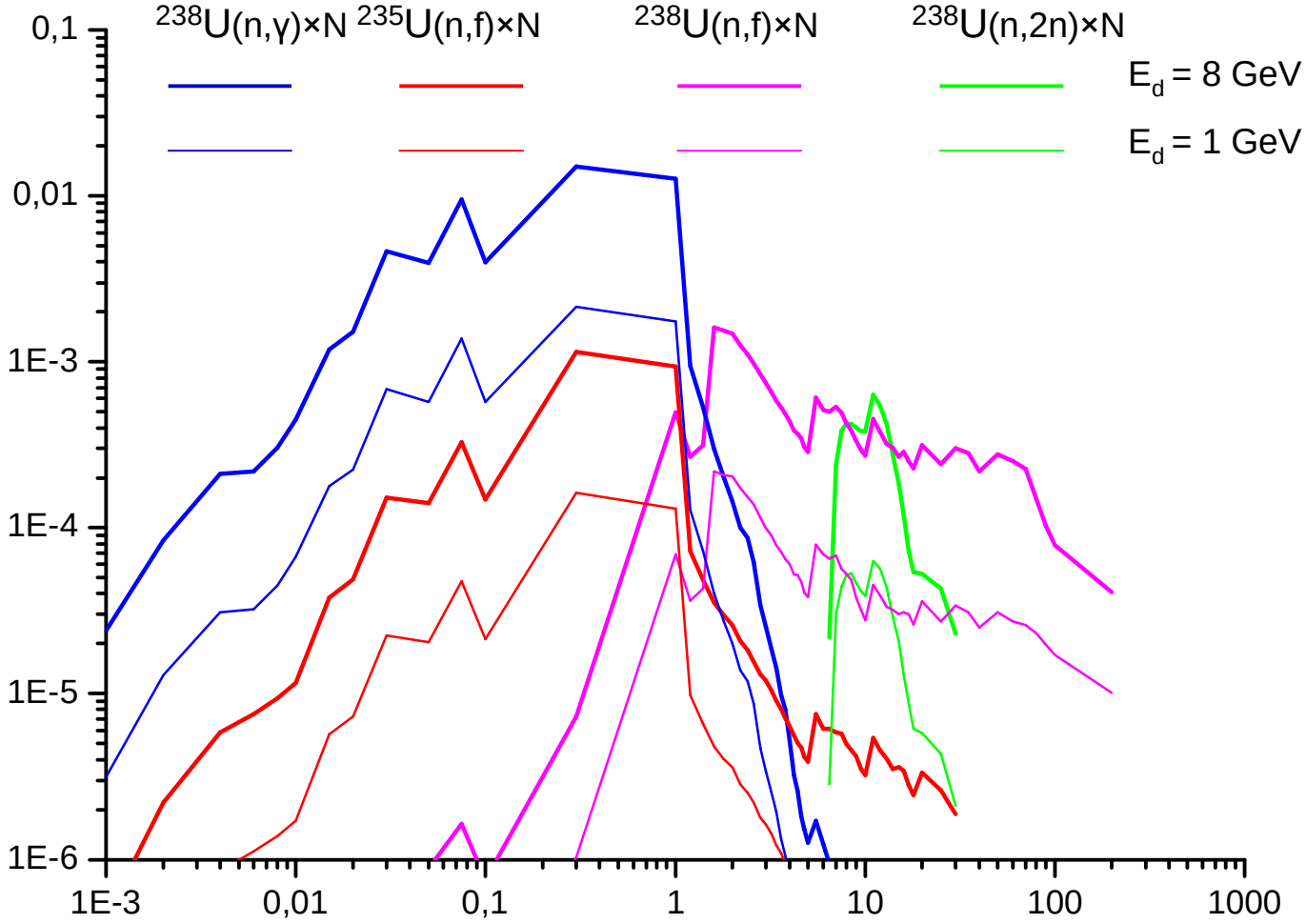
Total numbers of fission $N_f(tot)$ and produced ^{239}Pu nuclei ®

experiment v.s. calculation

Typical calculated neutron spectra at positions R= 0 and 12 cm, Z=64,5 cm (fourth plate)

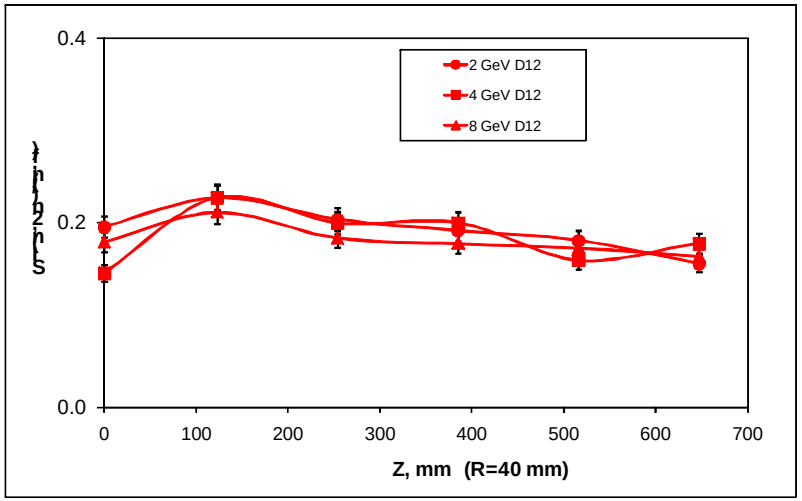


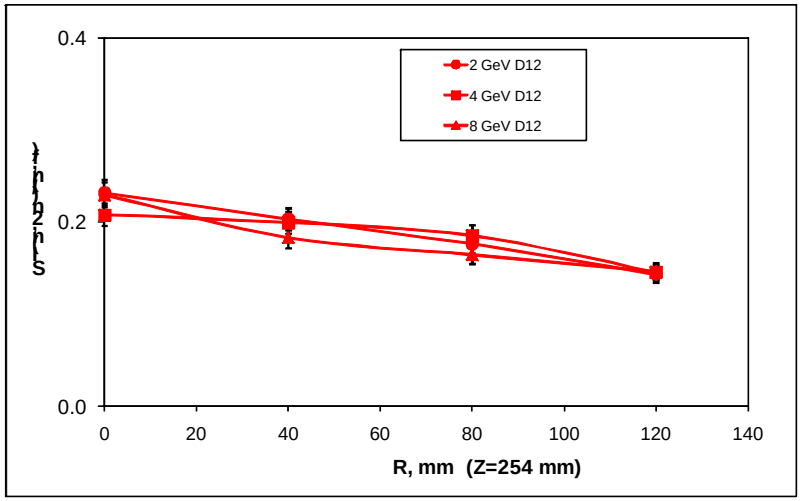
Convolution of cross-sections (n,γ), (n,f) and (n,2n) with neutron spectra at position R=12 cm in the fourth plate



Spatial distribution of Spectral Indices
$SI = N_{(n,2n)}(R,Z)/ N_f(R,Z)$

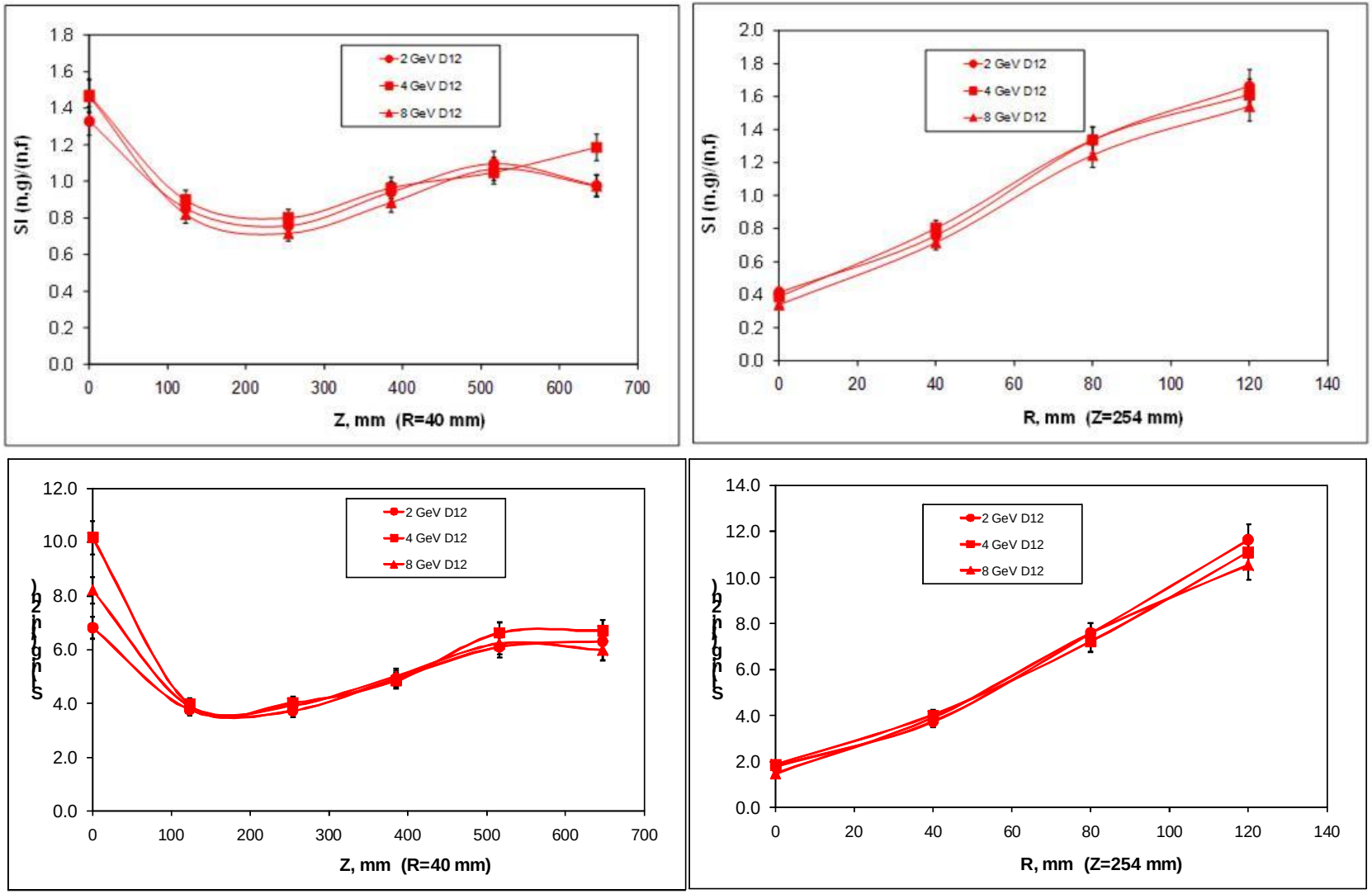
- Spectral indices are relative values. Their uncertainties are much smaller than for each $N_{(n,2n)}(R,Z)$, $N_f(R,Z)$ or $N_g(R,Z)$ values. ***SI*** do not depend on beam normalization
- The threshold of (n,f)-reaction ~ 1.2 MeV and for (n,2n) ~ 7 MeV
- Constancy of *SI* depending on *Z, R* and Ed indicates that the ratio of parts of the neutron spectrum responsible for these reactions varies slightly over volume of uranium target for studied interval of deuteron energy.



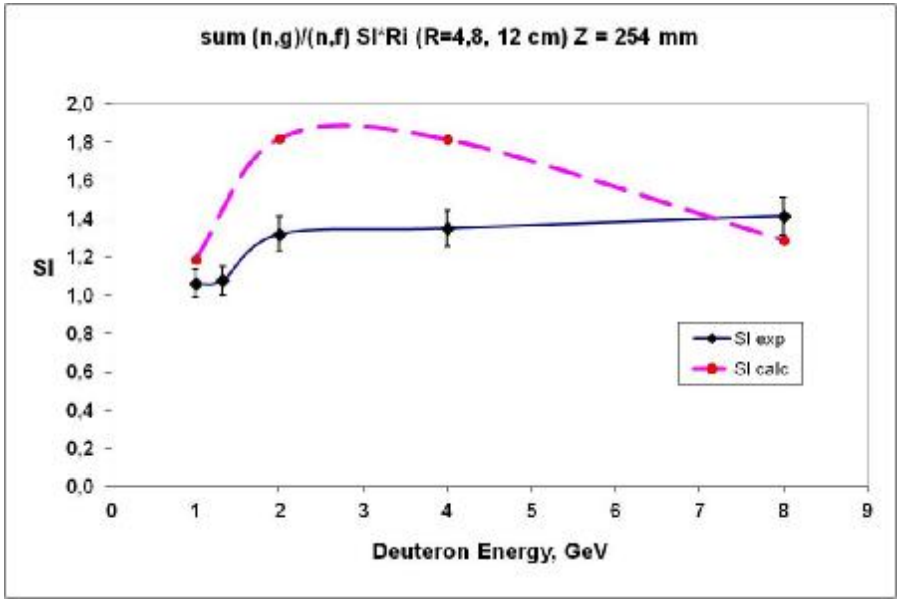


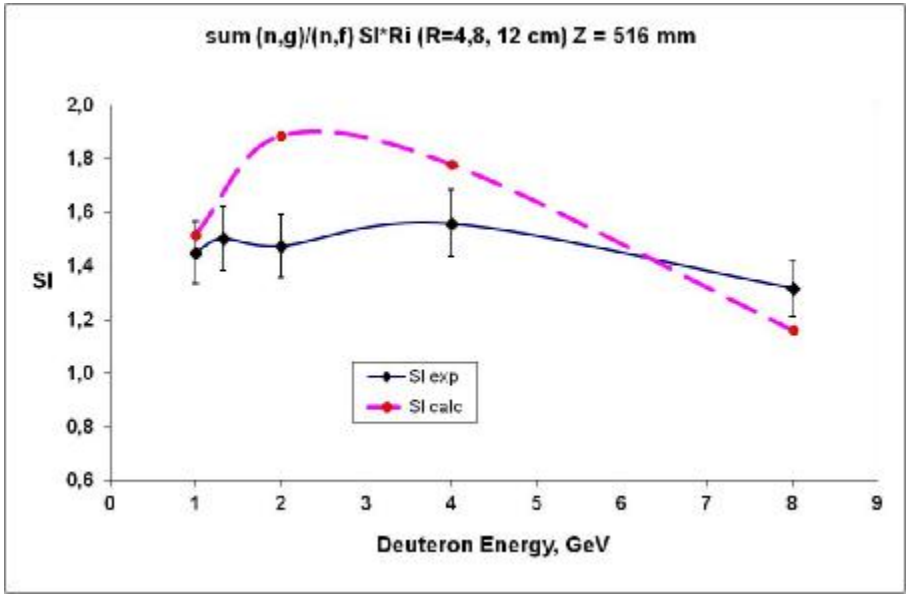
Spectral indices SI (n,γ)/(n,f) and SI (n,γ)/(n,2n)

Neutron spectrum becomes “softer” with increase the distance from beam axis



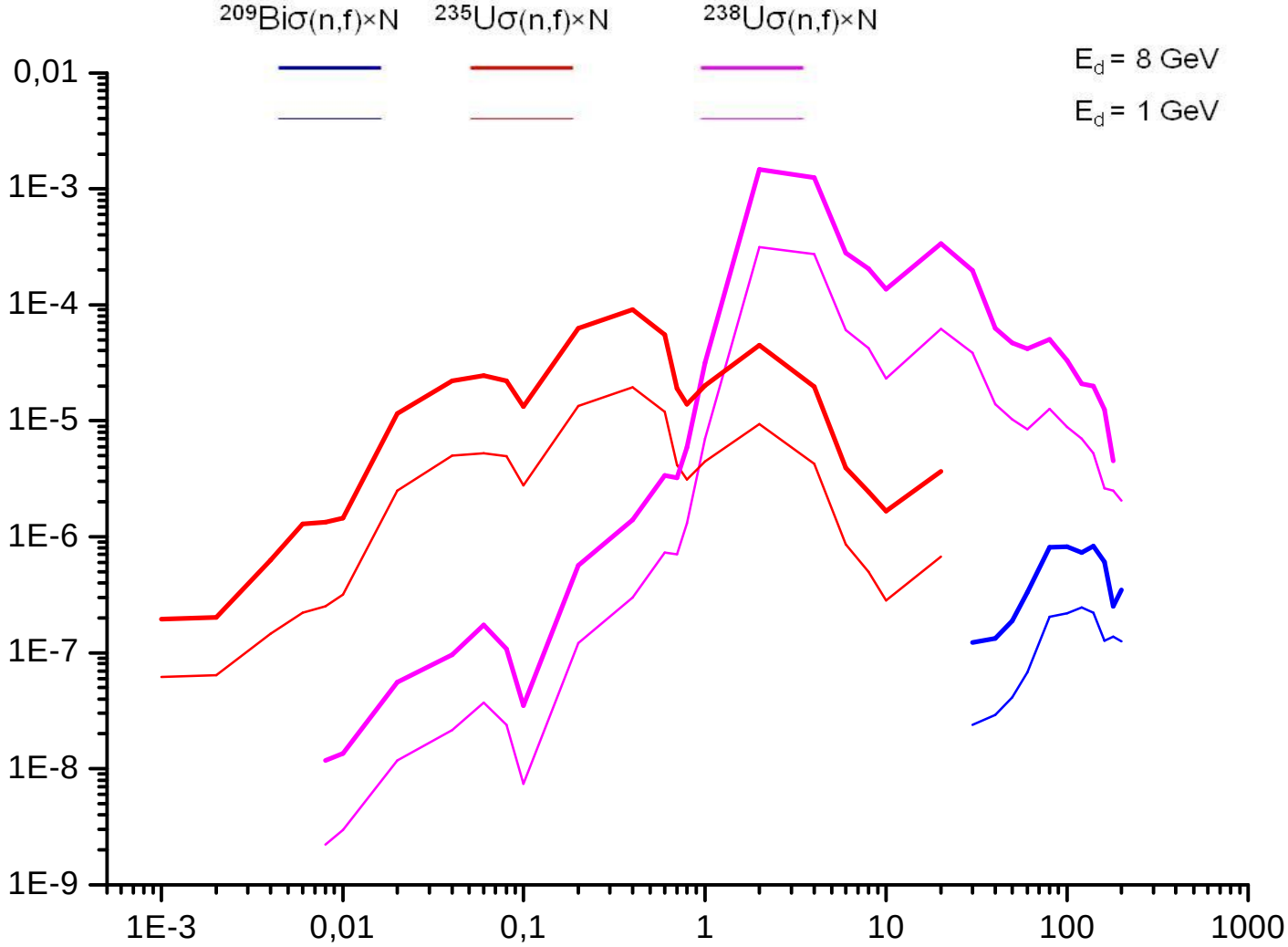
Comparison of experimental and calculated spectral indices SI (n,γ)/(n,f) in dependence on deuteron energy



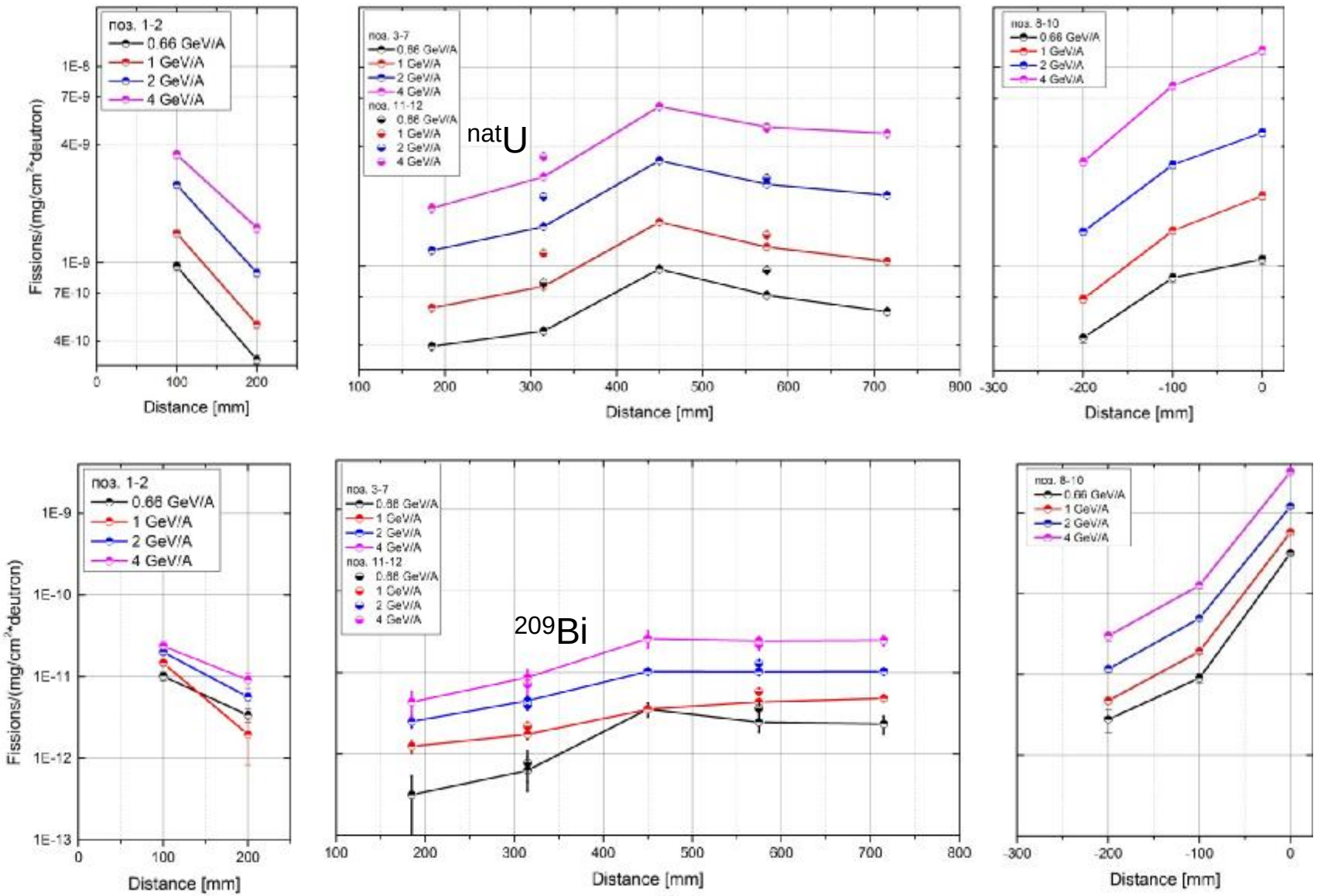


- **Share ratio of the neutron spectrum in the energy ranges En < 1.2 MeV and 1.5 Mev < En< 20MeV remains approximately constant with increase of incident energy**
- **But what about En > 20 MeV ?**

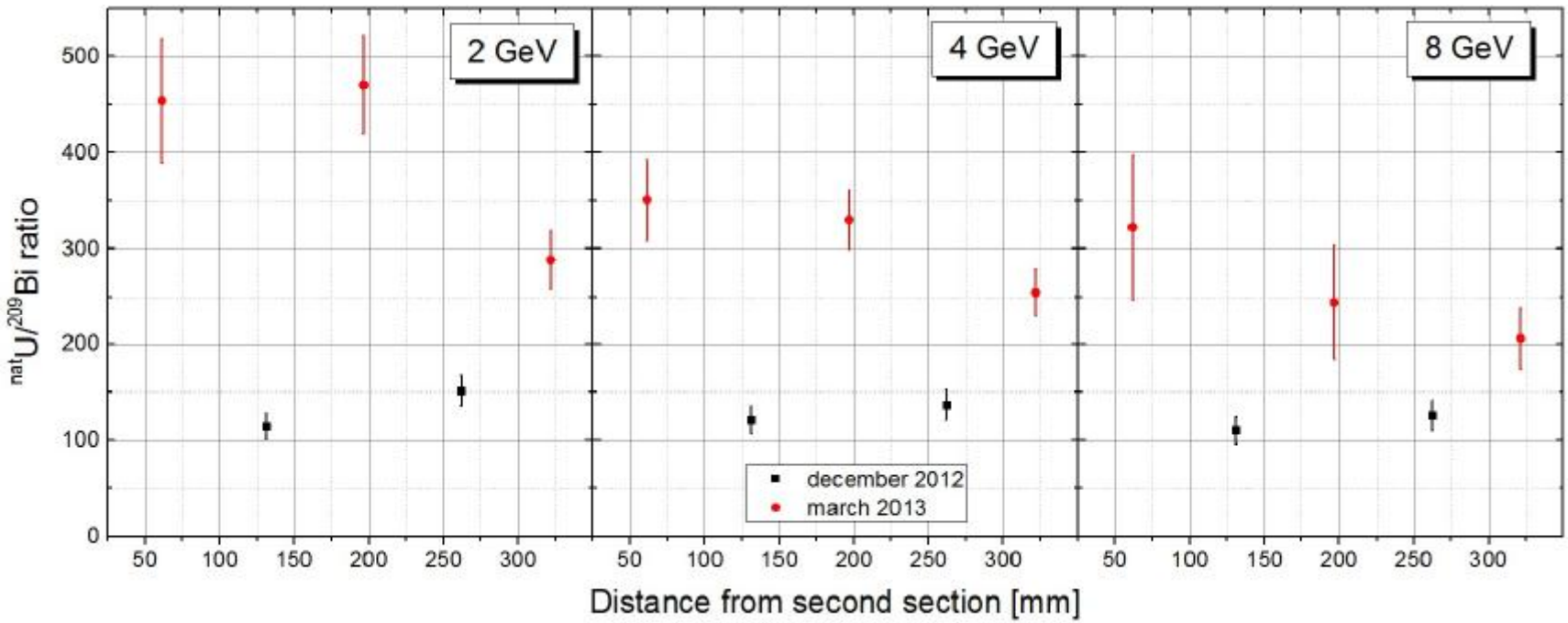
Convolution of neutron spectra with σ(n,f) for $^{nat.}U$ and ^{209}Bi at position R=12 cm in the fourth plate (Z=64,5 cm)



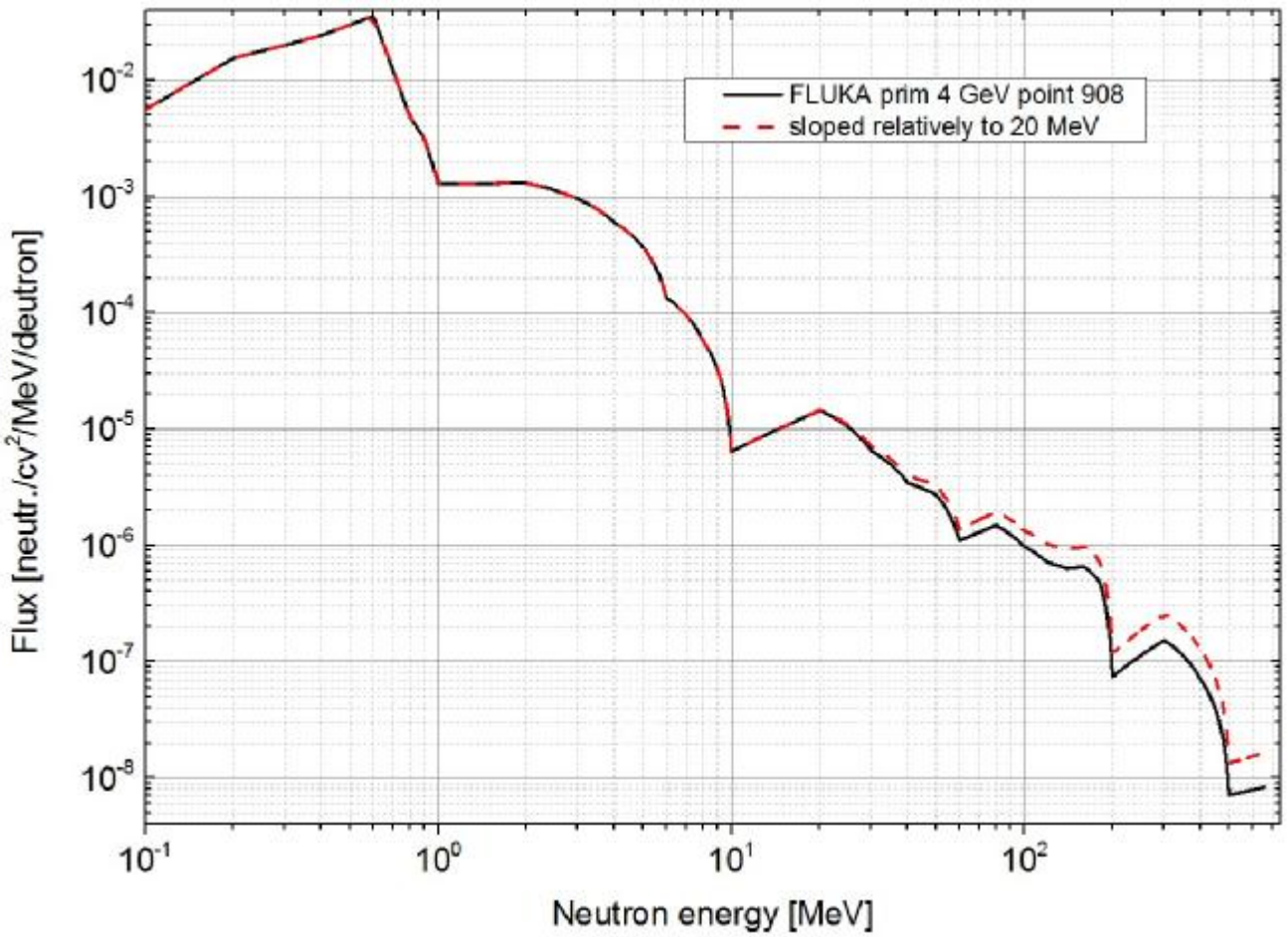
Fission rates for ^{nat}U and ^{209}Bi measured on surface TA



Ratios of $^{nat}U/^{209}Bi$ (n,f)-reaction rates
*(**black** – inside of TA QUINTA, **red** – on the surface)*



Neutron spectrum modified to fit experimental ratio $^{nat}U/^{209}Bi$ (n,f)-reaction rates



Spectral characteristics of leakage neutrons on QUINTA surface

E_d, GeV	1,32	2,0	4,0	8,0
Numbers of leaked neutrons N, n/(d·GeV)				
$N_{En>0,1\ MeV}$ (exp),	46,8	49,1	51,0	51,8
$N_{En>20\ MeV}$ (exp))	2,97	3,16	3,76	6,23
N_{total} (calc. MARS, FNAL))	49,2	47,9	42,8	39,1
$N_{En>20\ MeV}$ (calc. MARS, FNAL))	0,68	0,70	0,50	0,62
Ratios $N_{En>20\ MeV}$ / N_{total} , %				
$N_{En>20\ MeV}$ (calc) / N_{total} (calc)	1,38	1,46	1,17	1,60
$N_{En>20\ MeV}$ (exp) / $N_{En>0,1\ MeV}$ (exp)	6,35	6,43	7,38	12,0

E_d, **ГэВ**	1,0	2,0	4,0	8,0
Total neutron multiplicity, n/(d·GeV)				
M(exp)	59±11	62±12	63±12	62±12
M(calc. , MCNPX)	76,9	74,9	67,0	61,1
M(calc., MARS)	77,5	74,7	67,5	60,5
M(calc., MCNP [8])	60.7	62,7	57,3	-

Neutron multiplicities

[8]

Deuteron energy Ed, GeV	1	2	4	1	2	4
Models	Without Pb blanket			With Pb blanket		
Bertini-ABLA	56.4	115.7	209.6	60.7	125.5	229.6
Bertini-Dresner	53.4	108.9	197.3	57.4	118.2	216.5
CEM03	58.5	118.3	213.1	60.4	123.6	224.9
INCL-ABLA	54.0	112.6	203.3	57.8	122.5	225.2
INCL-Dresner	50.7	104.1	186.6	54.6	113.5	207.6
ISABEL-ABLA	56.8	113.6	201.9	61.0	123.7	222.9
ISABEL-Dresner	52.8	105.8	189.3	57.0	115.5	209.0

Neutron balance in TA QUINTA- experiment v.s calculation

Energy Ed,GeV	1,32	2	8	1,32	2	8
Nuclear reactions	Neutron production			Neutron “utilization”		
Fast cascade reactions	51,6	74,6	230,4	-	-	-
(n,xn) calculation	13,7	21,0	76,6	5,2	8,1	29,9
(n,2n) EXP				1.7±0.2	3.4±0.4	11±1.3
(n,f) calculation	35,6	53,1	178,5	11,2	16,5	54,0
(n,f) EXP				13±1.7	21±2.6	74±10
(n,γ) calculation	-	-	-	15,8	23,2	73,4
(n,γ) EXP				15±1.8	25±2.7	82±9
Other reactions	-	-	-	3,8	5,4	15,8
Neutron leakage calc.	-	-	-	64,9	95,6	312,4
Neutron leakage EXP				62±12	98±20	414±80
Multiplicity calc.	100,9	148,6	485,5	100,9	148,6	485,5
Multiplicity EXP.				77±13	123±21	496±83

Neutron balance in TA QUINTA- experiment v.s. calculation

Energy Ed,GeV	1,32	2	8	1,32	2	8
Nuclear reactions	Neutron production			Neutron “utilization”		
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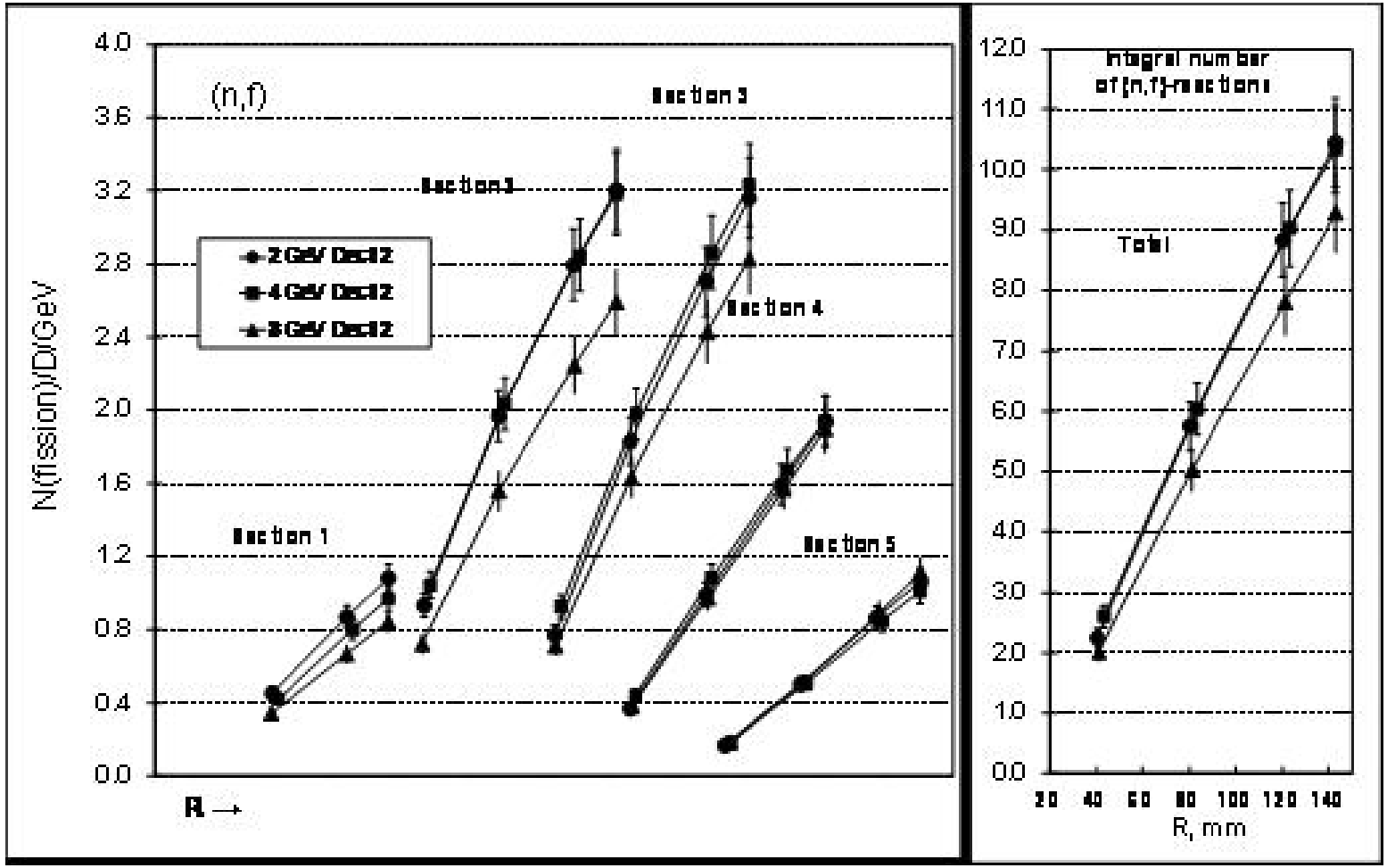
Neutron leakage EXP				62±12	98±20	414±80
Multiplicity calc.	100,9	148,6	485,5	100,9	148,6	485,5
Multiplicity EXP.				77±13	123±21	496±83
Leakage EXP %				80±19	80±19	83±20

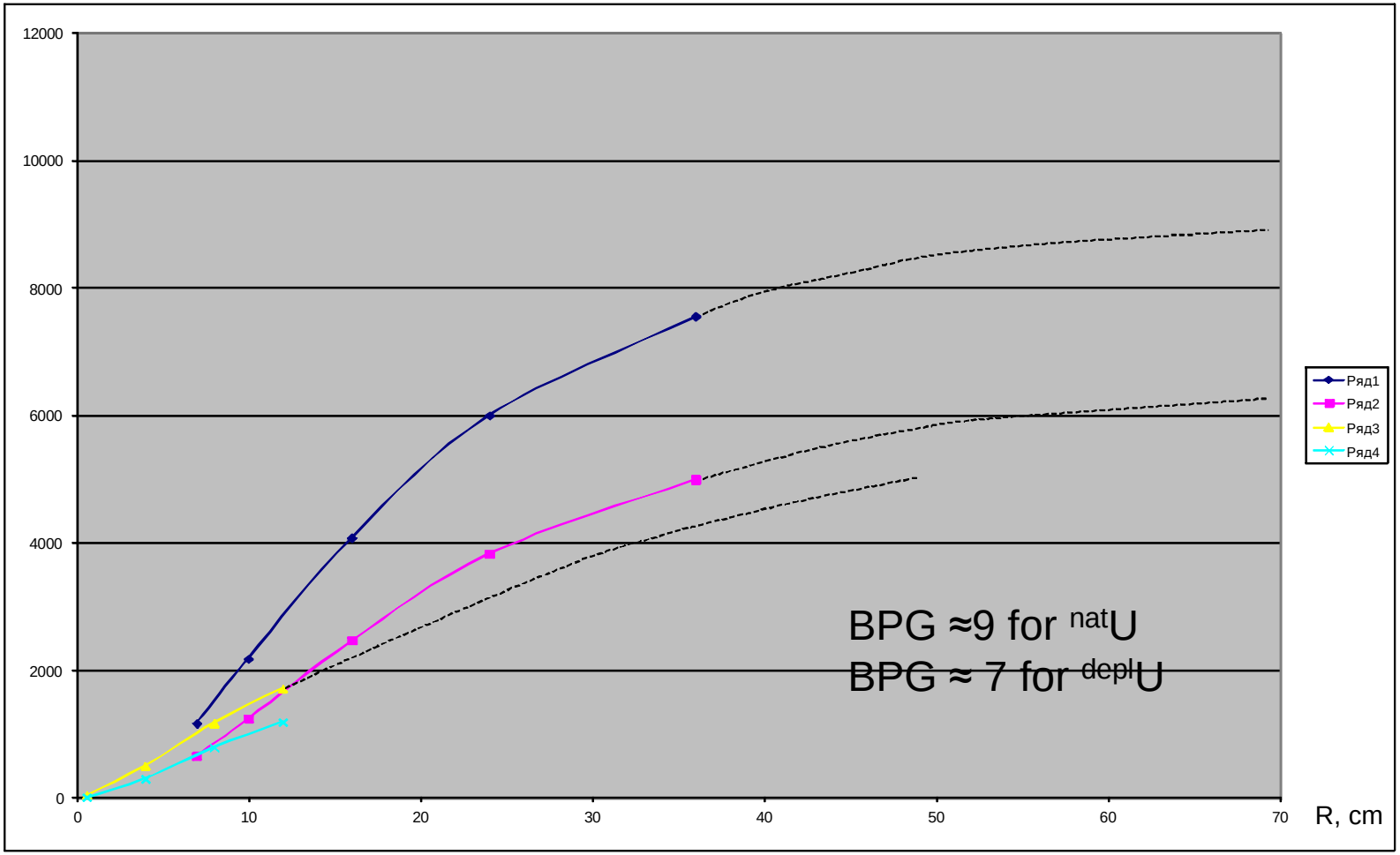
Beam power gain v.s. size of AC

- Beam power gain ***(BPG = $E_{release}/E_{beam}$)*** is key characteristics of any ADS. It defines practical applicability of the system.
- Minimal value of BPG providing a zero energy balance is BPG ≈ 9
- TA QUINTA shows BPG ≈ 2. This is due its relatively small size of (R ≈ 15 cm) and as consequence the large neutron leakage ≈ 80%.
- But BPG as function of radius (size) AC does not show saturation not only in TA QUINTA but even in ~ 3 tons ^{nat}U TA studied in experiment by Vassil'kov, Gol'dansky , Pimenov, Pokotilovsky and Chistyakov (1978).

Integrated numbers of fission up to given R (for each sections and total over whole TA)



Integrated numbers of fission up to given R (a.u., blue - Z = 245 mm, Z = 655 mm – lilac [VGPPC], yellow – Z= 52 mm and light blue – Z= 65 mm QUINTA)



Beam power gain v.s. incident energy

- In experiment of VGPPC BPG ≈ 7.4 for ^{nat}U and ≈ 6 for ^{depl}U (uranium mass – 3.2 tons, neutron leakage ~ 10%).
- But this result was obtained at E_p = 0.66 GeV and it was not reproduced by any calculations.
- For example calculation by Batyaev et al. (2008) for 30 t of ^{depl}U target gives BPG ≈ 3 for incident proton energy range from 1 to 10 GeV.
- Similar results were obtained in our calculations by MCNP and MARS codes for QI TA BURAN - BPG ≈ 3.8

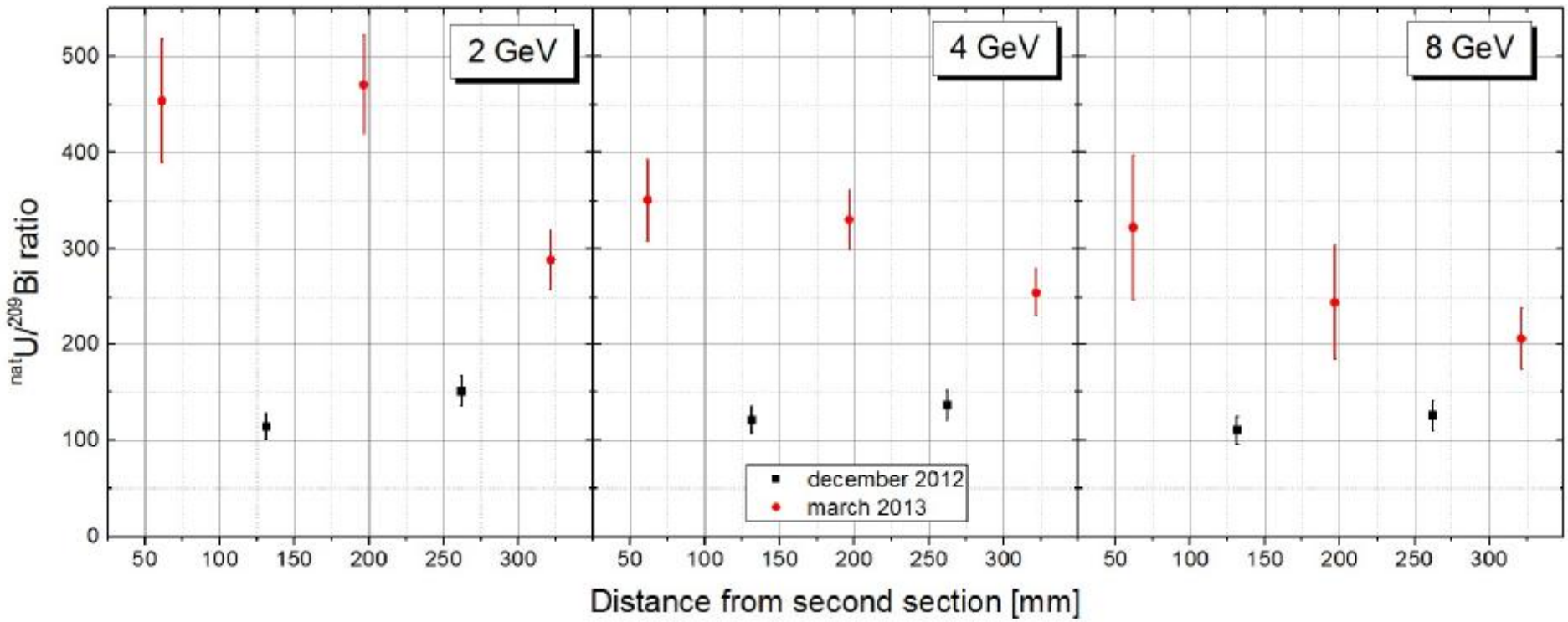
Modeling of TA BURAN

	Protons			Deuterons		
$E_{p(d)}$, GeV	1	6	12	1	6	12
Total neutron multiplicity	126	770	1450	125	794	1455
Number N (n,γ)	70	440	826	70	452	837
Number N(n,f)	16	100	183	15	100	183
$K_{BPG} = E_{tot}/E_{p(d)}$	*3.82*	*3.75*	*3.5*	*3.82*	*3.85*	*3.55*

Beam power gain v.s. incident energy

- Small calculated values of BPG (total number of fission in AC) may be associated with a significant underestimation in all used codes of the proportion of high-energy neutron spectrum share discussed above.
- Moreover as show our measurements of ratios $^{209}Bi/^{nat}U$ fission rates with TA QUINTA neutron spectrum becomes more “hard” with increase of incident energy.

Ratios of $^{nat}U/^{209}Bi$ (n,f)-reaction rates
*(**black** – inside of TA QUINTA, **red** – on the surface)*



Beam power gain v.s. incident energy

- Such tendency in neutron spectra provides a chance to get an additional increase of BPG for higher incident energy.
- But only experiments with QI TA BURAN are able to clear out of real dependence of BPG on incident energy and to establish its absolute value.
- This is the subject of “E&T RAW” collaboration work for 2014-2016

Instead of conclusion

REACTION RATE R(exp.)[E-27](err.)

Reaction products	Ed = 2 GeV Reza et al.	Ed = 4 GeV Adam et al.	Ed = 6 GeV Adam et al.
Th(n,g)**Pa-233**	76.9(39)	142(4)	176(3)
Th(n,2n)**Th-231**		51.4(15)	71.2(23)
Th(n,6n)**Th-227**		3.8(15)	4.4(4)
Th(n,p6n)**Ac-226**	1.13(8)	2.98(21)	3.41(11)
Th(n,p8n)**Ac-224**	0.62(5)	1.37(6)	3.1(3)
Th(n,**fission**)	**54.4(40)**	**118(10)**	**159(7)**
Th(n,…)**Zr-97**	1.9(9)	3.77(14)	5.2(17)
Th(n,…)**Mo-99**	1.99(11)	5.14(6)	5.75(18)
Th(n,…)**I-131**	1.15(14)	1.92(8)	2.26(6)
Th(n,…)**I-133**	1.04(6)	2.44(5)	3.12(5)
Th(n,…)**Ce-143**	1.06(8)	2.61(5)	3.25(6)
Th(n,**spallation**)	**17.9(25)**	**-**	**194(30)**

Main aim of experiments with TA BURAN for 2014-2016

- Investigation of BPG dependence on energy of incident particles (protons or deuterons) to determine its optimal value for this type of particle.
- Determination of reaction rates of long-lived isotopes of processing of spent nuclear fuel.
- Getting a complete set of experimental data required for verification and modification of existing theoretical models and transport codes that can reliably describe and predict the properties of accelerator driven systems with QI active cores.

Thanks for your attention