Research of accelerator-driven systems at JINR and their development prospects

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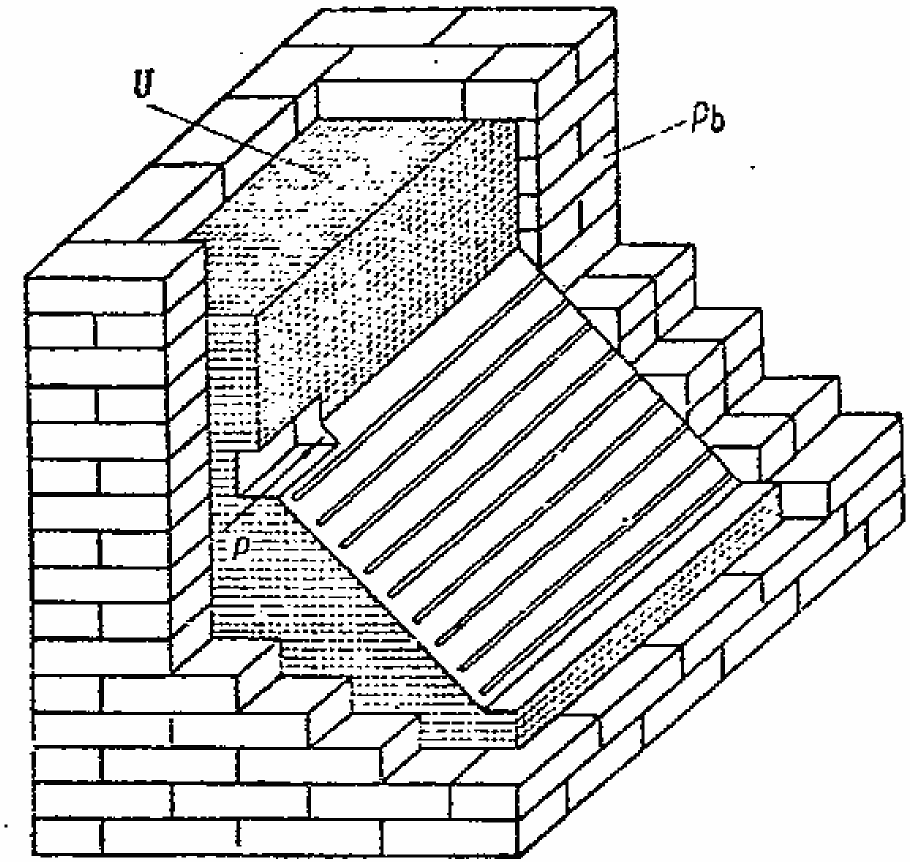
Introduction

- The key problem of global atomic energy is effective utilization of spent nuclear fuel (SNF).
- One of the ways to solve this problem considered seriously last ten years is to use the Accelerator Driven subcritical Systems (ADS).
- State programs develop ADS for transmutation of highly toxic, long lived components of SNF currently adopted in Europe, China, India and South Korea.
- ADS are much more safe than any reactors exploiting chain reaction of nuclear fission ($k_{eff} < 1$ in all in all operating modes).

- But practical use of ADS for utilization of SNF presents many technological challenges.
- First of all is to create a reliable and highly effective accelerator MW power
- No less serious problem finding and licensing of radiation-resistant materials for the active core (ADS target) and cooling systems.
- *So even the creation of the first full scale prototype ADS will take at least a decade of hard work (example – MYRRHA project)*

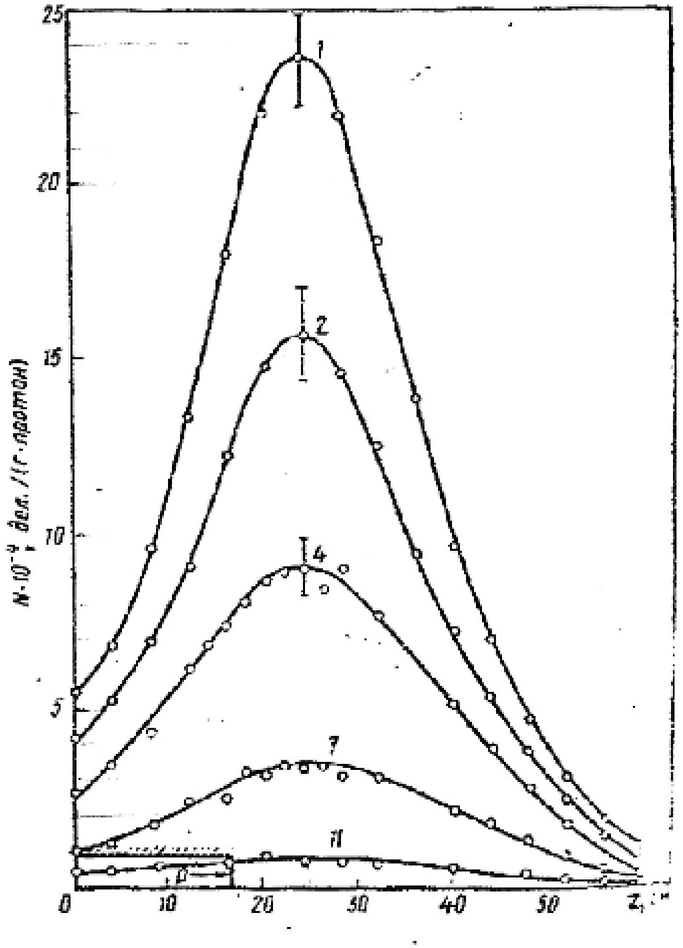
- But ADS study has more than 50 years of history.
- In particular first pioneering experimental works in this direction were performed at JINR in the 60-70s on Synchrocyclotron (*Vassil'kov, Gol'dansky, Pokotilovsky*) and Synchrophasotron (*Tolstov, Yurevich, Krivopustov et all*).
- Beside that in the same time extensive theoretical study of different kinds of ADS were implemented by *Barashenkov, Toneev and their colleagues.*
- May be conscious, but rather as a practical matter they studied deeply subcritical system of natural or depleted uranium irradiated by GeV protons or deuterons.

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



Dimensions 56x56x64cm

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



N_f axial distributions for different radial channels

Natural U

$N_f = 18.5 \pm 1.7$

$N_{Pu} = 46 \pm 4$

$BPG = 7.4 \pm 0.7$

Depleted U

$Nf = 13.7 \pm 1.2$

$N_{Pu} = 38 \pm 1.2$

$BPG = 5.7 \pm 0.5$

$Ep = 0.66$ GeV

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

- *Such neutron spectrum should be rather 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*

- *What is known additionally about the characteristics of the neutron field of similar ADS to higher incident 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*
- *W/E_d is a share of deuteron energy E_d expended in the formation of neutrons. It grows with incident energy !*

For ø20x60cm lead target irradiated by GeV protons
from V. Yurevich et al (2006)
Total neutron yield Y and partial yield Y_{20} (for En > 20 MeV)

E_p, GeV	Neutron yield per one proton, exp.		Ratio, %
	Y	Y_{20}	Y/Y_{20}
0.994	24.1±2.9	2.1±0.4	8.8±2.5
2.0	44.4±5.3	4.7±0.8	10.6±3.0

- *Share of neutron energy spectrum with E_n>20MeV is 10%*

Motivation

- *So it is very important to study ADS with quasi-infinite deep subcritical core for incident energy above threshold of fast fragmentation (~ 1.5 GeV) , i.e. in conditions when total neutron multiplicity (and integral fission yield respectively) should essentially increase*
- An investigation of 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 .
- *The project supported by grants of plenipotentiaries of Belarus, Bulgaria, Kazakhstan, Poland and Czech Republic*

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.*

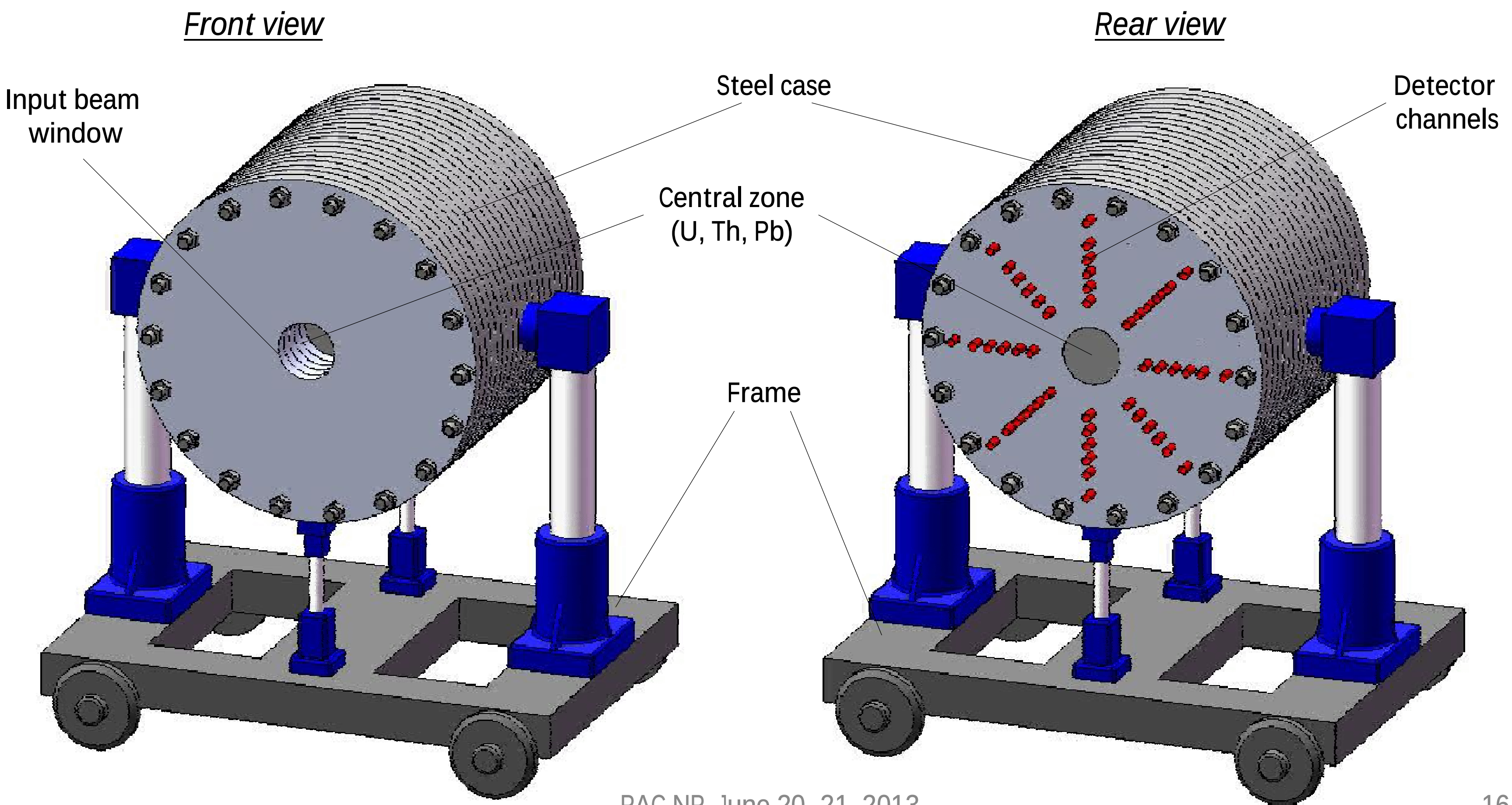
- *For full scale modeling of the considered ADS at JINR there is the quasi-infinite (~20 tones) AC – Big depleted URANium target assemly (BURAN) and high energy beams of super conducting accelerator NUCLOTRON.*

- *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.*

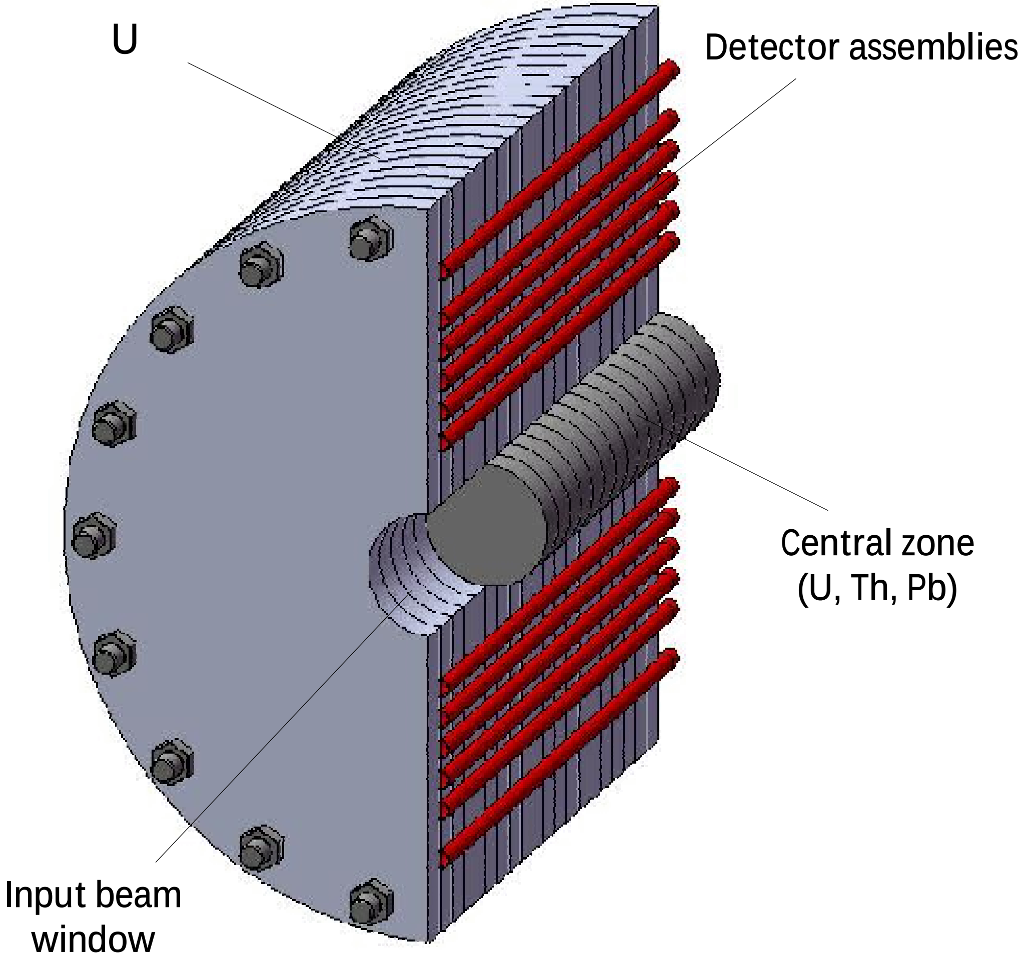
Quasi-infinite depleted uranium target (BURAN) with replacement central zone

Mass of uranium	– 19.5 т.	Materials of central zone	– U, Th, Pb.
Diameter	– 1,2 м.	Diameter of central zone	– 0,2 м.
Length	– 1 м.		

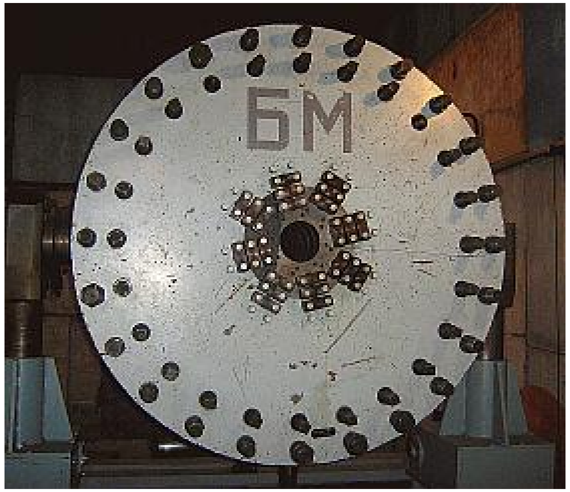


Quasi-infinite depleted uranium target (BURAN) with replacement central zone

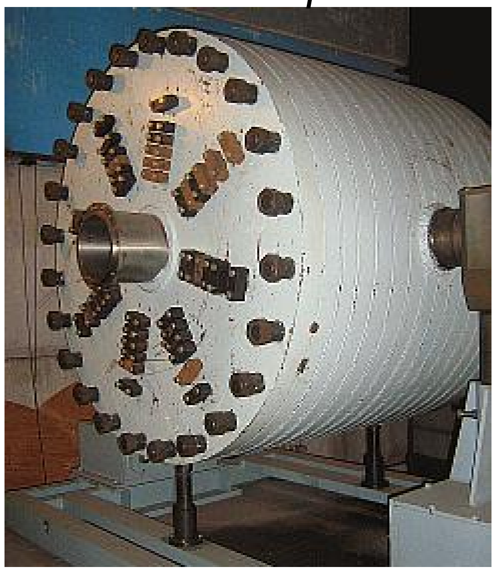
Longitudinal section of TA BURAN together with central zone and detector channels



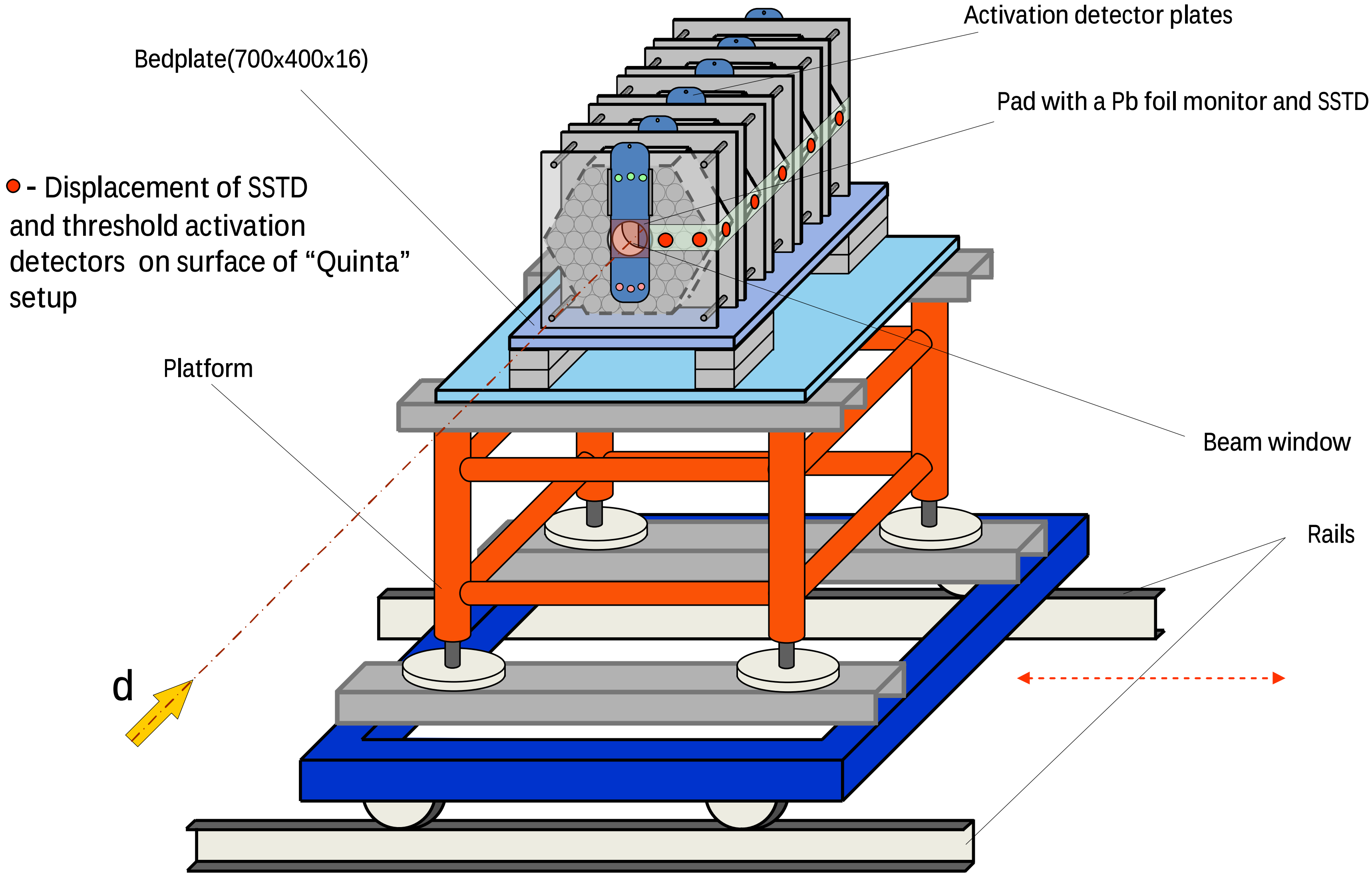
Front view photo



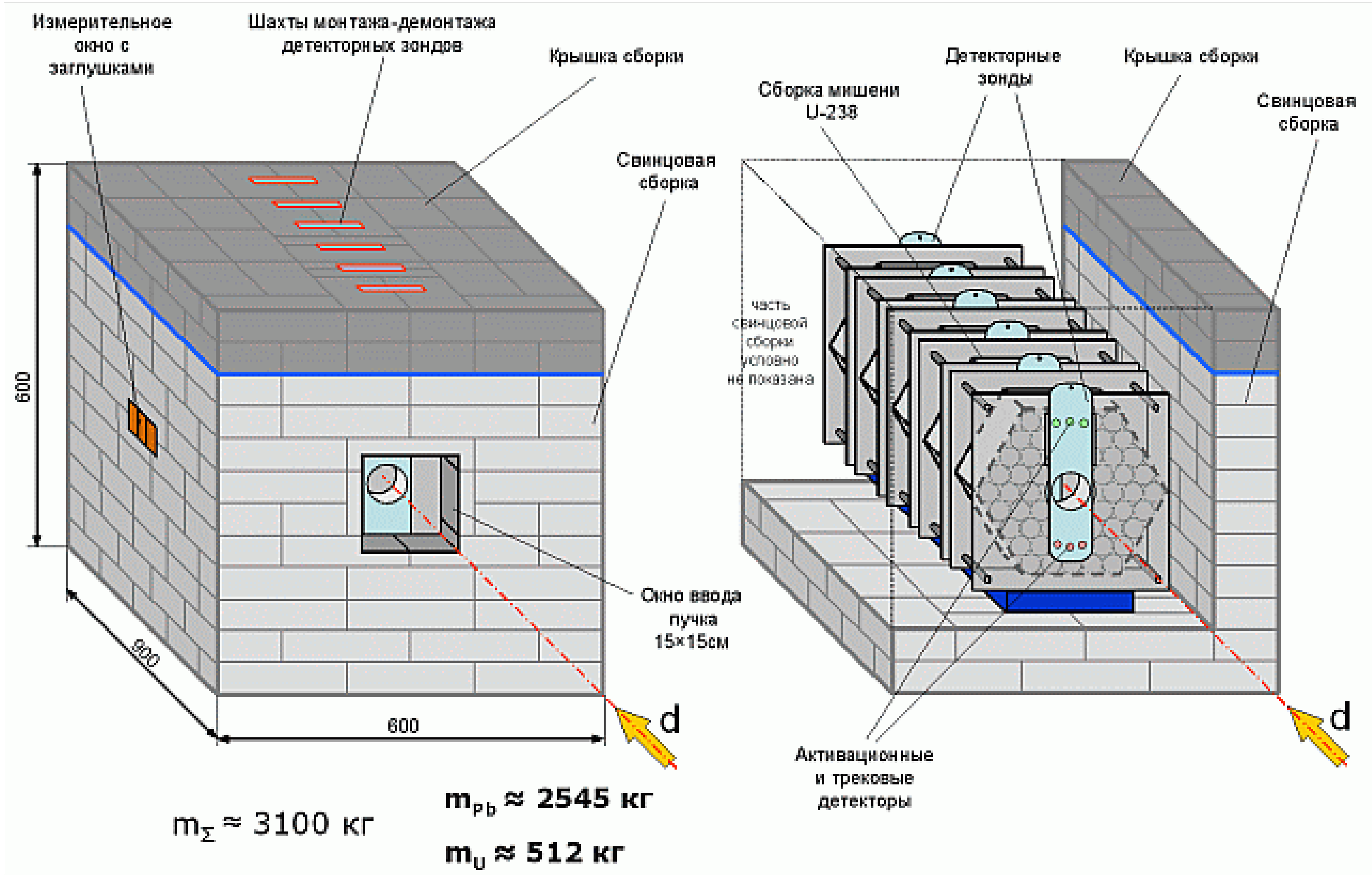
Rear view photo



Target assembly “Quinta” at the irradiation position (March 2011)



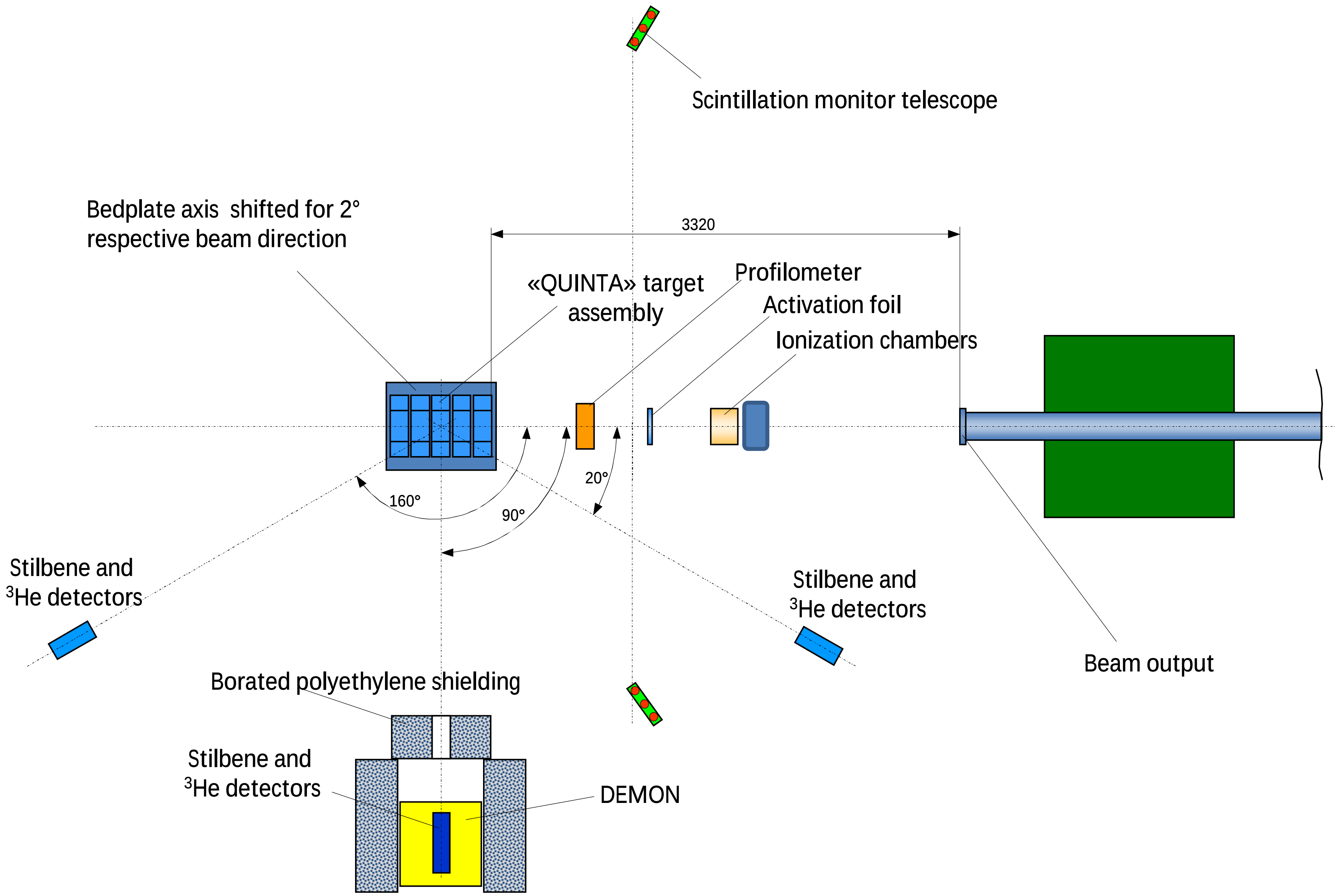
TA QUINTA with lead blanket



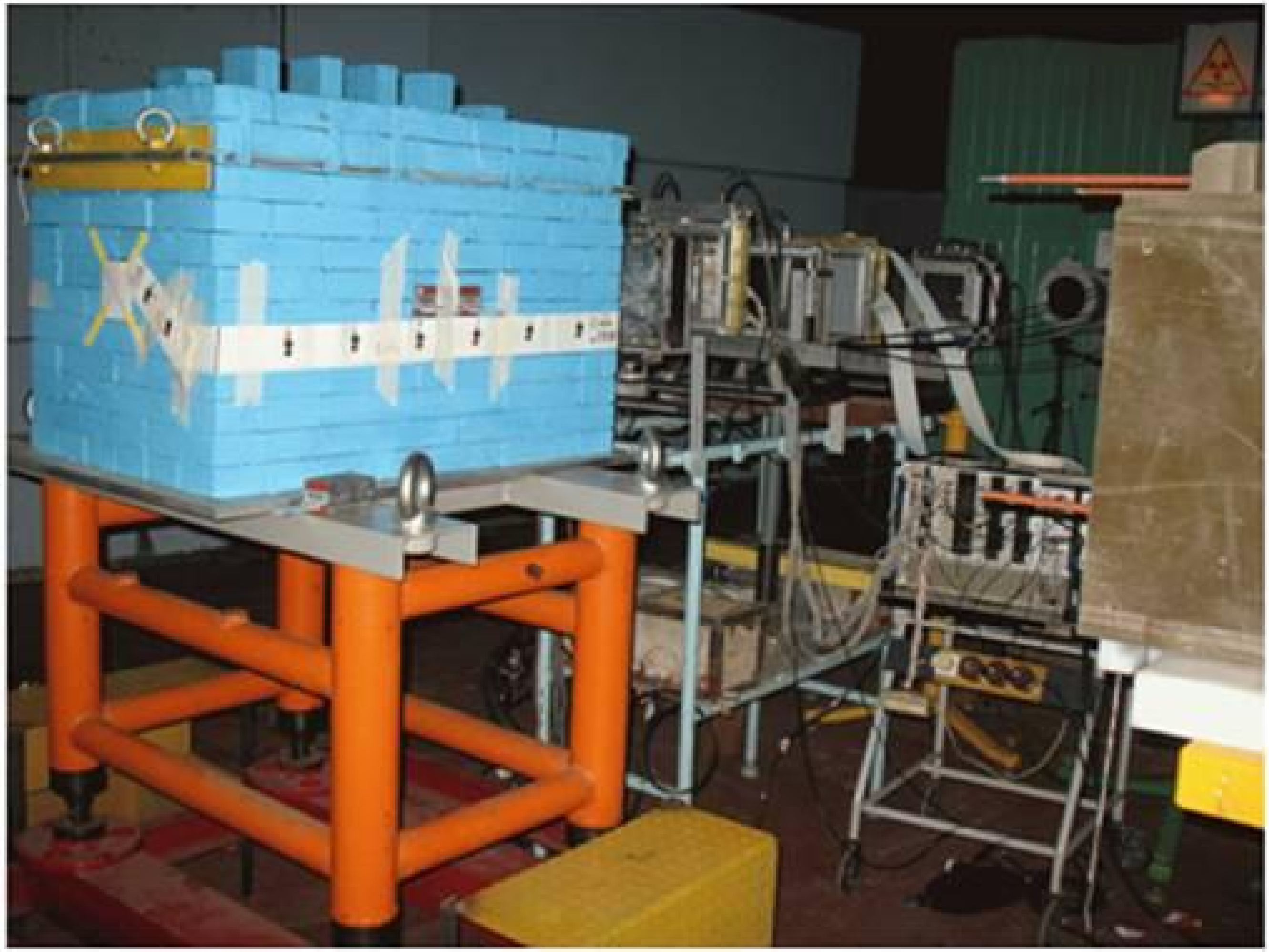
Results of experiments 2011-2013

- *The main results of measurements carried out with TA QUINTA during 2011-2013 and obtained data analysis will be presented below*

Scheme of experiment with “QUINTA” setup

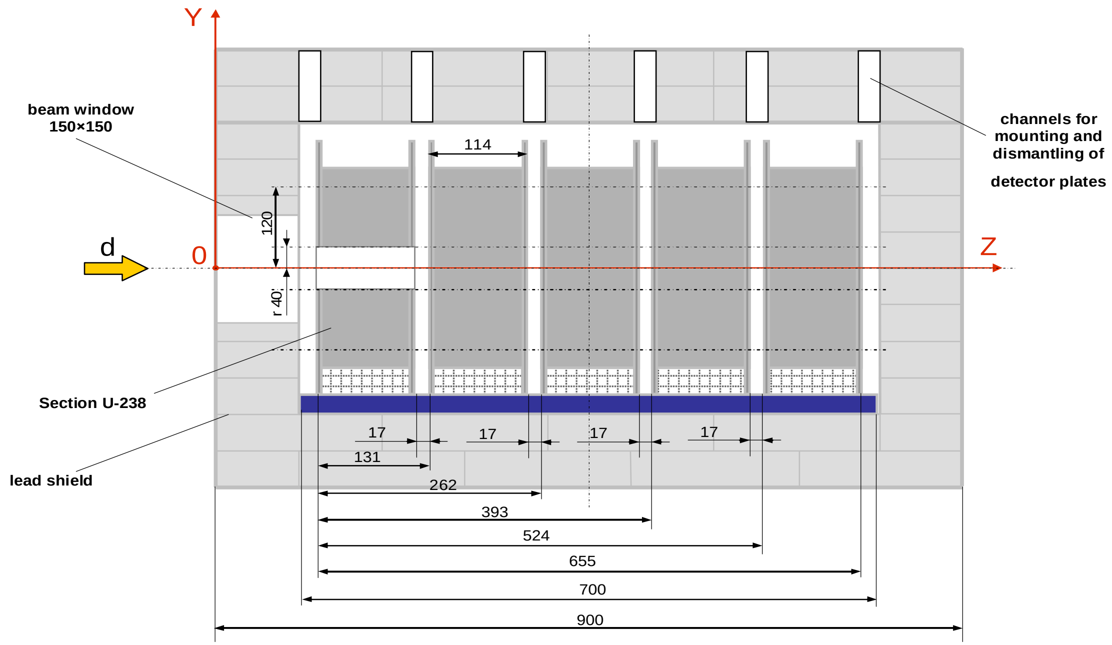


TA QUINTA ON IRRADIATION POSITION (March 2013)



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) $\beta- \rightarrow {}^{239}Np$ (2,36 days) $\beta- \rightarrow {}^{239}Pu$

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.

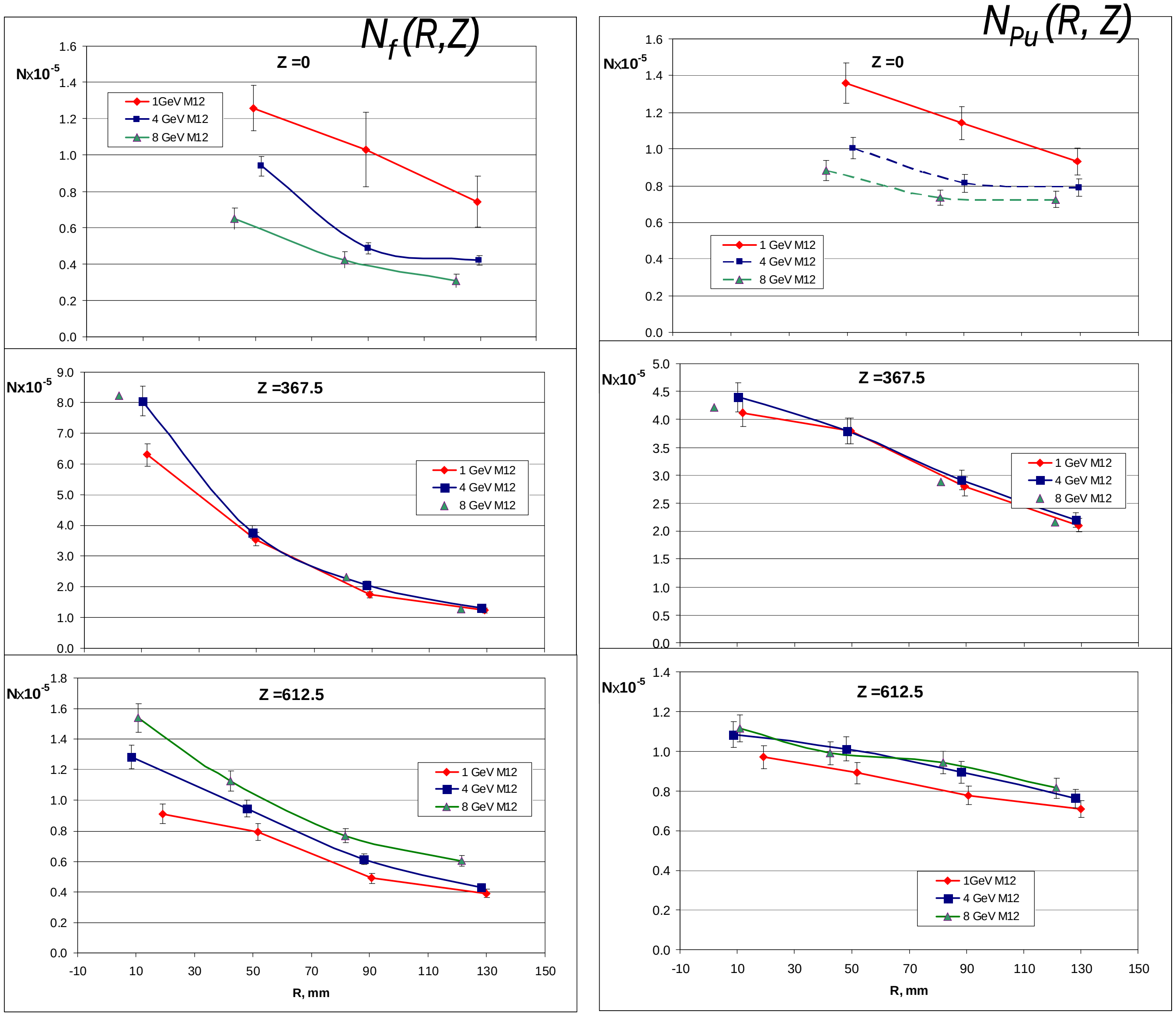
Number of fissions in the place of measurements defines by averaging of following fission product yields:

^{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*

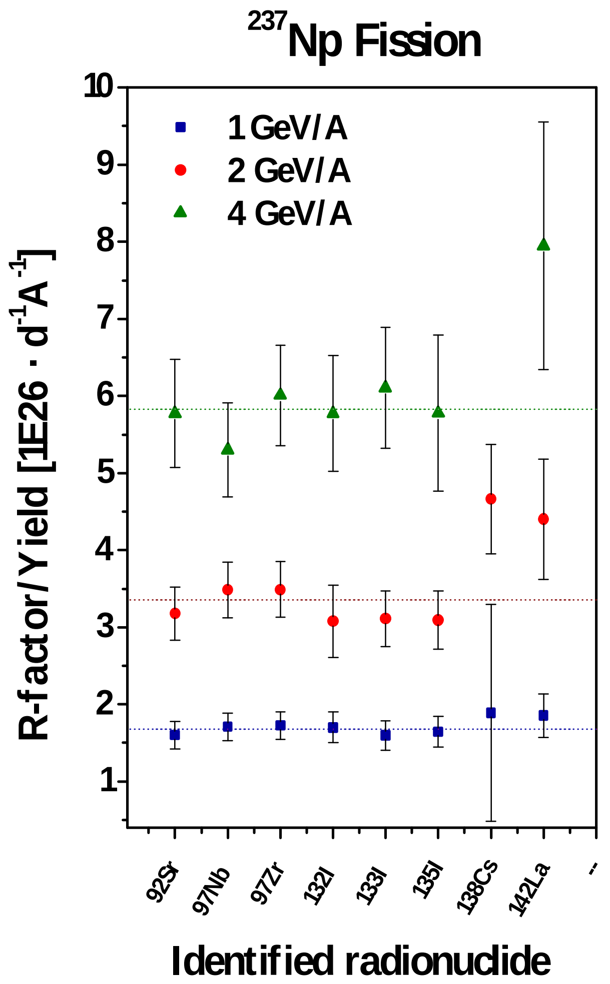
Total numbers of fission N_f(tot) and produced ^{239}Pu nuclei ®

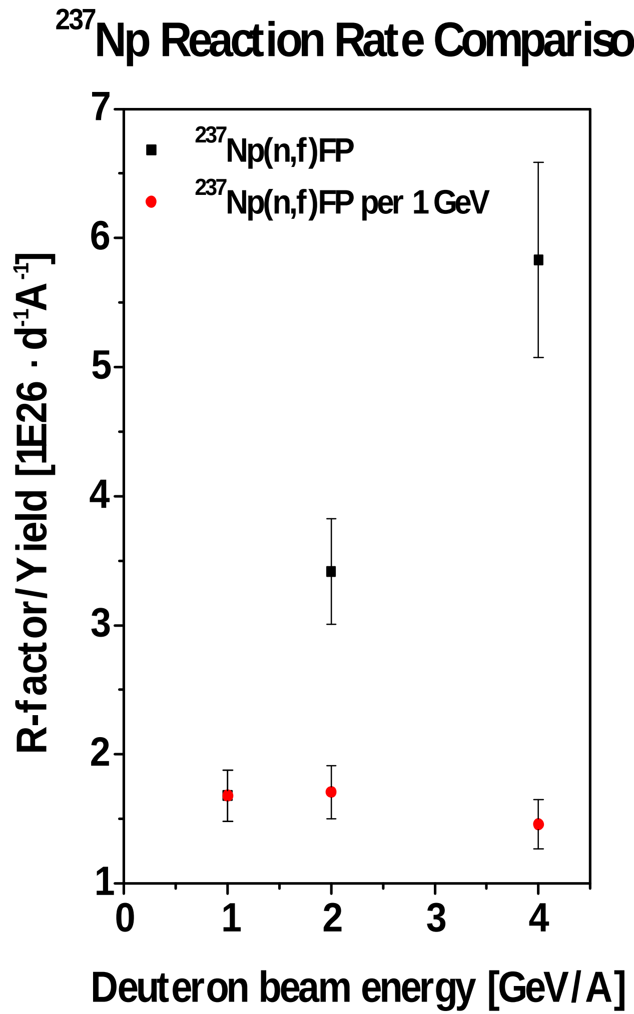
experiment v.s. calculation

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

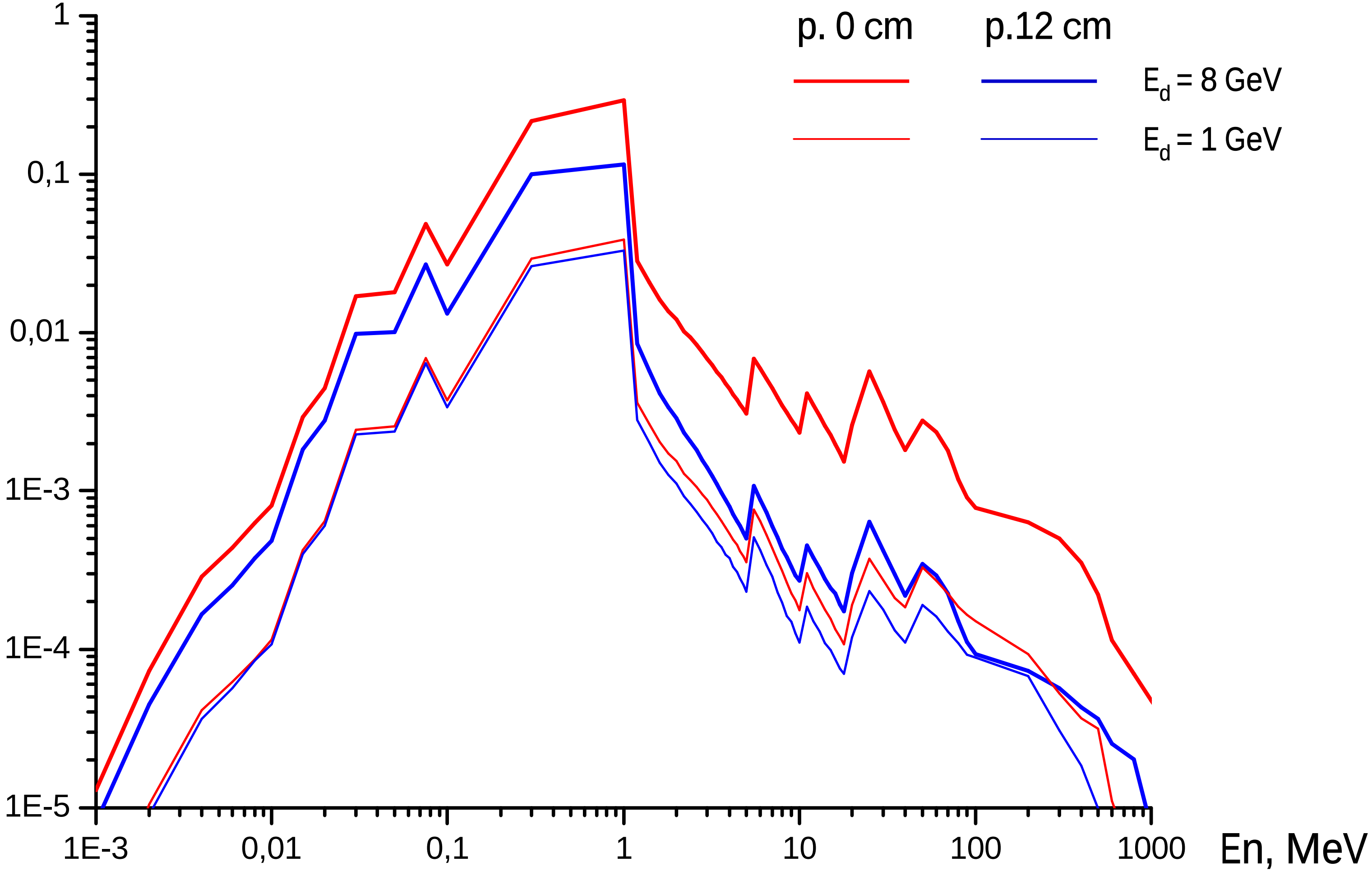
Reaction products	Ed = 2 GeV Reza et al.	Ed = 4 GeV Adam et al.	Ed = 6 GeV Adam et al.
Th(n,g)**Pa-233**	76.9(39)	142(4)	176(3)
Th(n,2n)**Th-231**		51.4(15)	71.2(23)
Th(n,6n)**Th-227**		3.8(15)	4.4(4)
Th(n,p6n)**Ac-226**	1.13(8)	2.98(21)	3.41(11)
Th(n,p8n)**Ac-224**	0.62(5)	1.37(6)	3.1(3)
Th(n,**fission**)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

Incineration of the threshold minor actinide from SNF
example of ^{237}Np

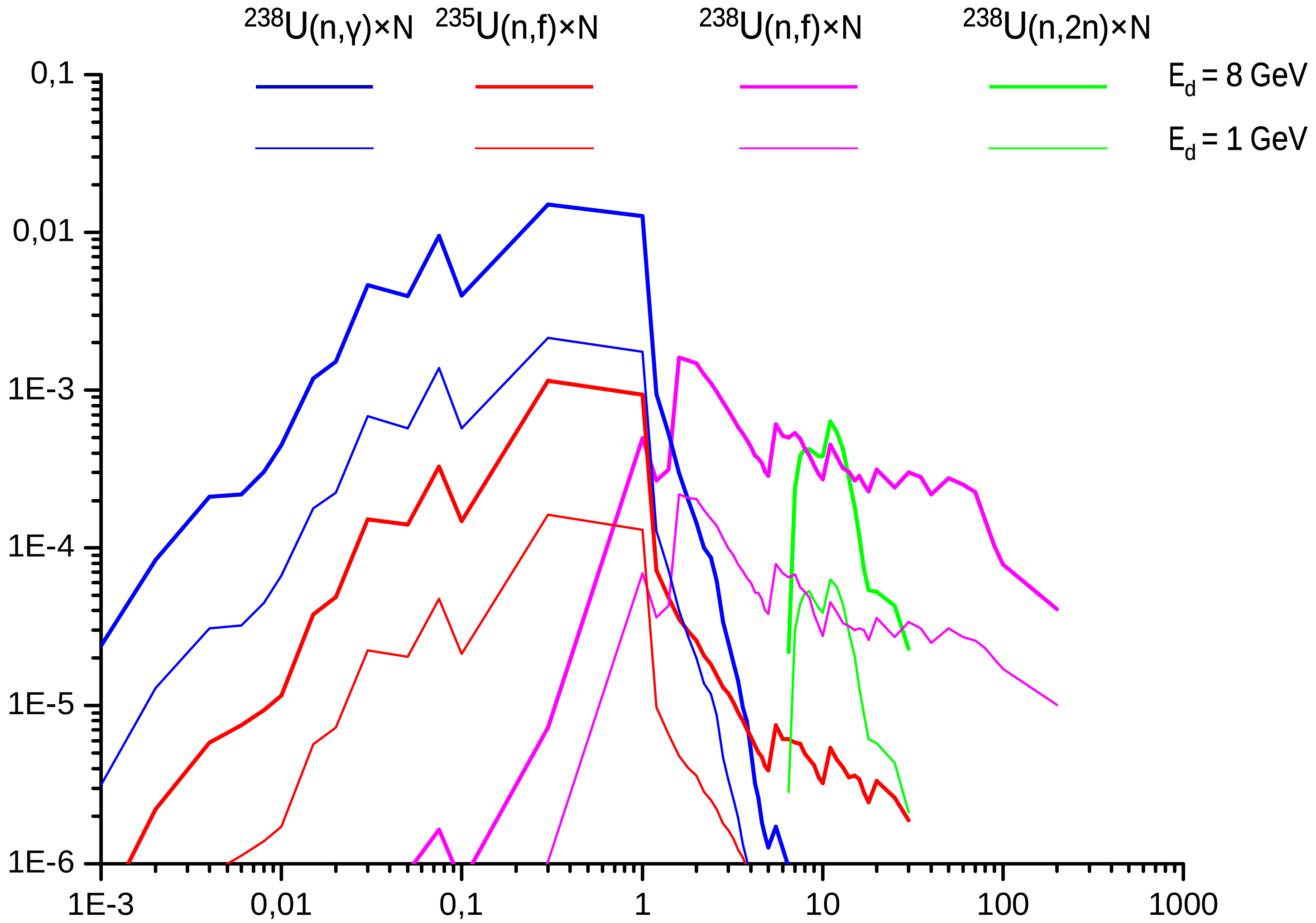




Typical calculated neutron spectra ($n/d/\Delta E_n$) at positions R= 0 and 12 cm, Z=64,5 cm (fourth plate)

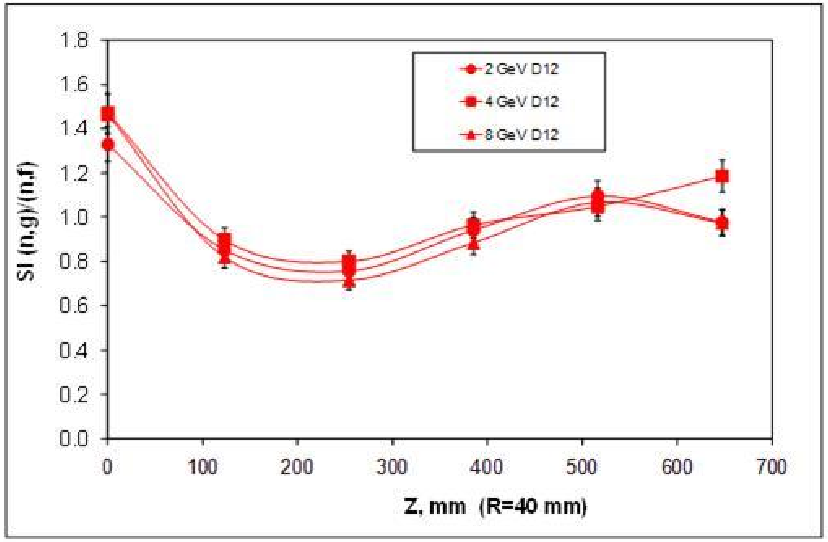


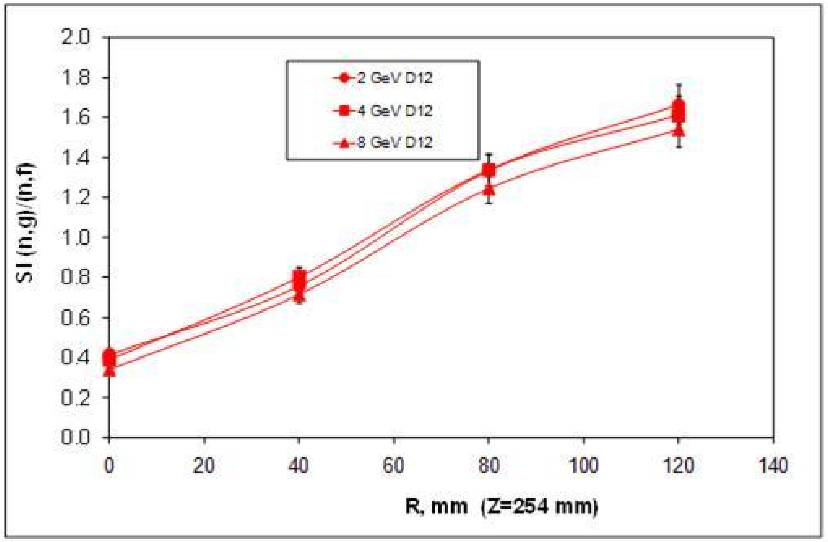
Convolution of cross-sections (n,γ), (n,f) and (n,2n) with neutron spectra at position R=12 cm in the fourth plate



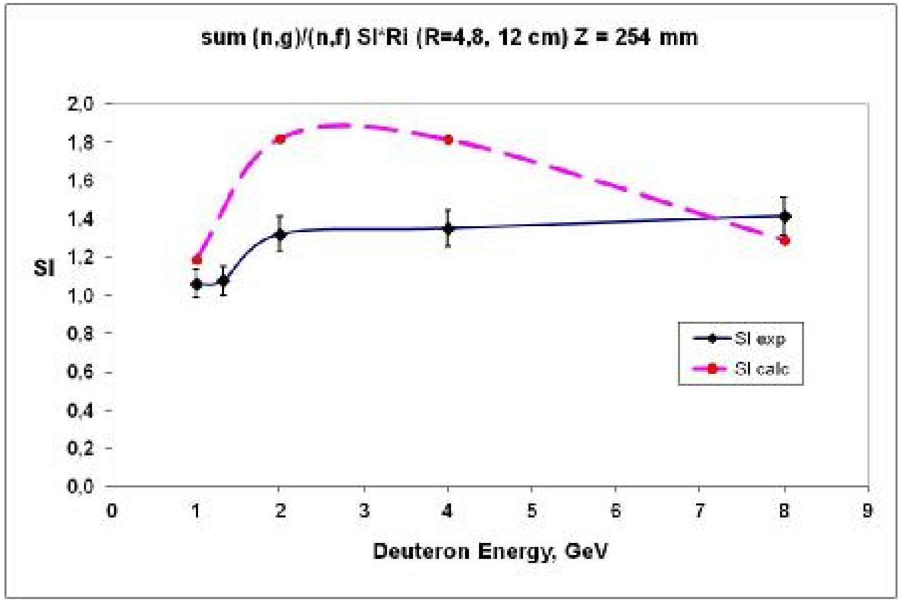
Spatial distributions of spectral indices SI (n,γ)/(n,f)

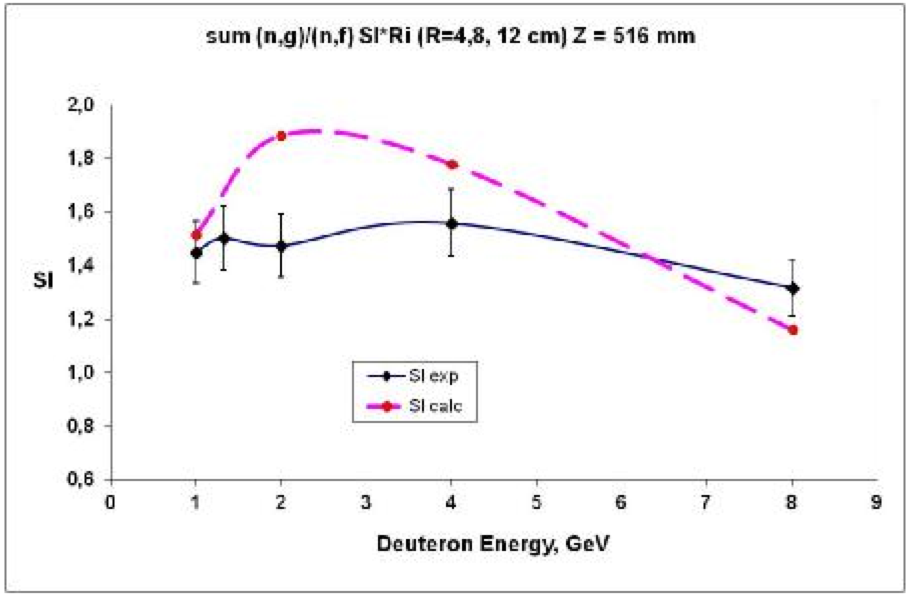
- *Spectral indices are relative values. Their uncertainties are much smaller than for each $N_{(n,2n)}(R,Z)$, $N_f(R,Z)$ or $N_g(R,Z)$ values. SI do not depend on beam intensity normalization*
- *Neutron spectrum becomes "softer" with increase the distance from beam axis*





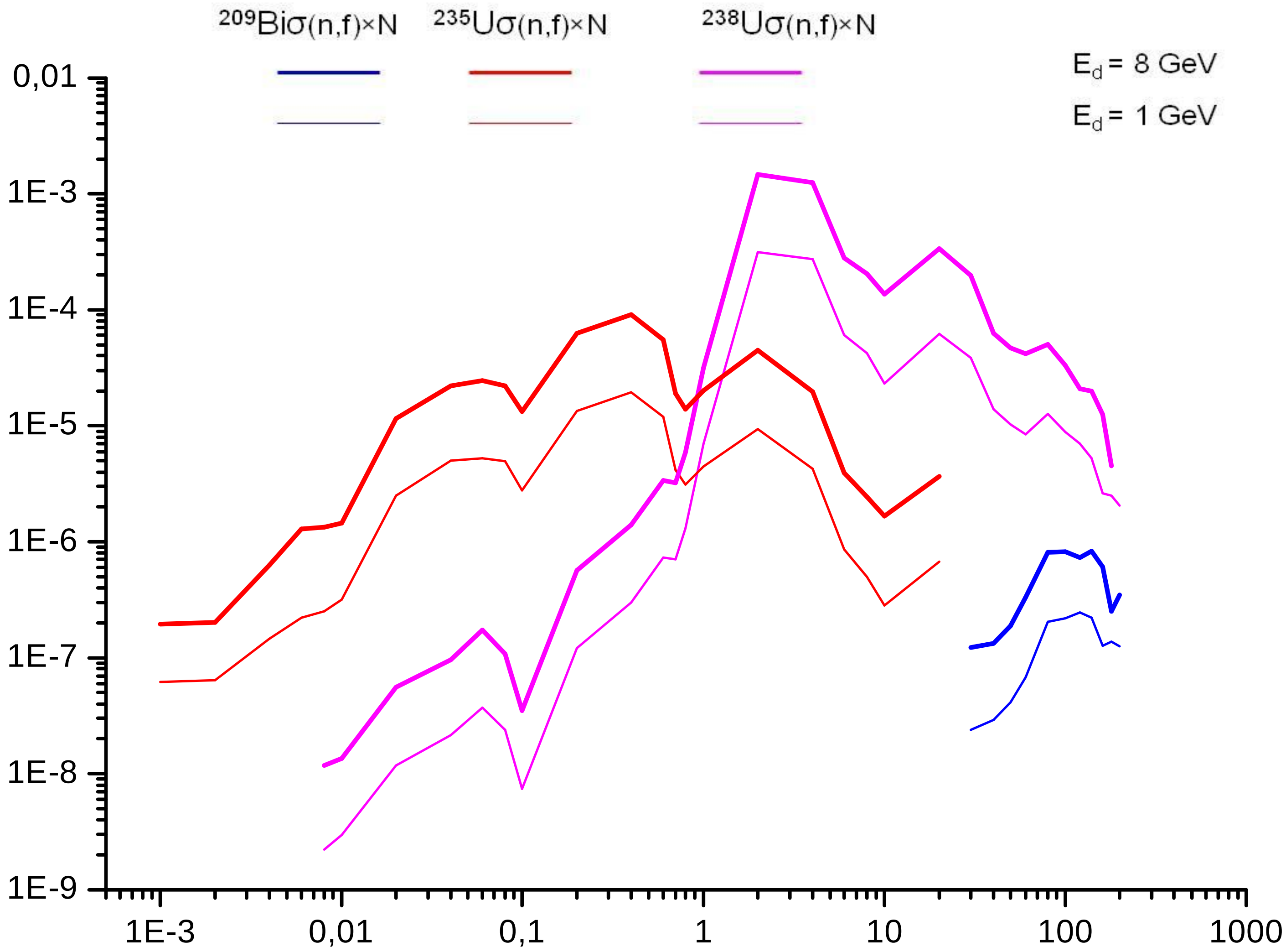
Comparison of experimental and calculated spectral indices SI (n,γ)/(n,f) in dependence on deuteron energy



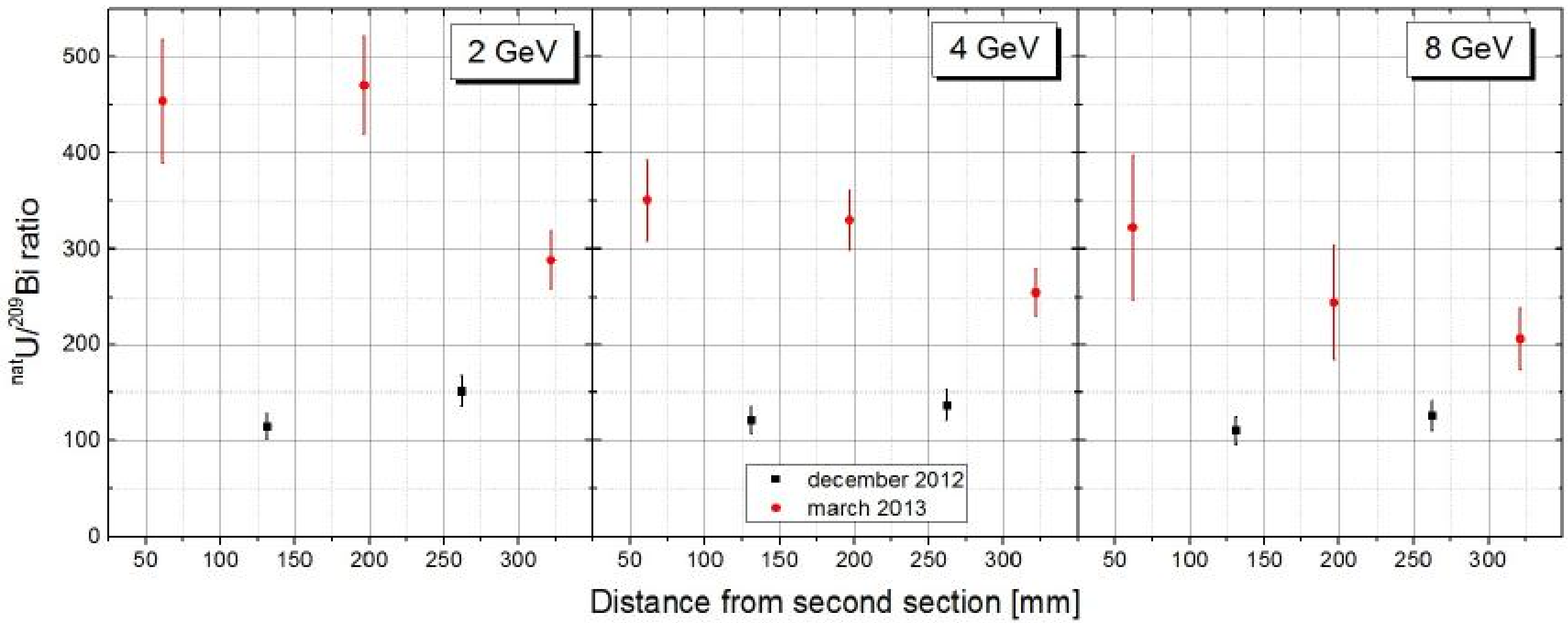


- *Share ratios of the neutron spectrum in the energy ranges En < 1.2 MeV and 1.5 Mev < En< 20MeV remain approximately constant with increase of incident energy*
- But what about En > 20 MeV ?

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)

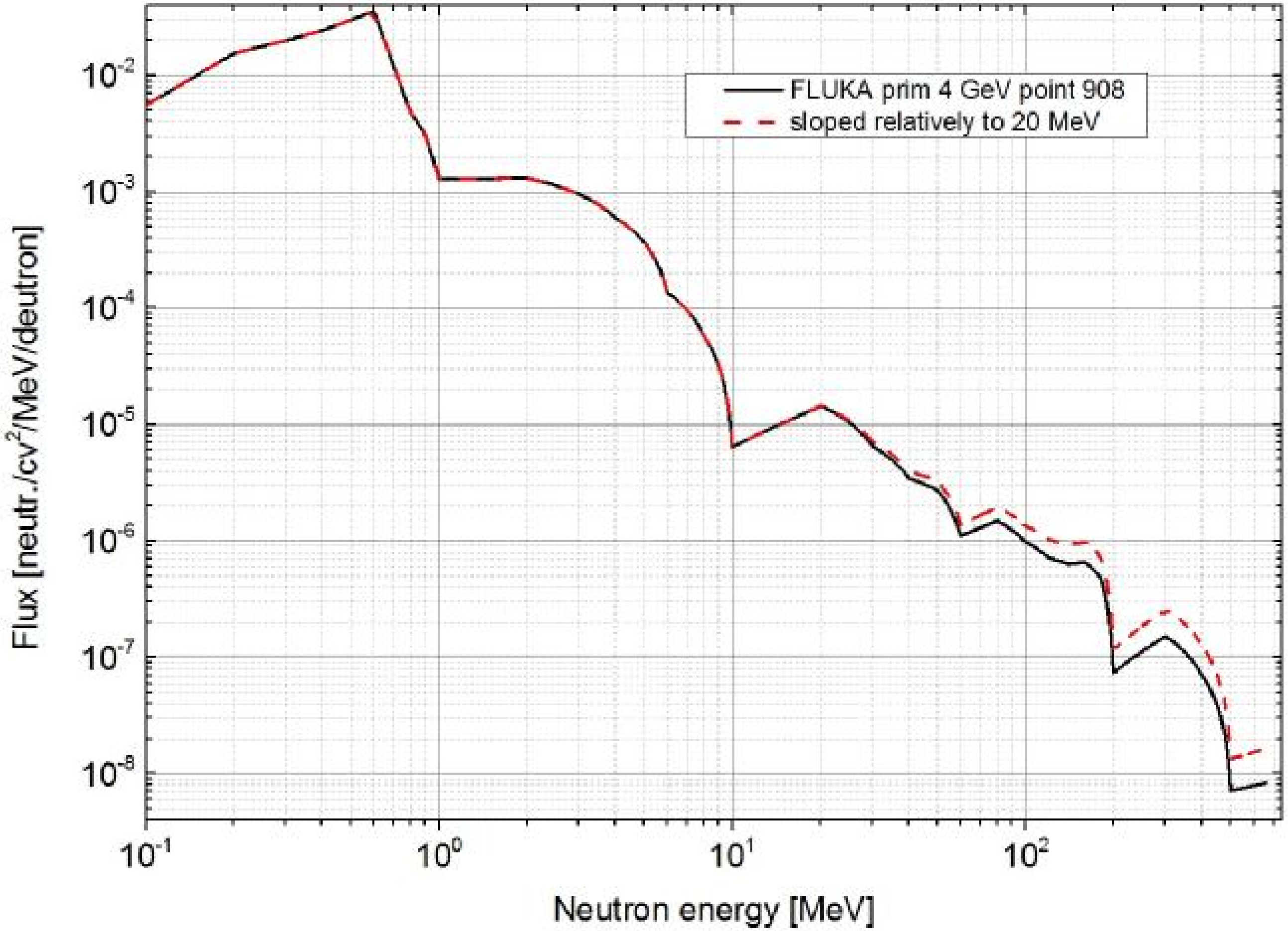


Ratios of $^{nat}U/^{209}Bi$ (n,f)-reaction rates
(black – inside of TA QUINTA, red – on the surface)



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-5, FNAL)	49,2	47,9	42,8	39,1
$N_{En>20\ MeV}$ (calc. MARS-5, 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

Beam power gain vs. size of AC

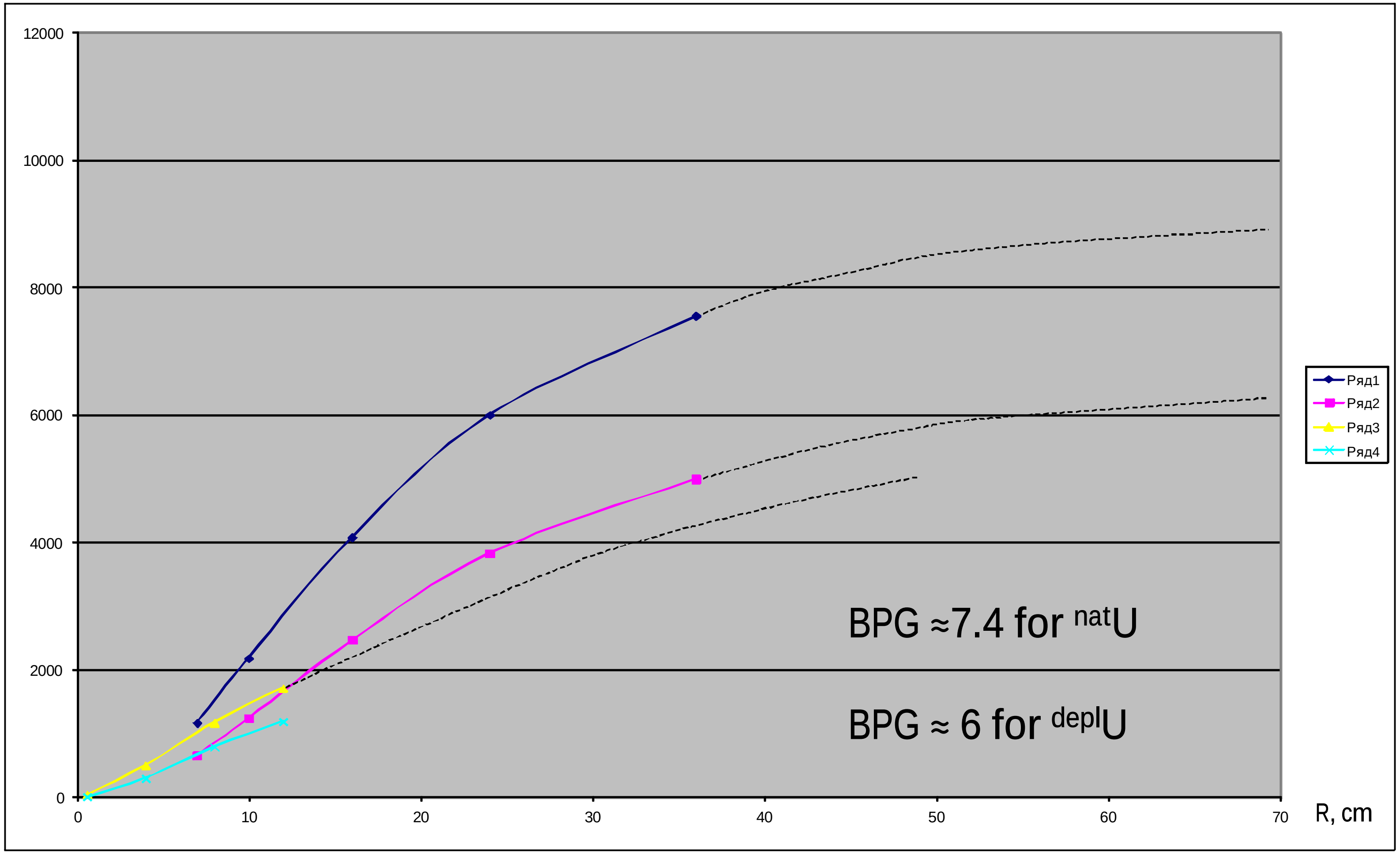
- *Beam power gain ($BPG = E_{release}/E_{beam}$) is key characteristics of any ADS. Its value depends mainly on the total number of fissions within AC and determines practical applicability of given ADS.*

- *Minimal value of BPG providing a zero energy balance of whole ADS operation is in better case 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 – 3.5 tons, neutron leakage ~ 10%).*

- *This result was not reproduced by any calculations – the difference with the experiment up to two times.*

- *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 (Vassil'kov&Goldansky result BPG ≈ 6).*

- *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,γ) 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 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*

<u>Conclusion</u>(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