Study of neutron fields and nuclear reactions in massive natural uranium target irradiated by deuteron Nuclotron beams with energy (0.5 - 4) GeV/nucleon and perspectives for ADS with deep subcritical active core

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Introduction

- The problems of rapidly growing energy consumption in the world can not be solved without the use of nuclear energy.

- The key issue here is the availability of an adequate supply of nuclear fuel. In the long term aspect, the use of such materials as enriched ^{235}U or artificial ^{239}Pu can not solve the problem of global energy.

- Indeed, obtaining of ^{235}U or ^{239}Pu is very energy costly, and its total amount is rather limited and certainly does not exceed the forecast amount of hydrocarbon fuel.

- So only involvement in the production of energy is practically unlimited reserves of natural (depleted) uranium and thorium can provide long-term prospects for nuclear energy.

- But inevitable and dangerous in the long term consequence of nuclear power – radioactive wastes- requires radical solution
- As an alternative of underground waste disposal recently seriously considered the transmutation of its long-lived components with the Accelerator Driven subcritical Systems (ADS) (example – MYRRHA project)
- Such systems in principle could transmute ("burn") wastes and simultaneously produce energy

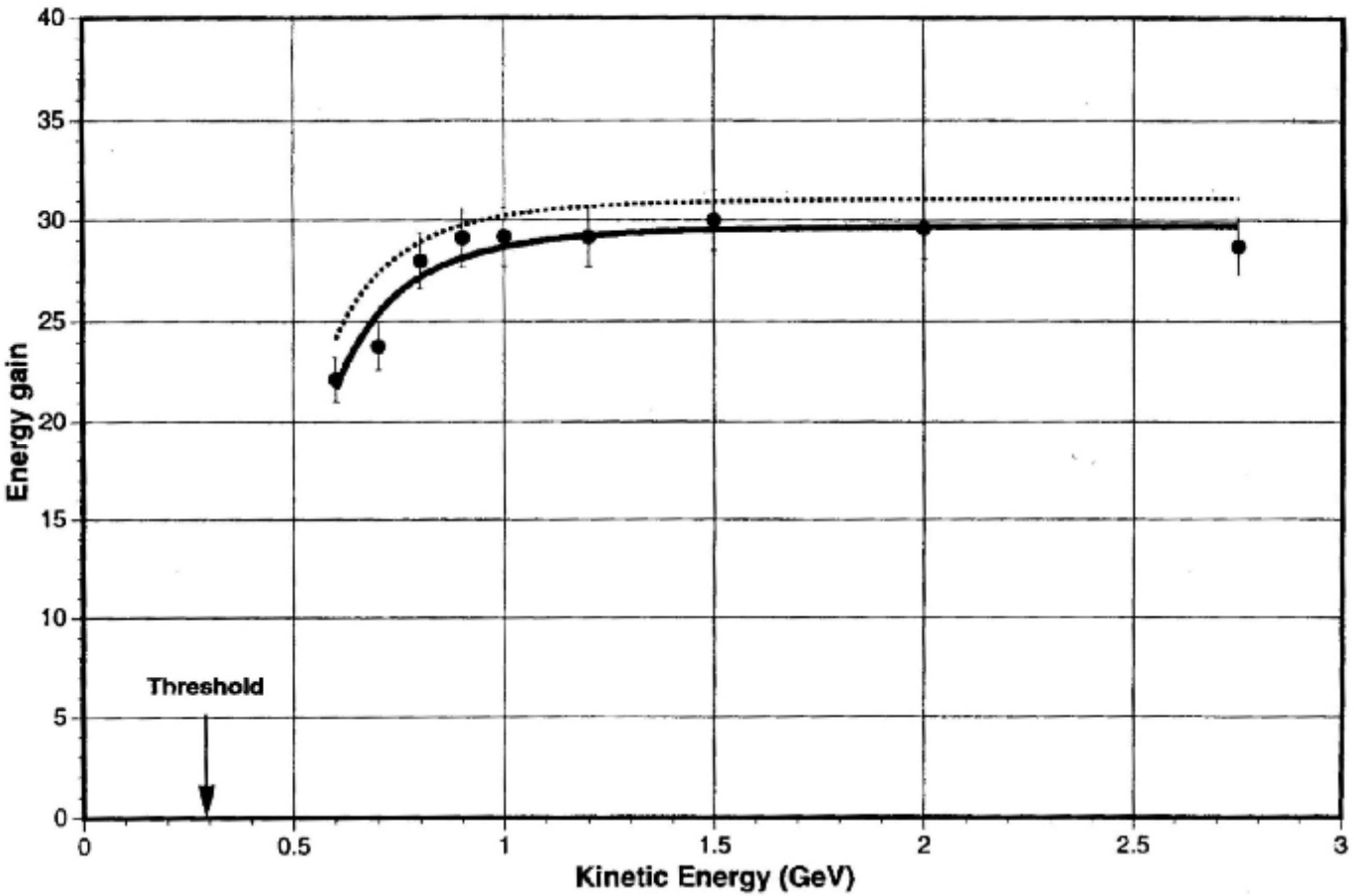
Problems of natural uranium and thorium use

- Carlo Rubia proposed the idea of so-called energy amplifier based on ADS with multiplying target (active core) from $^{nat.}$U or Th .

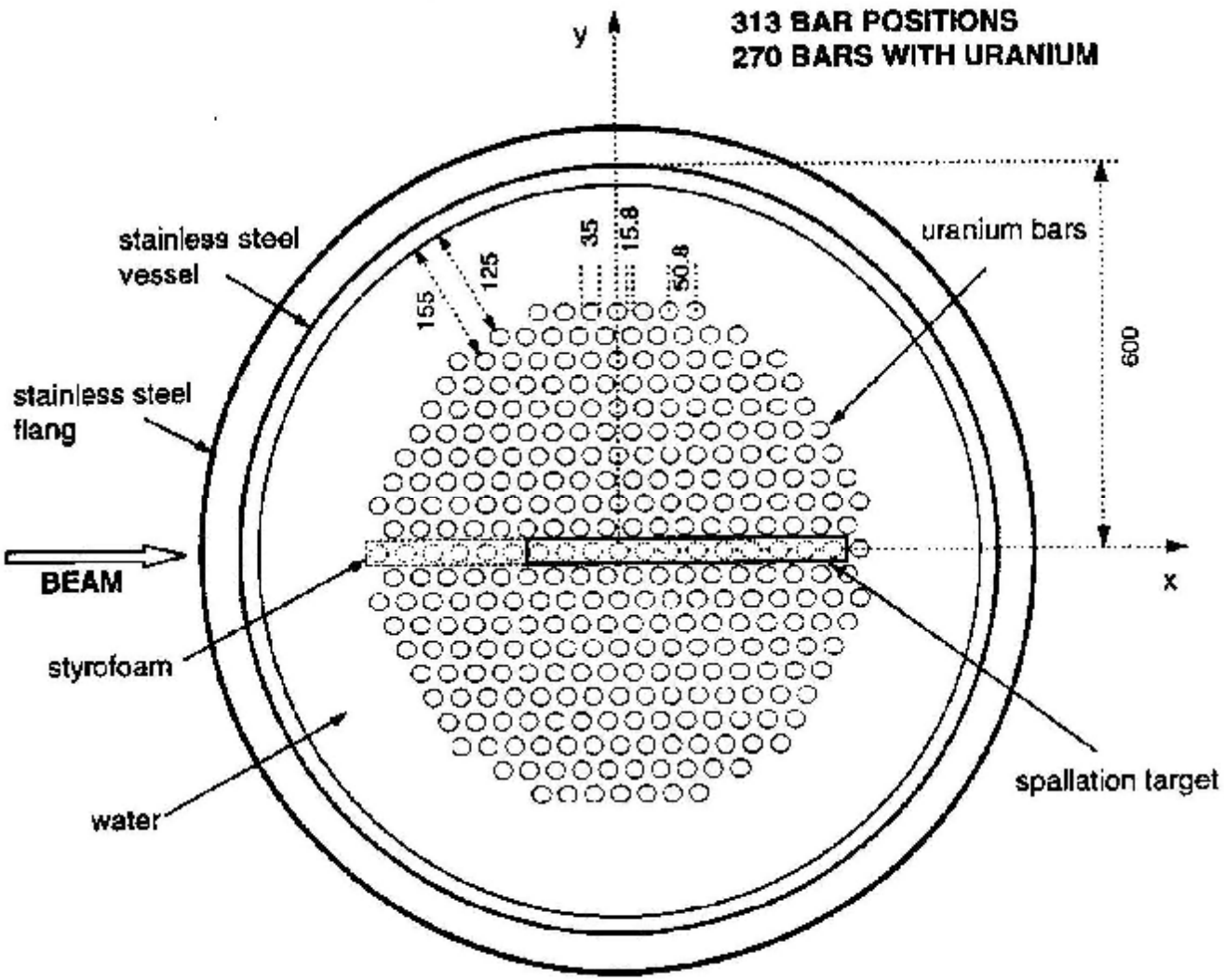
- The main positive finding of a key experiment FEAT performed at CERN by C. Rubbia group, that it is possible to reach the gain power of the incident proton beam (BPG)* around 30 at an energy of 1 GeV. With increasing proton energy up to 2.7 GeV this value goes to constant.

* *The ratio of energy released in the subcritical target to the energy of the incident proton beam is the Beam Power Gain (BPG)*

Main result of the FEAT experiment
(S. Andriamonje et al.,CERN/AT/94-45(ET))



Top view of the FEAT subcritical assembly. Sizes in mm.



Problems of natural uranium and thorium use

- But in this experiment the massive uranium target (~3.5 tones) was embedded into light water moderator. As consequence the neutron spectrum inside of active core was practically fully thermalized and neutron multiplication coefficient k_{eff} of this system was near 0.9.

- In these circumstances in spite of rather promising BPG ≈ 30 it is practically impossible to use the base core material (natural uranium or thorium) for energy production because of their high fission thresholds (~1.2 MeV and ~5 MeV respectively).

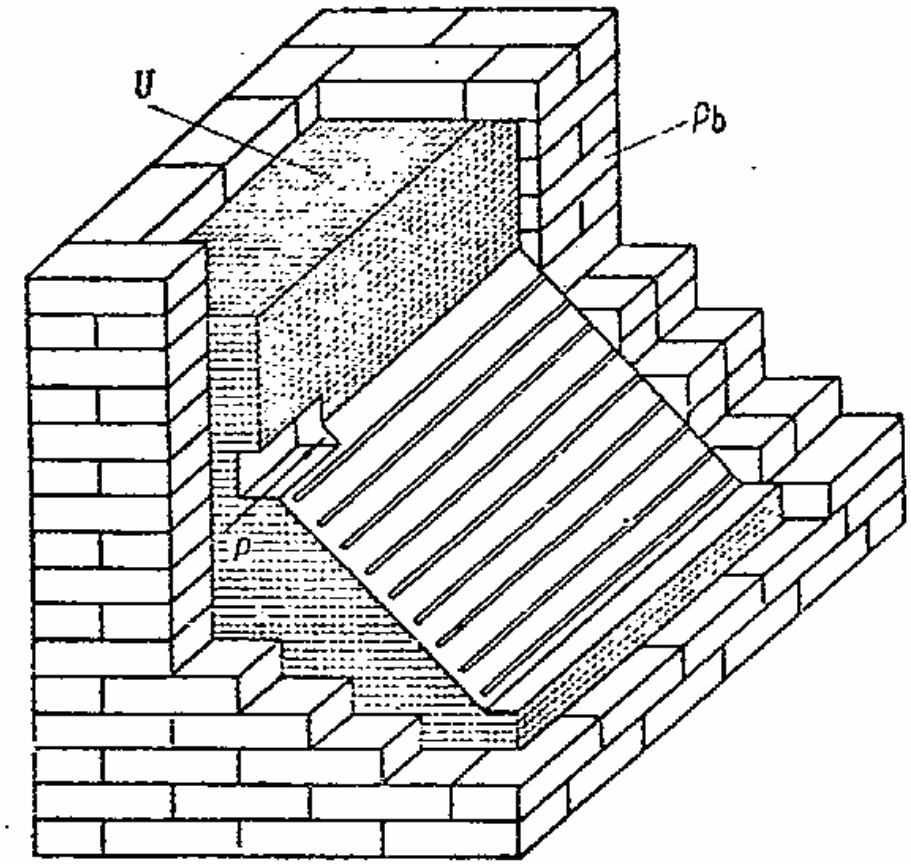
- 3500 kg 238U → 25 kg 235U → BPG !!!

- And actually proposed EA options must move on to the enriched fuel !?

New is a good forgotten old …

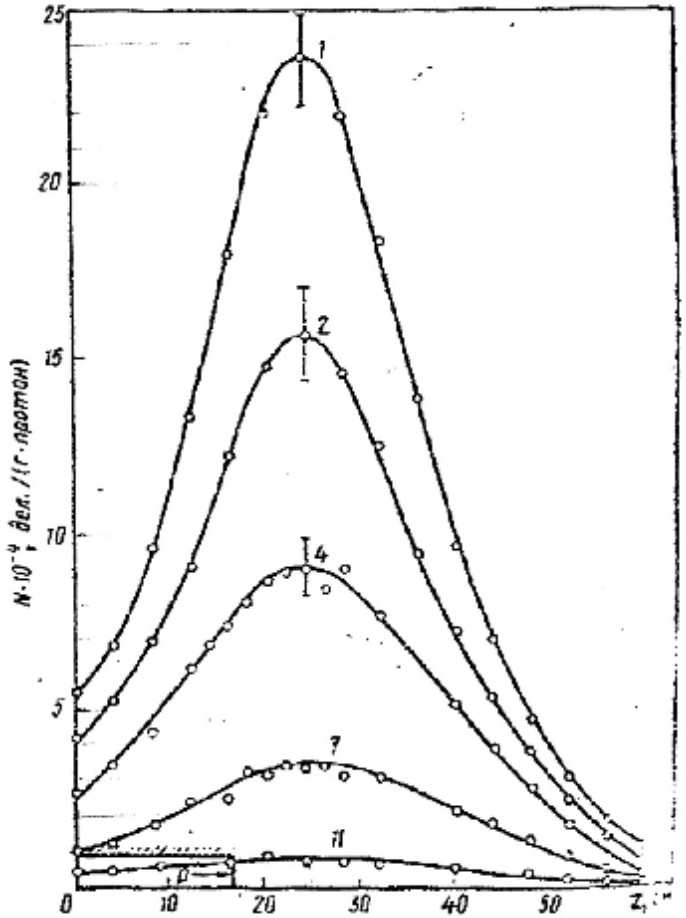
- About 50 years ago at Dubna by Vasil'kov-Goldansky group there was obtained BPG ≈ 7.4 with only 0.66 GeV protons and 3.2 tones of natural uranium metallic target.

Target assembly of experiment by R. Vassil'kov et al (1978)



Dimensions
56x56x64cm

- *No moderator*
- *Special geometry – assymmetric beam input → 7 tons effective mass*
- *10 cm lead blanket*
- *Rather small (~10%) neutron leakage*



N_f axial distributions for different radial channels

Natural U

N_f = 18.5 ± 1.7

N_{Pu}= 46 ± 4

BPG = 7.4 ± 0.7

Depleted U

Nf = 13.7 ± 1.2

N_{Pu}= 38 ± 1.2

BPG = 5.7 ± 0.5

Ep = 0.66 GeV

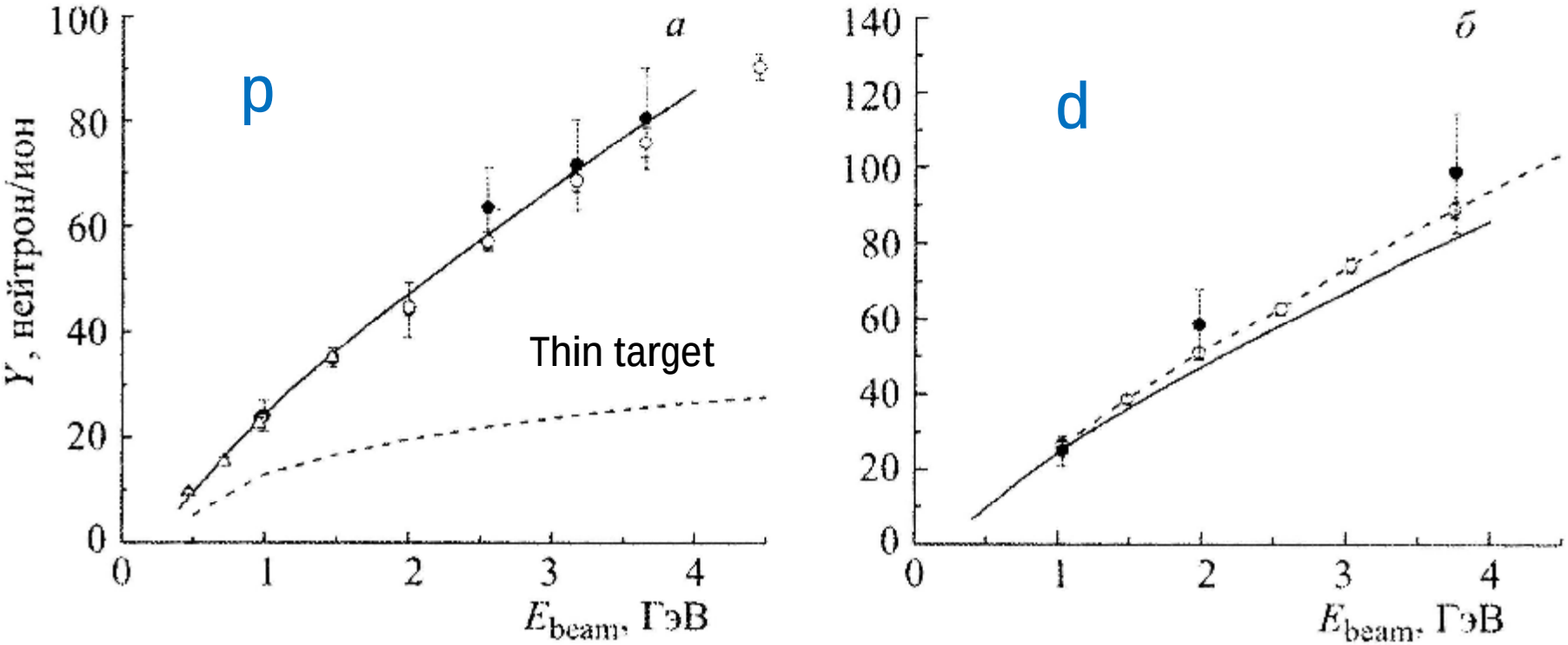
- *In this of "quasi-infinite" (QI) deep subcritical active core (DS AC) maximally hard neutron spectrum has been realized with rather low $k_{eff} \sim 0.4$*

- *Such neutron spectrum is very effective for burning (via (n,f)- reaction) of threshold minor actinides and transmutation of long lived fission products (via (n,xn)-reactions) from spent nuclear fuel (SNF) composition*

- *So it is very attractive to investigate ADS with such type of active core for higher incident energy*

- *What is additionally known from study of massive target irradiated by GeV particles?*

Mean neutron multiplicities for thick (ø20x60cm) lead target in dependence on incident particle energy



For ø20x60cm lead target irradiated by GeV deuterons
from V. Yurevich et al (2006)
TOF study of spectral characteristics of leaked neutrons

E_d GeV	<En> MeV	En kin MeV	En kin/E_d %	W MeV	W/E_d %
1.03	6.5	162	16	336	32.6
1.98	7.9	460	23	870	43.9
3.76	10.4	1025	27	1717	45.7

- *Very hard neutron spectrum < En > ® (6.5 ¸ 10.4) MeV*
- *Share of neutron energy spectrum with E_n>20MeV is 10%*
- *W/E_d is a share of deuteron energy E_d expended in the formation of neutrons. It grows with incident energy !*

- *So for multiplicative quasi-infinite deep subcritical target it is plausible to expect an essential growth of total neutron multiplicity (and integral fission yield) with increase of incident energy*

- *To study this problem is just an aim of JINR project "Energy and Transmutation Radioactive Waistes" ("E&T RAW") adopted for realization during 2011-2016 on the basis deuteron (proton) beam of JINR NUCLOTRON in incident energy range 1- 10 GeV/N and natural (depleted) massive uranium targets available at JINR .*

Motivation

Main goals of E&T – RAW project are

- *to study basis characteristics of neutron fields inside quasi-infinite deep subcritical AC, spatial distributions of the core nuclei fission, ^{239}Pu production and transmutation reaction rates for long lived minor actinides and fission products in hard neutron spectrum*
- *As well as to define optimal energy of incident beam for transmutation RAW and simultaneous energy production.*

- *For full scale modeling of the considered ADS at JINR there is the quasi-infinite (~20 tones) AC – Big depleted URANium target assemly (BURAN) and high energy beams of super conducting accelerator NUCLOTRON.*

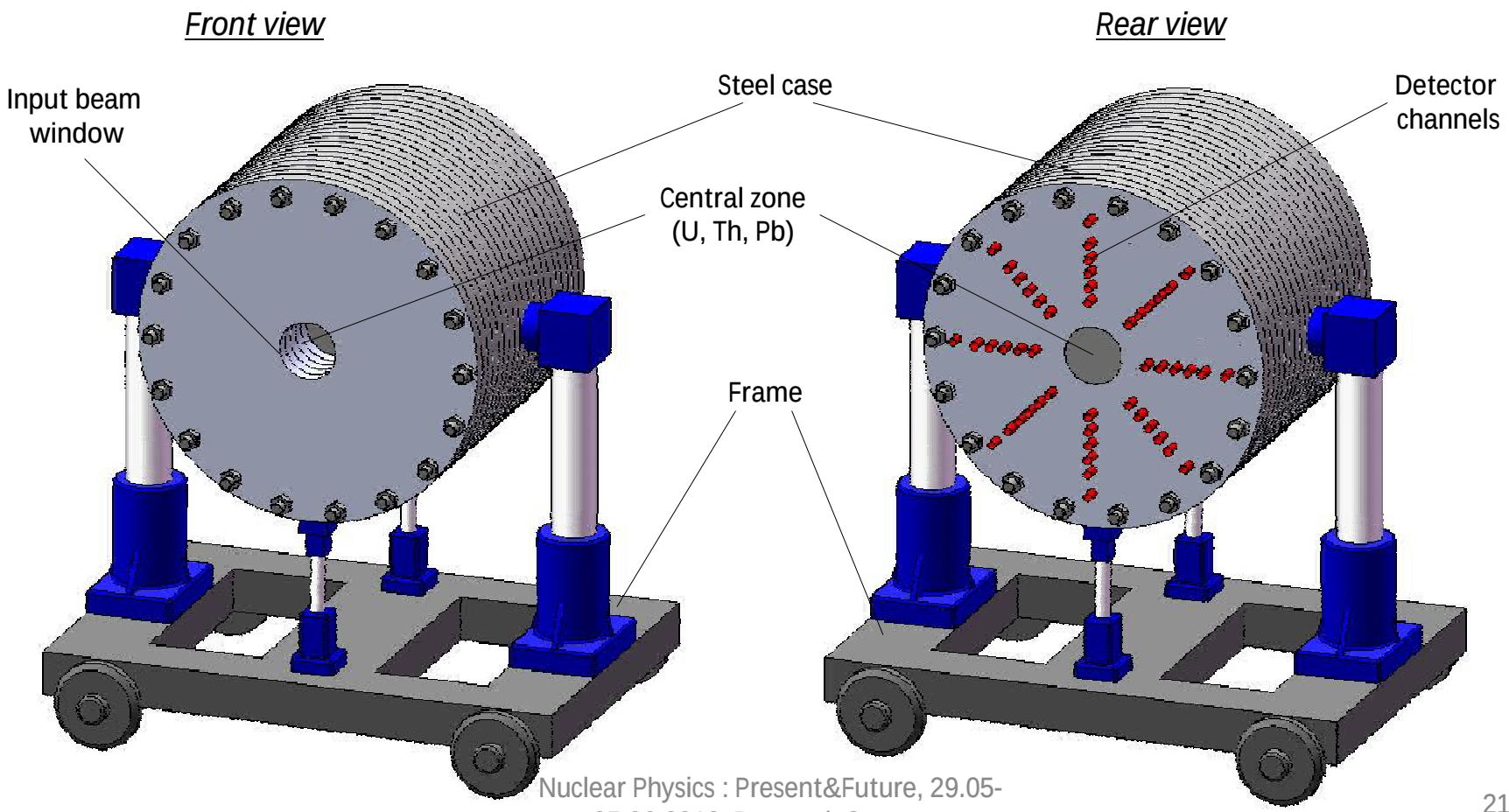
- *As a preparatory stage for main experiments with BURAN there was used an intermediate size massive (512 kg) target assembly (TA) QUINTA from natural metallic uranium.*

- *TA QUINTA models the central part of TA BURAN and allows to work out and to test the measurement and data analysis procedures for main experiments.*

Quasi-infinite depleted uranium target (BURAN) with replacement central zone

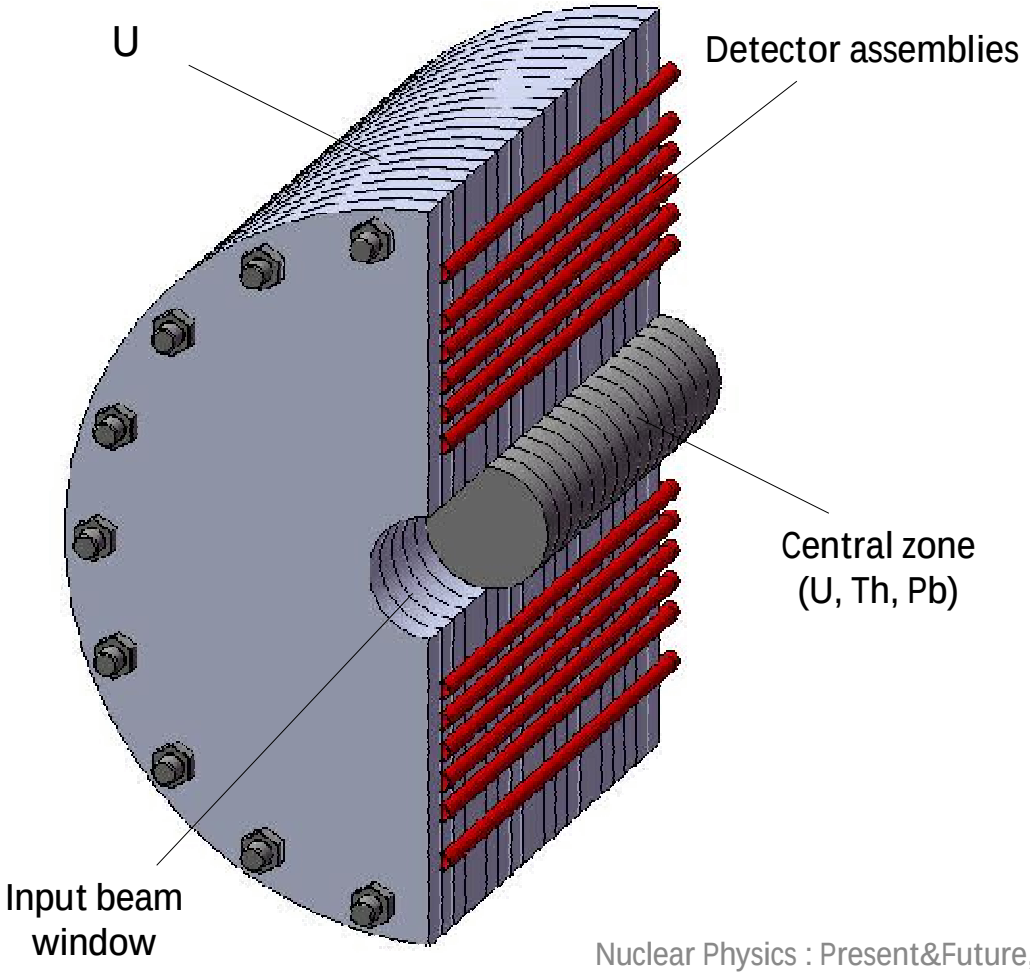
Mass of uranium – 19.5 **т.**
Diameter – 1,2 **м.**
Length – 1 **м.**

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

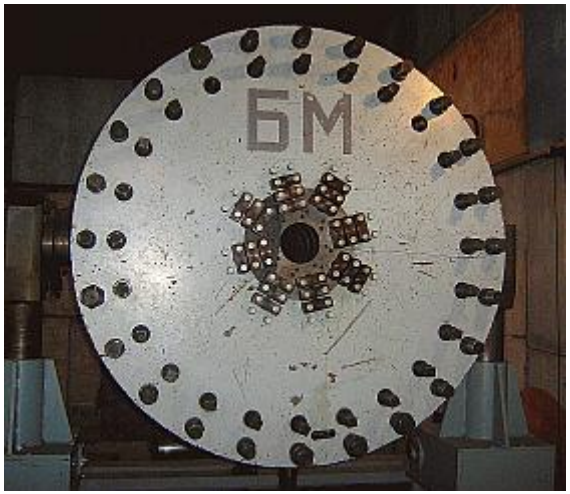


Quasi-infinite depleted uranium target (BURAN) with replacement central zone

Longitudinal section of TA BURAN together with central zone and detector channels

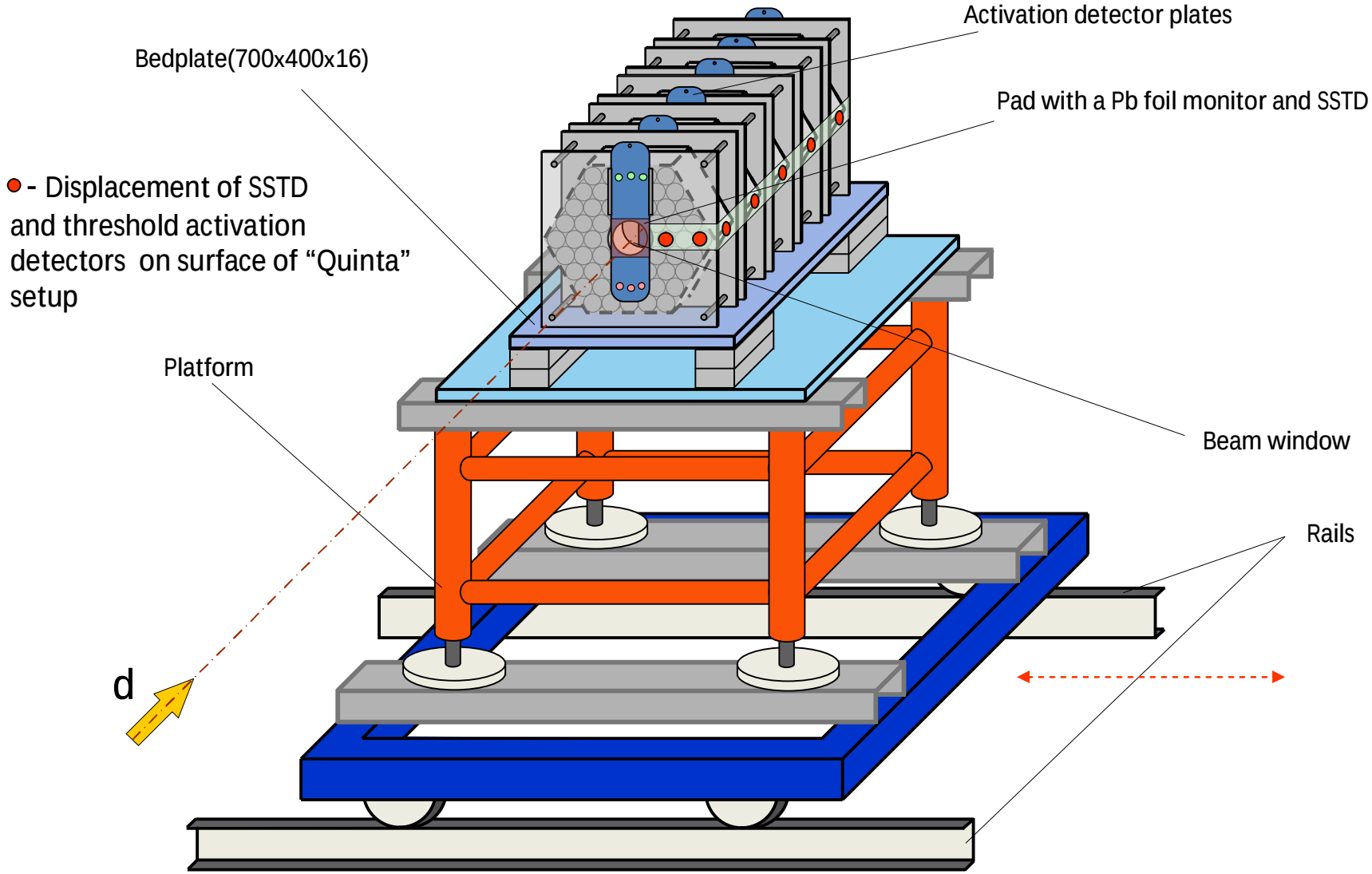


Front view photo

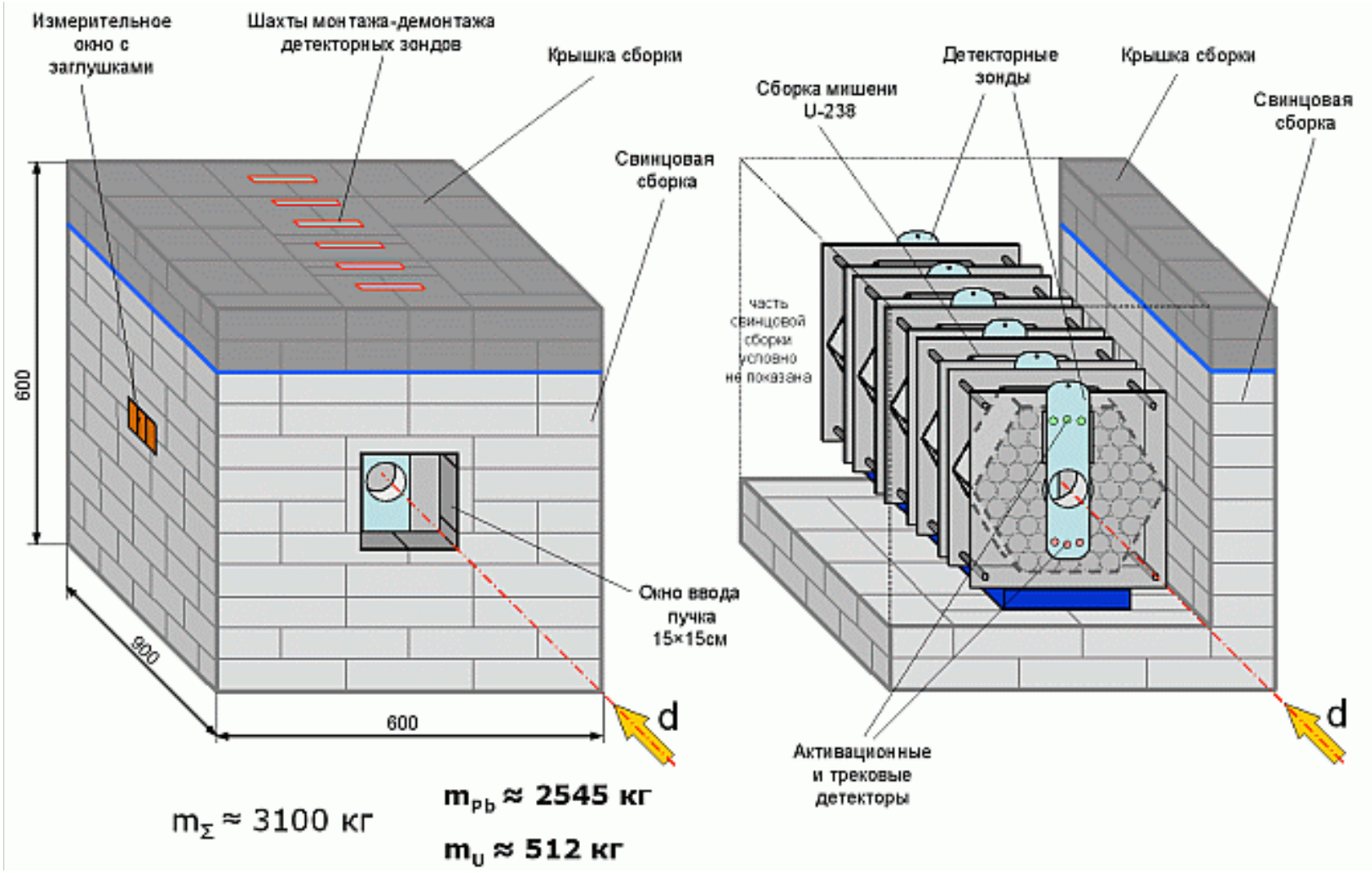


Rear view photo

Target assembly "Quinta" at the irradiation position (March 2011)



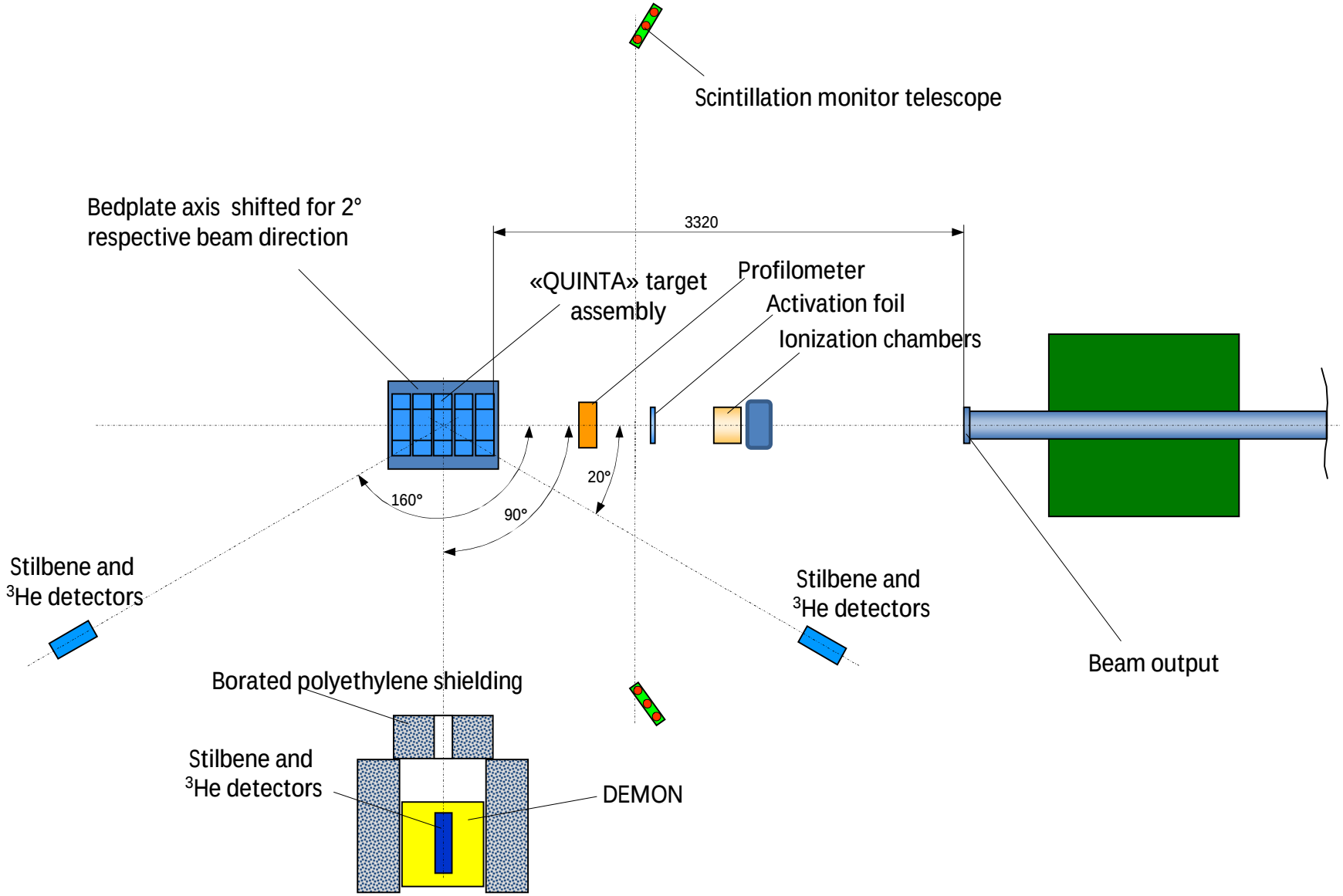
TA QUINTA with lead blanket



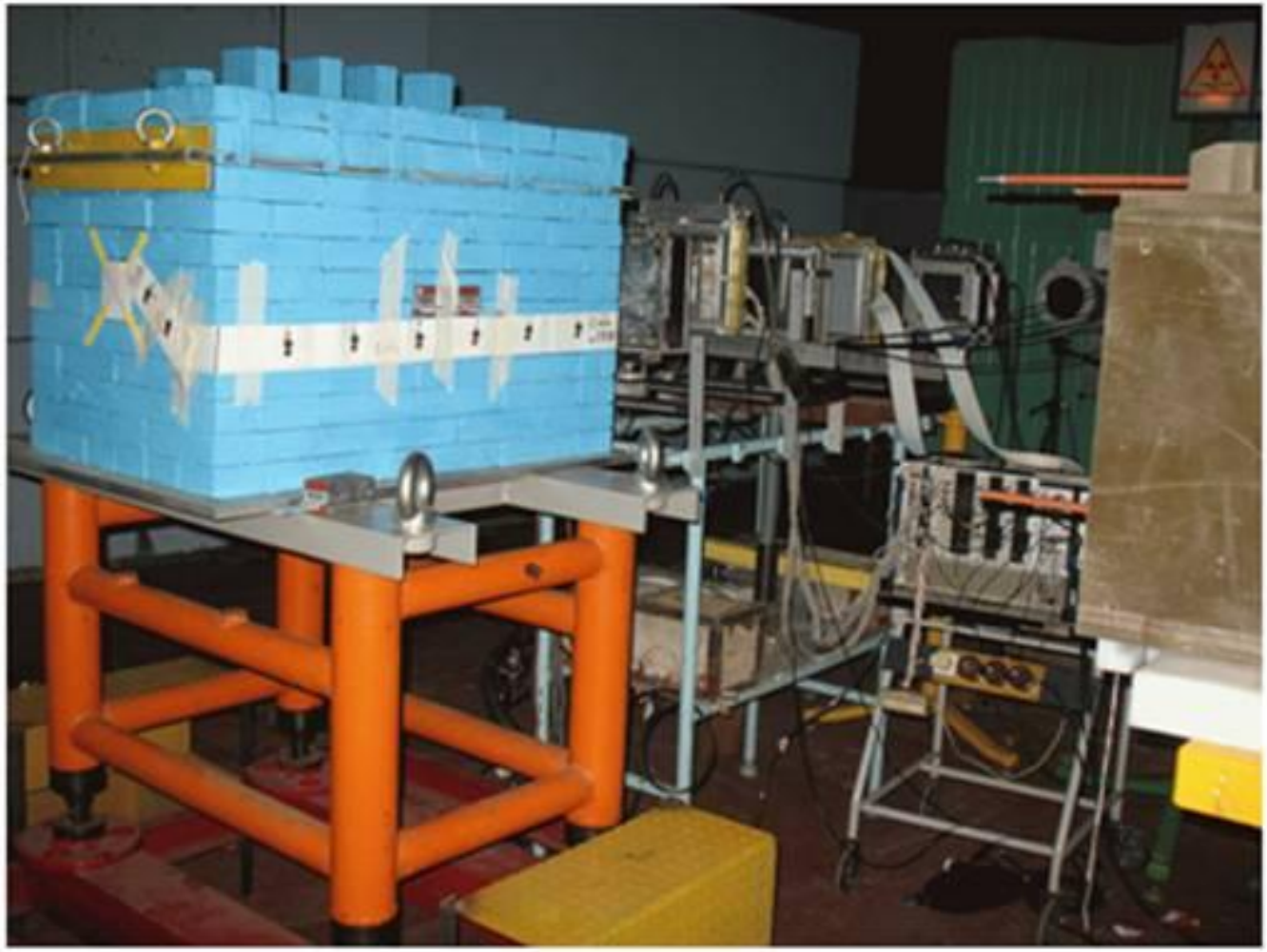
Results of experiments 2011-2013

- *In present talk it'll be discussed main results of experiments carried out with TA QUINTA during 2011-2013*

Scheme of experiment with "QUINTA" setup

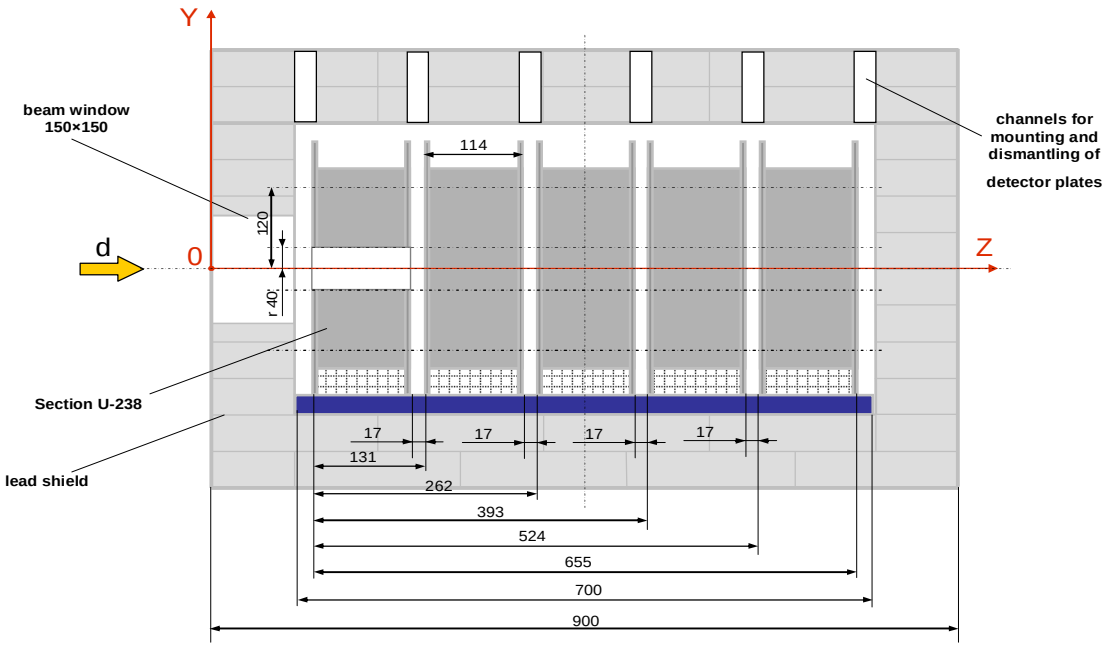


TA QUINTA ON IRRADIATION POSITION (March 2013)



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.

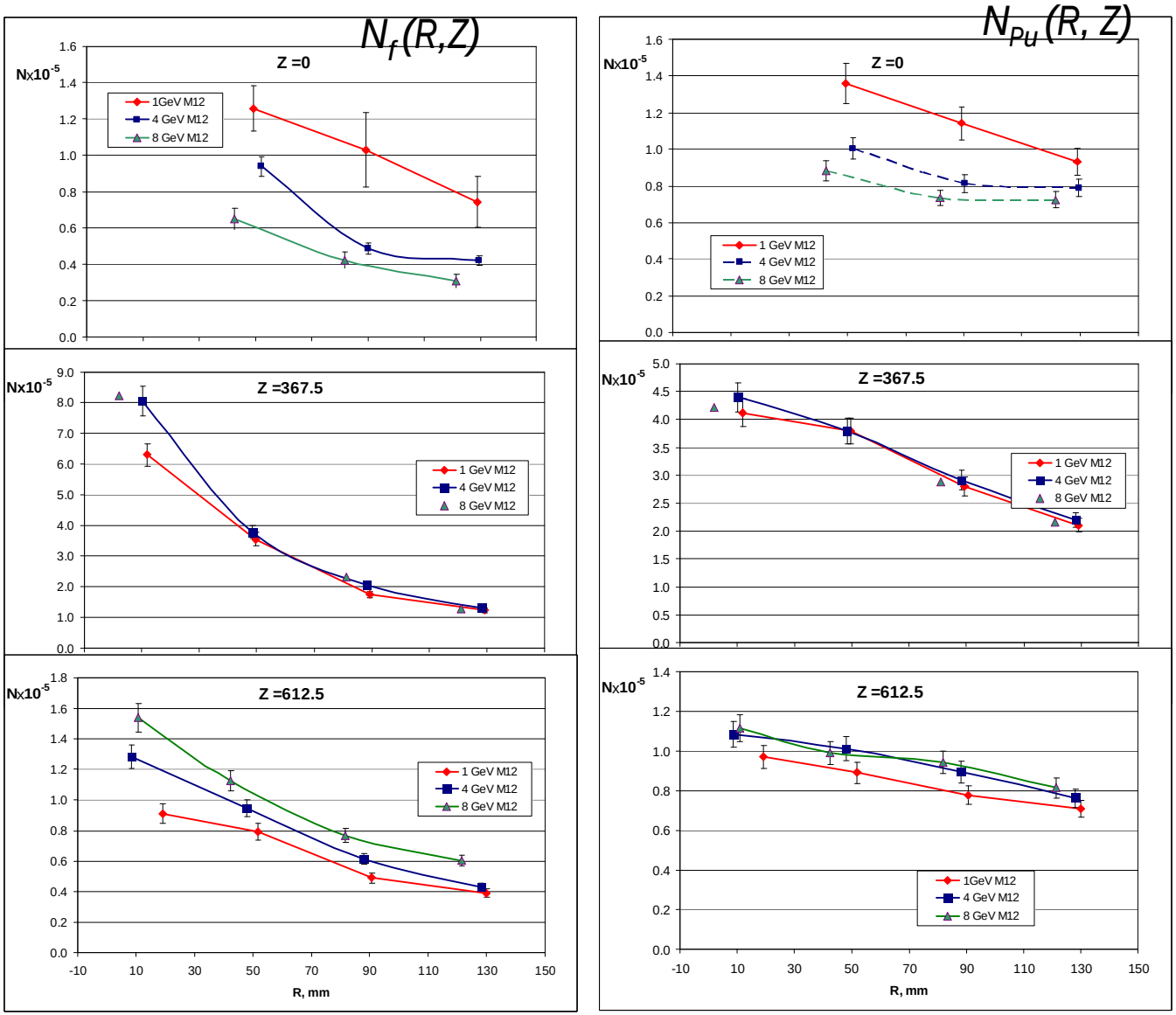
Number of fissions in the place of measurements defines by averaging of following fission product yields:

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

.

In brackets there are mean cumulative FP yields

Spatial distributions of ^{nat}U fission $N_f(R,Z)$ and ^{239}Pu production $N_{Pu}(R, Z)$ within TA QUINTA (in units – per 1 deuteron/ 1 g 238U/ 1 GeV)



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		
03.12 (SSTD)	8.9 ±1.5		8.1 ±1.5		9.2 ±1.6
03.12	(10.2±0.5) ±1.1		(9.6± 0.4) ±1.0		(9.4±0.5) ±1.0
12.12		(10.5±0.5) ±1.1	(10.3±0.5) ±1.1		(9.3±0.5) ±1.0
Calc.	*9,5*	*9,1*	*8.3*	*7,7*	*7,3*
	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		
03.12	(11.3±0.6) ±1.2		(11.0±0.5) ±1.1		(10.2±0.5) ±1.1
12.12		(12.5±0.7) ±1.3	(12.2±0.7) ±1.3		(10.3±0.5) ±1.1
Calc.	*12*	*11.8*	*11,6*	*10.8*	*9,2*

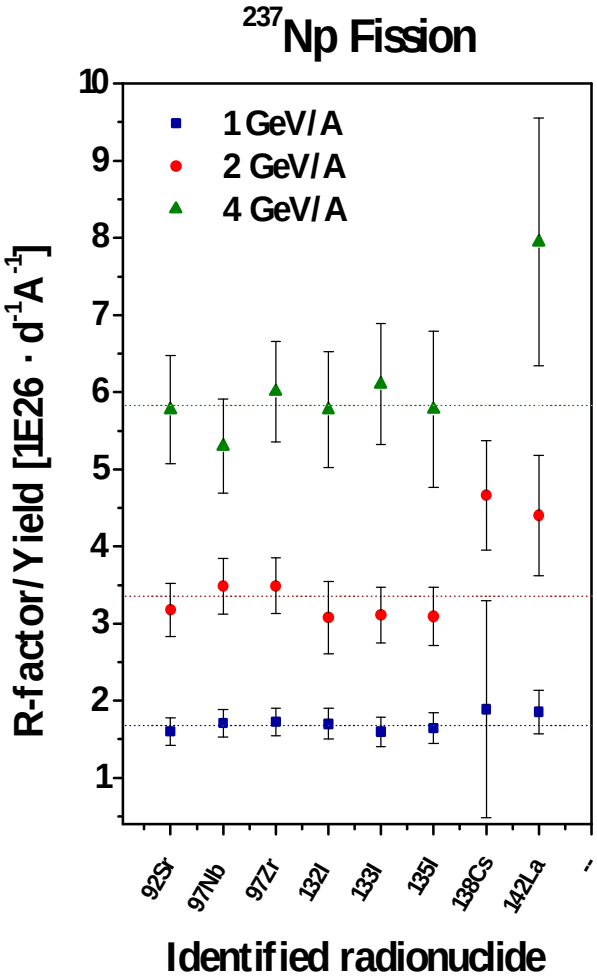
Total numbers of fission $N_f(tot)$ and produced ^{239}Pu nuclei ®

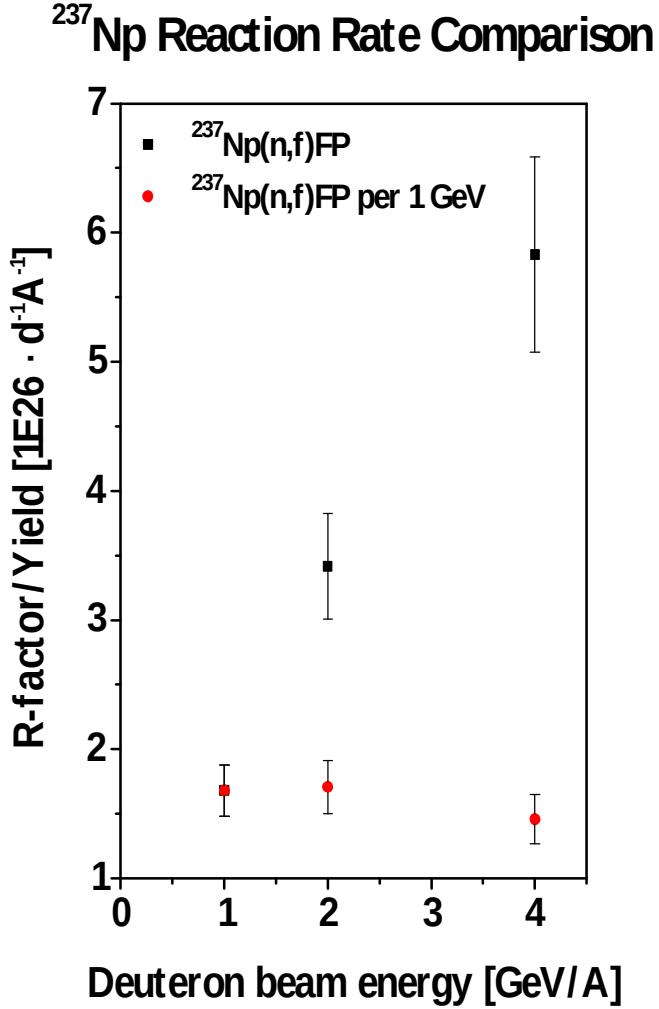
experiment v.s. calculation

*Reaction rates (atom^{-1}, d^{-1}, * [E-27])*

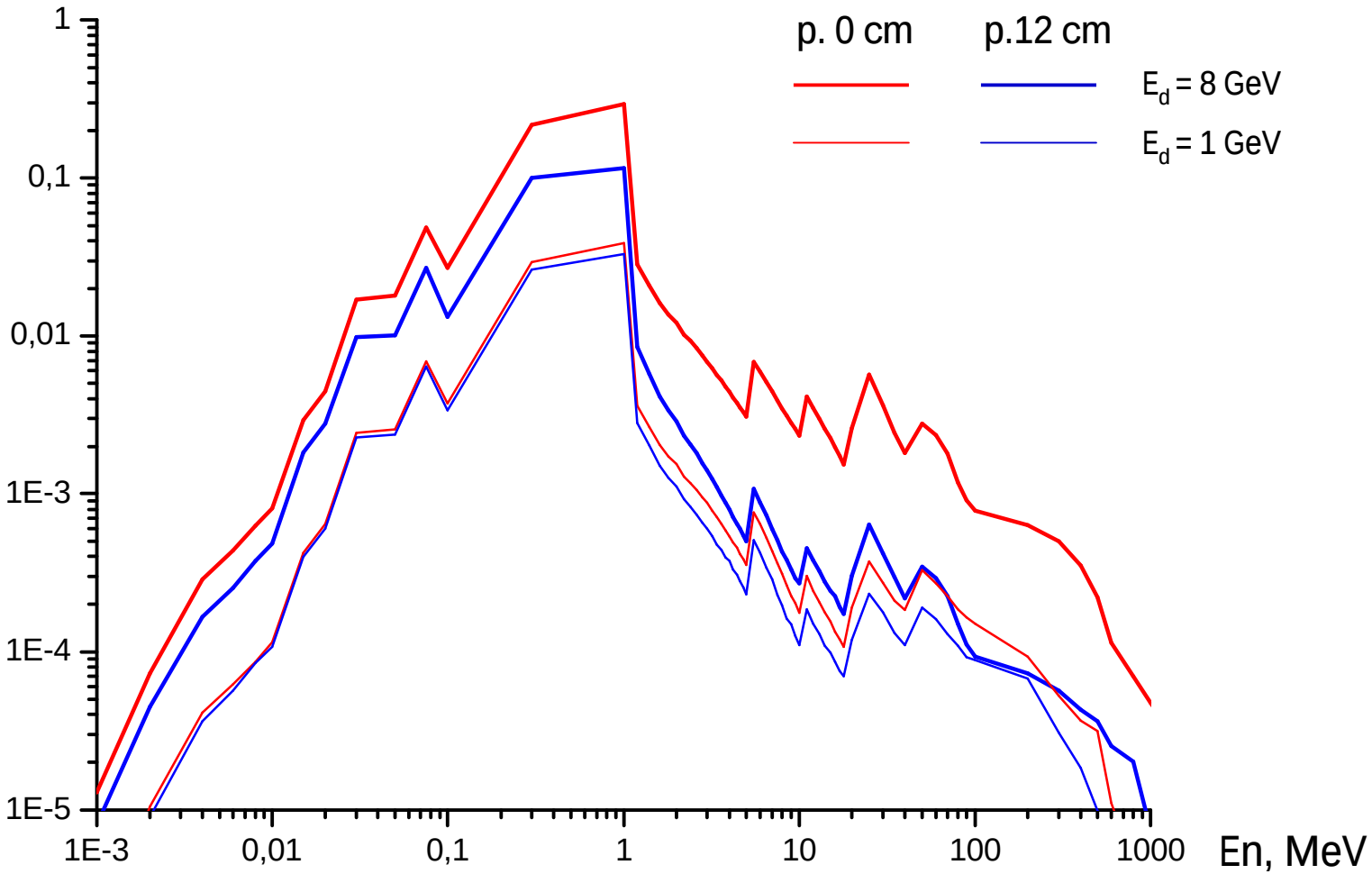
Reaction products	Ed = 2 GeV Reza et al.	Ed = 4 GeV Adam et al.	Ed = 6 GeV Adam et al.
Th(n,g)**Pa-233**	76.9(39)	142(4)	176(3)
Th(n,2n)**Th-231**		51.4(15)	71.2(23)
Th(n,6n)**Th-227**		3.8(15)	4.4(4)
Th(n,p6n)**Ac-226**	1.13(8)	2.98(21)	3.41(11)
Th(n,p8n)**Ac-224**	0.62(5)	1.37(6)	3.1(3)
Th(n,**fission**)	**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)**

^{237}Np Results

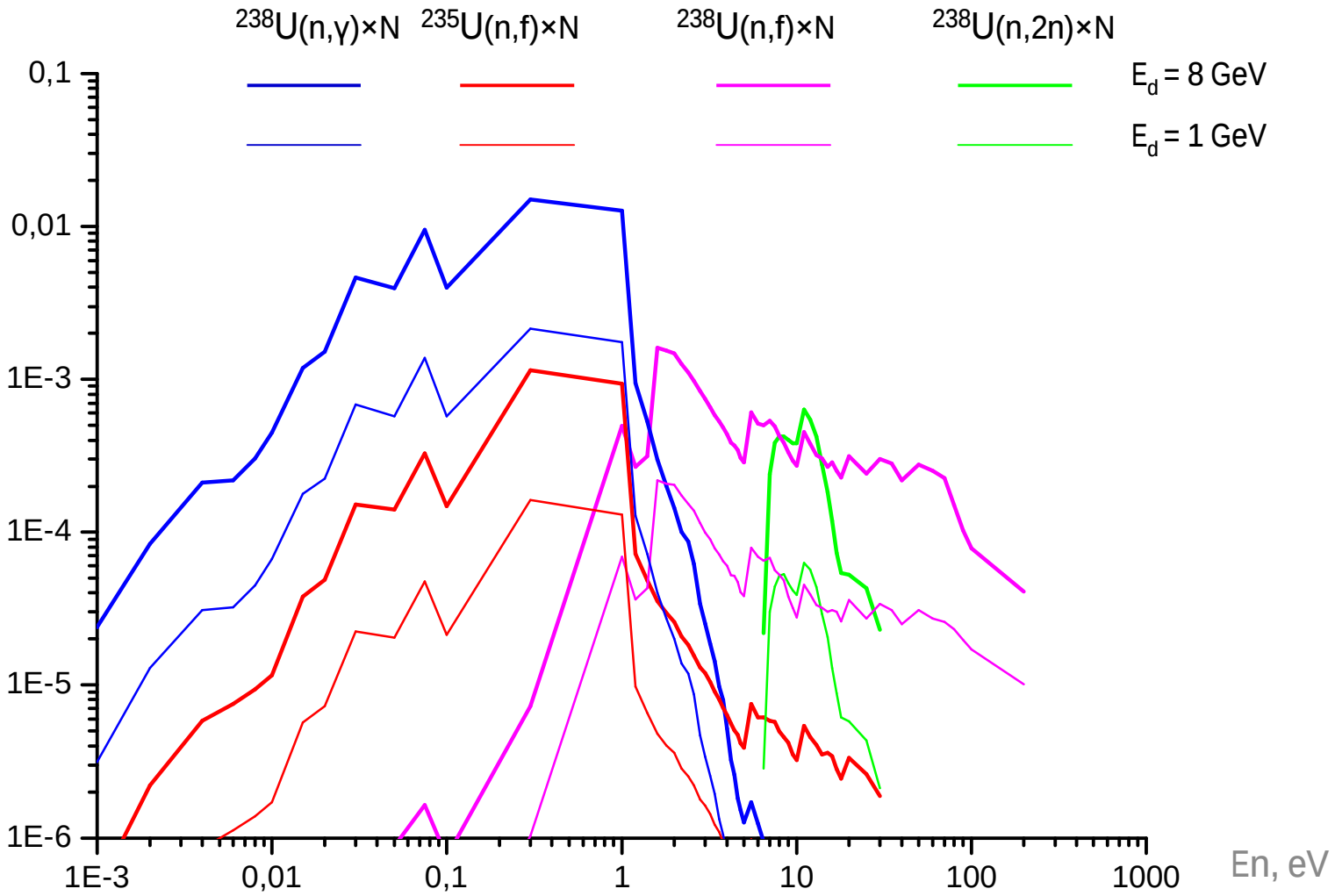




Typical calculated neutron spectra at positions R= 0 and 12 cm, Z=64,5 cm (fourth plate)

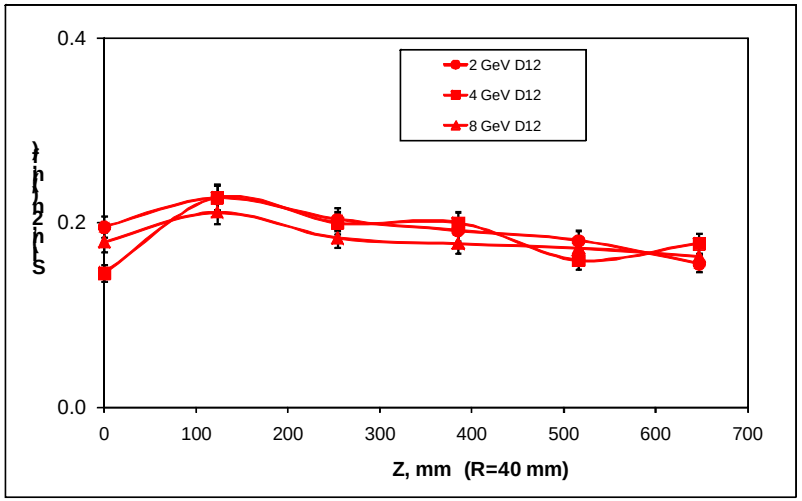


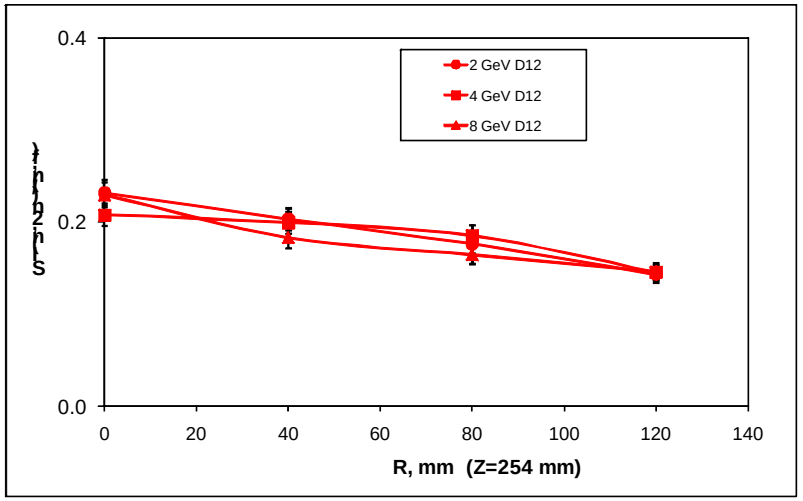
Convolution of cross-sections (n,γ), (n,f) and (n,2n) with neutron spectra at position R=12 cm in the fourth plate



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

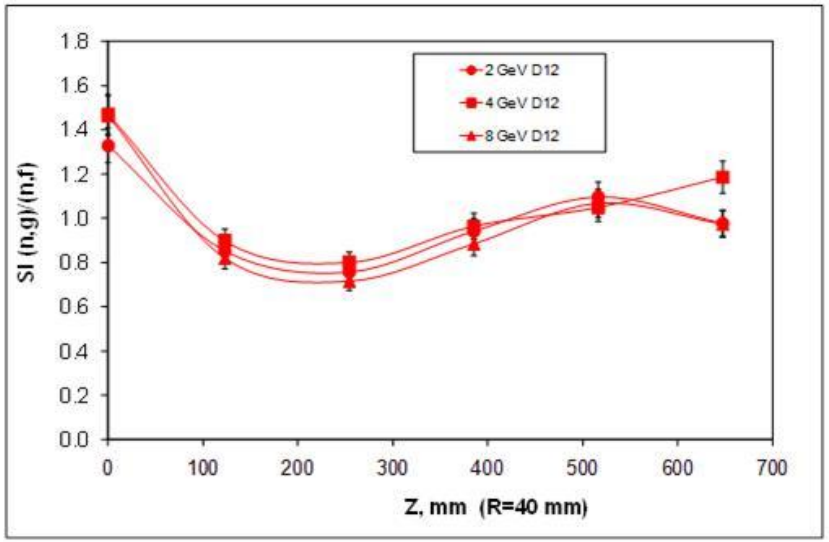
- *Spectral indices are relative values. Their uncertainties are much smaller than for each $N_{(n,2n)}(R,Z)$, $N_f(R,Z)$ or $N_g(R,Z)$ values. SI do not depend on beam normalization*
- *The threshold of (n,f)-reaction ~ 1.2 MeV and for (n,2n) ~ 7 MeV*
- *Constancy of SI depending on Z, R and Ed indicates 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.*

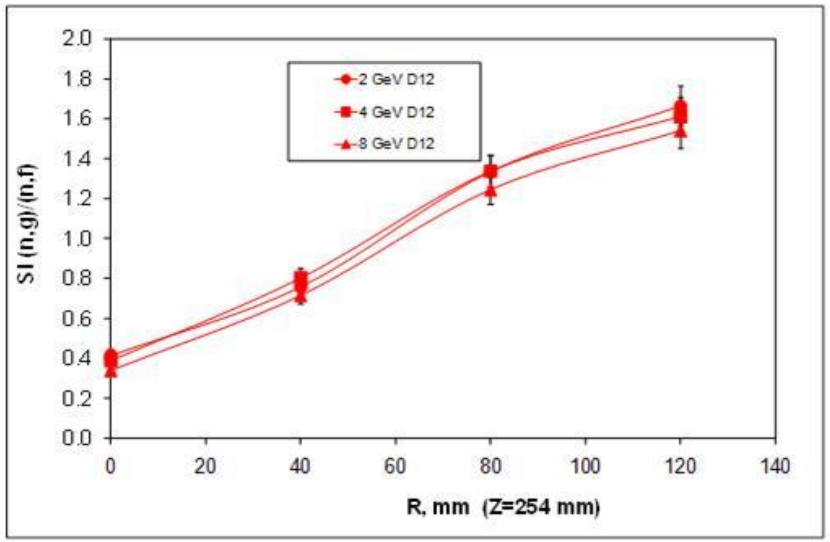




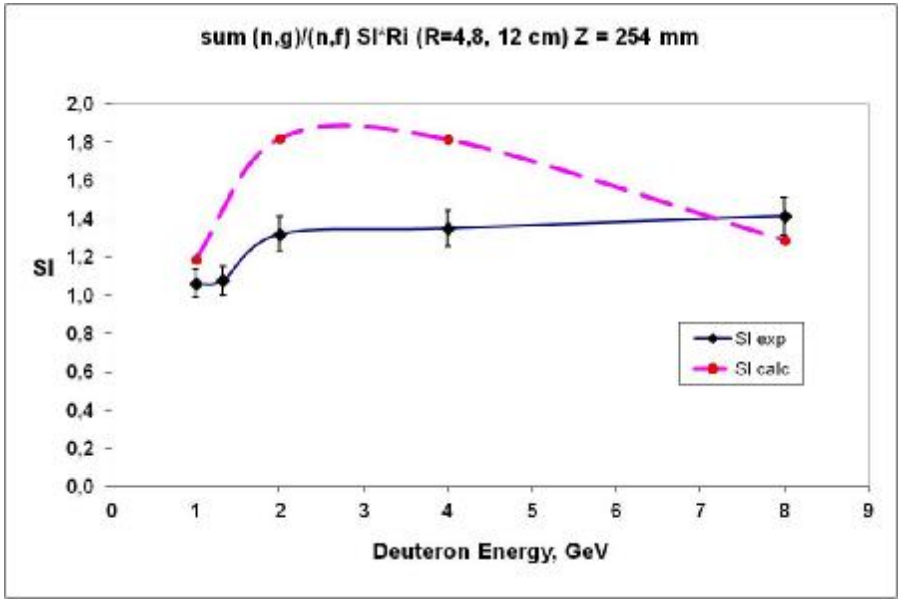
Spectral indices SI (n,γ)/(n,f)

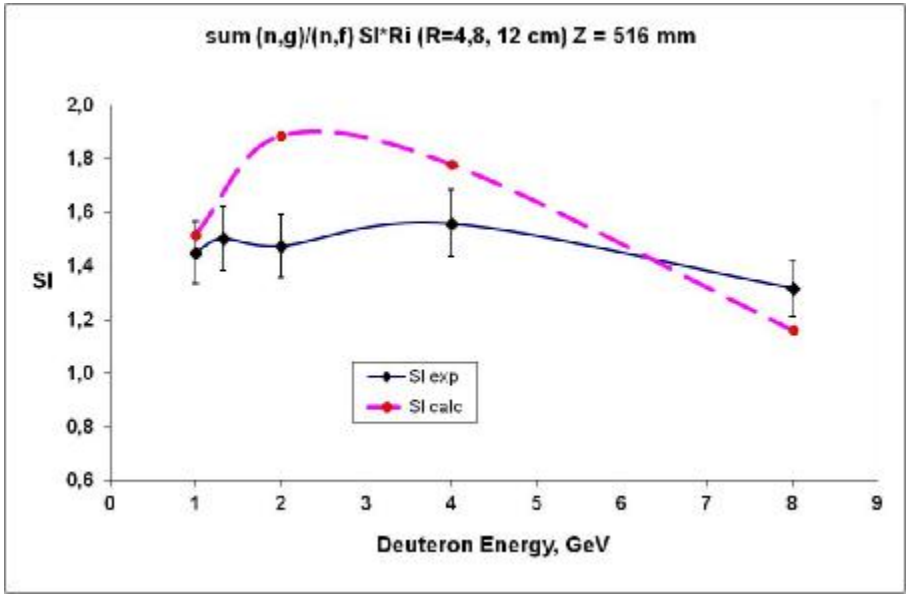
- *Neutron spectrum becomes "softer" with increase the distance from beam axis*





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



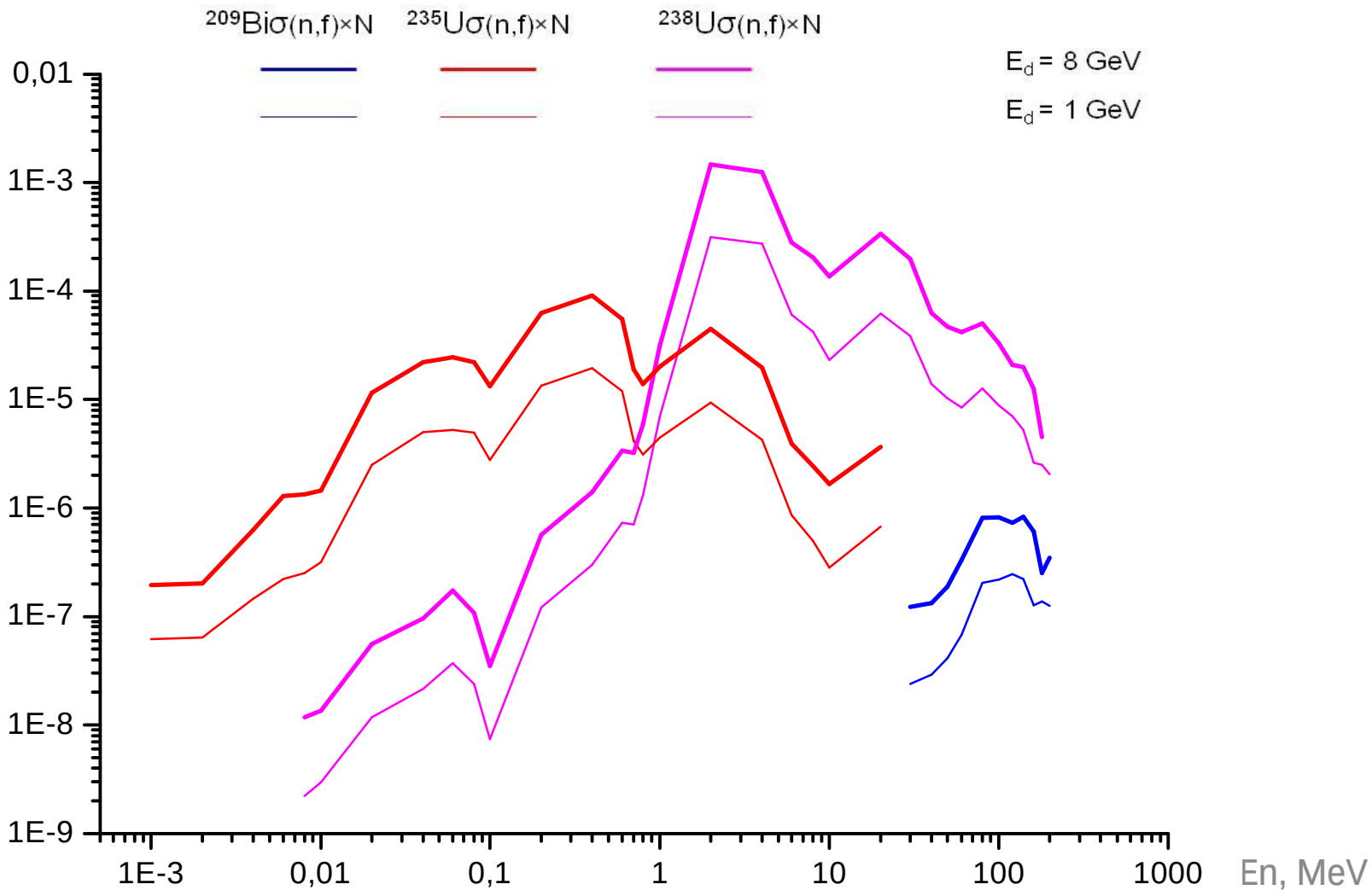


- *Share ratios of the neutron spectrum in the energy ranges*

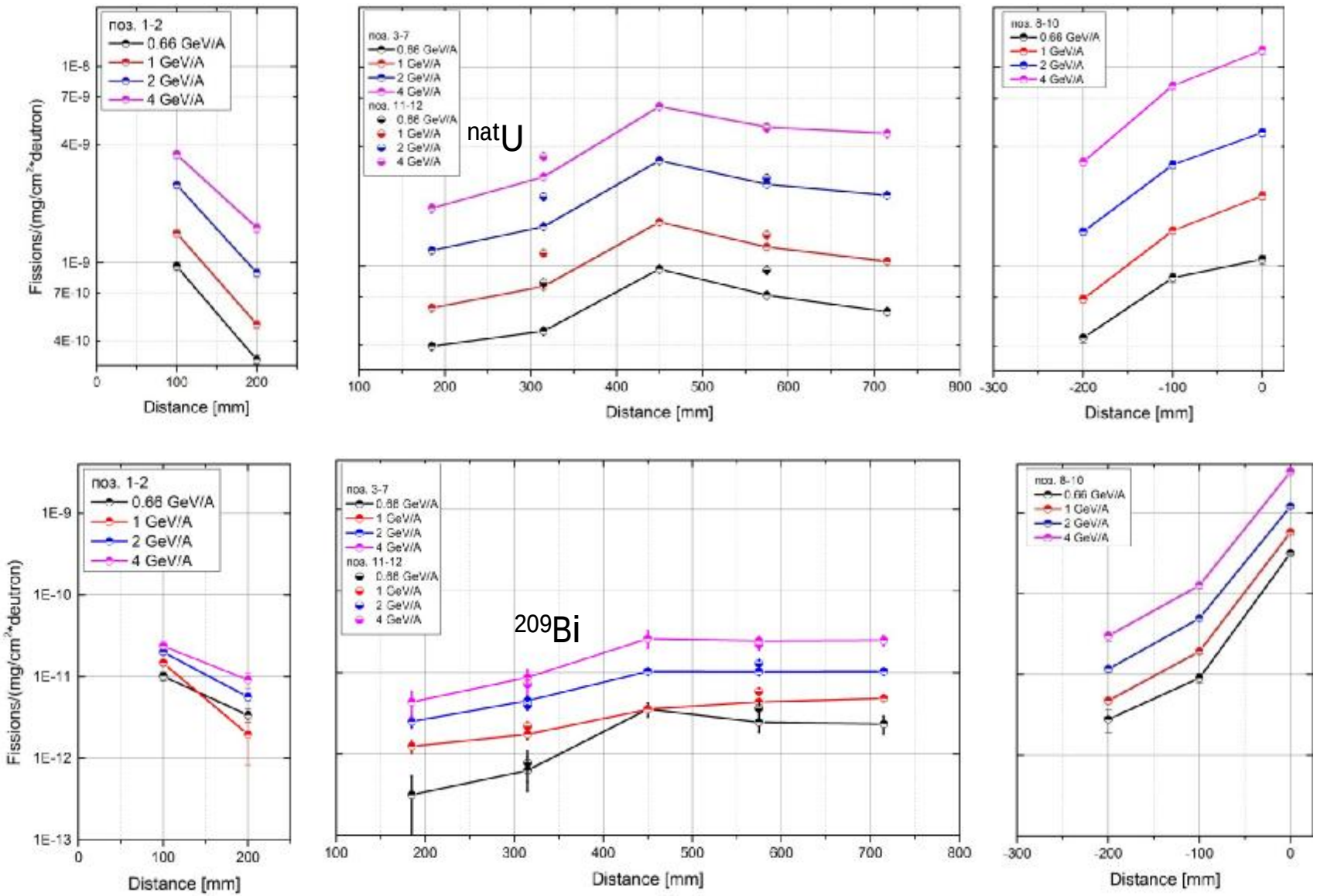
En < 1.2 MeV and 1.5 Mev < En< 20MeV remain approximately constant with increase of incident energy

- But what about En > 20 MeV ?

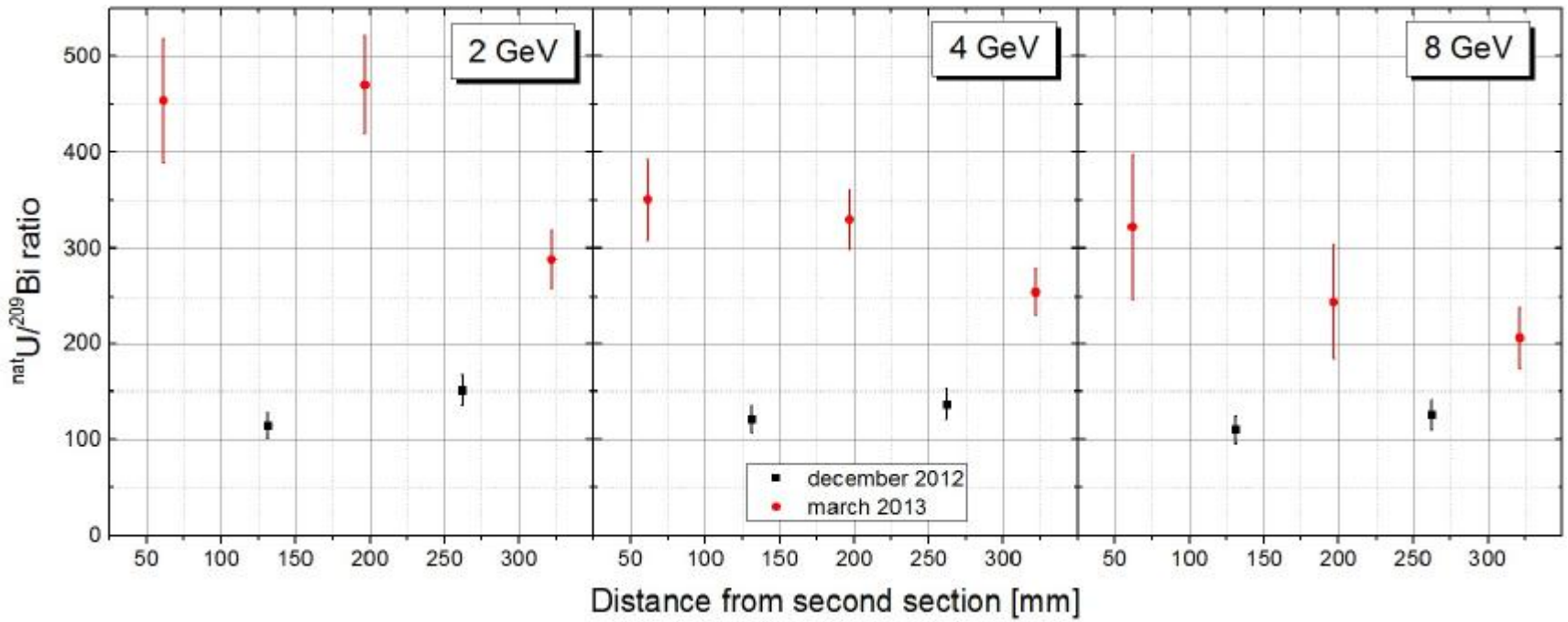
Convolution of neutron spectra with σ(n,f) for $^{nat.}U$ and ^{209}Bi at position R=12 cm in the fourth plate (Z=64,5 cm)



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

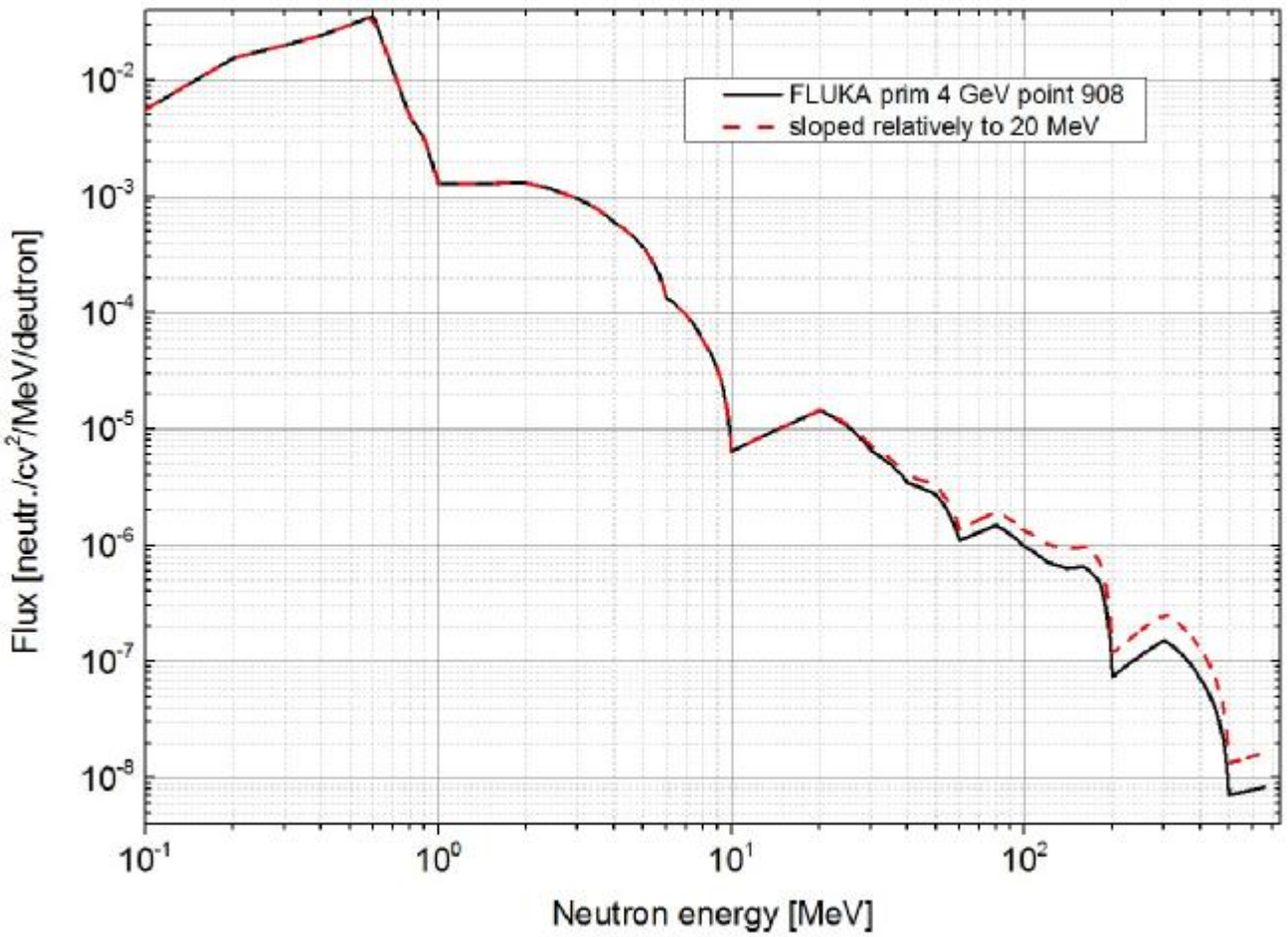


Ratios of $^{nat}U/^{209}Bi$ (n,f)-reaction rates
(black – inside of TA QUINTA, red – on the surface)



Energy spectrum of leakage neutrons becomes more "harder" with increase of deuteron energy !

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



Spectral characteristics of leakage neutrons on QUINTA surface

E_d, GeV	1,32	2,0	4,0	8,0
Total 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

Total neutron multiplicity and leakage vs. incident energy

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., MCNP)	60.7	62,7	57,3	-
M(calc., MARS)	77,5	74,7	67,5	60,5
N_L / M (exp), %	80±19	80±19	82±20	83±19

Beam power gain vs. size of AC

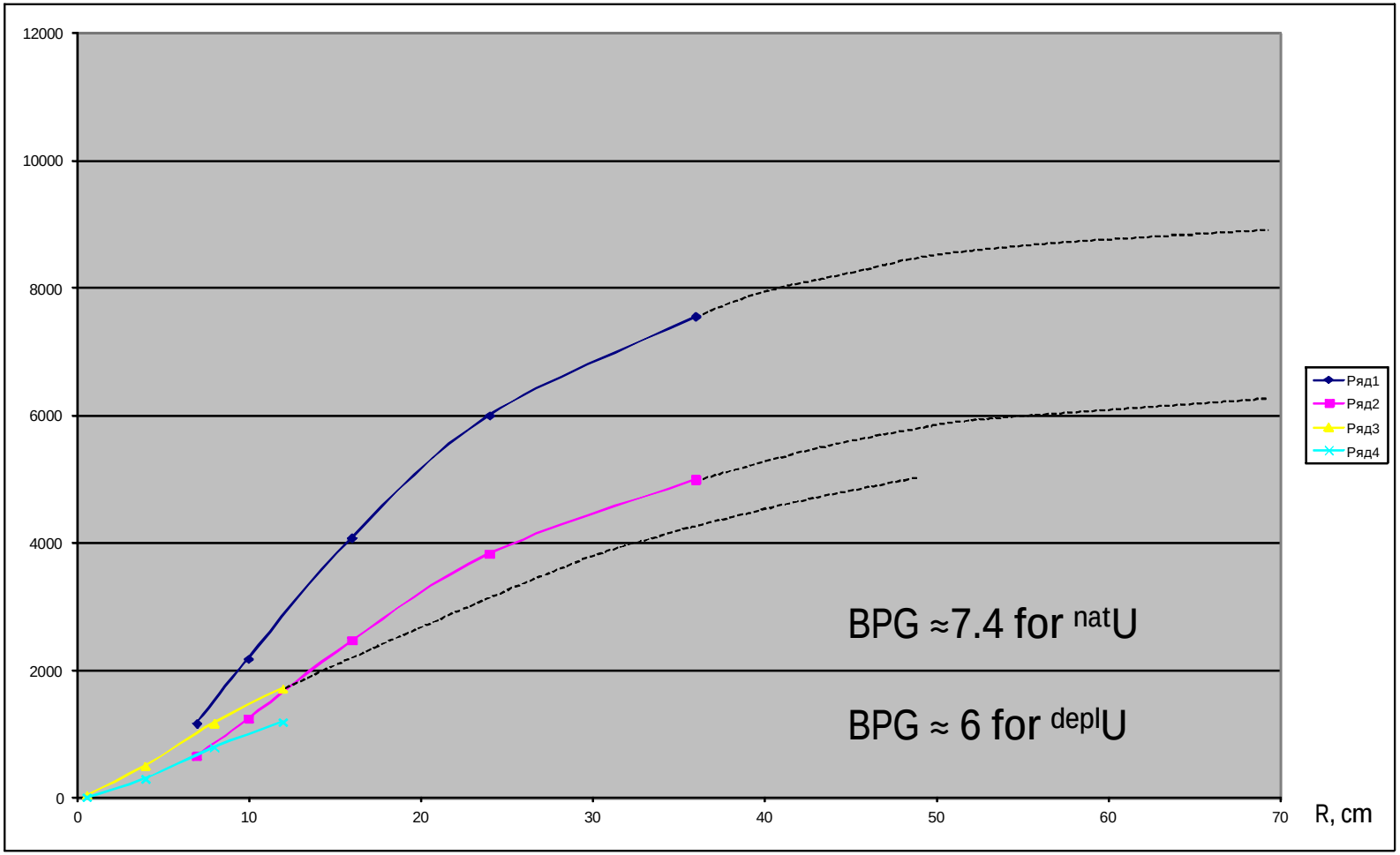
- *Beam power gain ($BPG = E_{release}/E_{beam}$) is key characteristics of any ADS. It defines practical applicability of the system.*

- *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 et al. (1978).*

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



Beam power gain vs. incident energy

- *In experiment of Vassilkov et al. BPG ≈ 7.4 for ^{nat}U and ≈ 6 for ^{depl}U (Ep=0.66 GeV, uranium mass – 3.2 tons, neutron leakage ~ 10%).*

- *This result was not reproduced by any calculations.*

- *Moreover calculations 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 →

(n,f), (n,ɤ) and M^{tot} linear growth with $E_{d(p)}$ but BPG ?!

	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 vs. 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 our measurements of ratios $^{209}Bi/^{nat}U$ fission rates with TA QUINTA show energy spectrum of leakage neutrons becomes more "hard" with increase of incident energy.*

Beam power gain vs. 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*

Main goals 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.*

Conclusion

- *The project "E&T RAW" aimed at study of ADS with deep subcritical AC suitable for utilization of SNF and energy production is implemented at JINR on base of NUCLOTRON beams and massive uranium targets QUINTA and BURAN*
- *The measurements performed in 2011-2013 shown that numbers of core nuclei fission and ^{239}Pu production grows linearly with increase energy of incident particles*
- *Energy spectrum of neutrons leaking TA QUINTA surface becomes harder with grows of incident particle energy*

Conclusion(cont.)

- *Calculations are not able to reproduce high energy (E_n >20 MeV) tail of neutron spectrum inside and outside of TA QUINTA which is very important when size of AC increase*
- *Experiments with quasi infinite AC BURAN should give decisive information on practical applicability studied ADS for utilization of SNF with simultaneous energy production*
- *In any case realization of scientific program with TA BURAN provides valuable benchmark for presently used INC models and transport codes*

Thanks for your attention