RECENT RESULTS ON THE STUDY OF ADS WITH 500kg NATURAL URANIUM TARGET ASSEMBLY QUINTA IRRADIATED BY DEUTERONS WITH ENERGIES FROM 1 TO 8 GeV AT JINR NUCLOTRON

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on the behalf of

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Dubna

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Content

Introduction

- **The problems of global world power can not apparently solved without the contribution of atomic energy. But the development of traditional nuclear power rests on the lack of resources commonly used fuel (enriched uranium or plutonium) and the key problem of utilization of radioactive waste (RAW).**
- **Recent decades as a way to solve the RAW problem more seriously considered Accelerator Driven Systems (ADS).**
- **In Europe, China and India have adopted state ADS programs. The most striking example is the project MYRRHA, already implemented in Belgium**

- **At the last XX Baldin Seminar was presented the main features of new promising ADS project aiming to recycle spent nuclear fuel (SNF) with simultaneous producing energy.**

- **This project is based on so called Relativistic Nuclear Technology (RNT) proposed recently by one of the institutions (*CPTP «Atomenergomash», Moscow)* participating collaboration.**

Basis features of RNT

1. **Using the deep subcritical active core (AC) of natural (depleted) uranium or thorium without any moderator the size of which provides minimal leakage of neutrons (quasi-infinite AC) and allow one to obtain maximally hard neutron spectrum inside.**

2. **An increase in energy of incident particles up to 10 GeV**
   instead of 1 GeV as in the traditional ADS schemes.
   It permits to decrease the accelerator flux and alleviate the problem of heat removal

3. **Using as a target for incident beam the material of AC**

4. **Application as a load of AC encupsulated fuel elements from natural (depleted) uranium or thorium, as well as from spent nuclear fuel, without its preliminary radiochemical reprocessing**

5. **Using the technology of high temperature helium coolant for primary circuit to preserve hard neutron spectrum in AC**

- **As for any ADS a necessary and key element is creation and use of powerful (~ 10-20 MWt) accelerator with good enough duty cycle.**

- **It is proposed to utilize an original Russian technology BWLAC for creation of reliable and compact MWt linac.**

- **All RNT engineering problems including creation of necessary accelerator have to be discussed practically after detailed study and verification of basic physics ideas of proposed approach.**

- ***This is just an aim of JINR project "Energy and Transmutation Radioactive Waiste" ("E&T RAW") adopted for realization during 2011-2013 on the basis deuteron and proton beams of JINR NUCLOTRON in incident energy range 1- 10 GeV/nucleon and available at JINR natural (depleted) massive uranium targets.***

Motivation

- ***Main goals of E&T – RAW project are to study basis characteristics of neutron fields inside quasi-infinite AC, spatial distributions of fission of core nuclei and 239Pu production and reaction rates of long lived minor actinides and fission products as well as to define optimal energy of incident beam for transmutation RAW and energy production.***

It is appropriate to note that quasi-infinite subcritical AC from natural or depleted uranium were studied earlier:

J.W. Weile et al.~19.3 tons nat. U+14 Mev neutrons (1961)
Wang Dalun ~20 tons depl.U+14 MeV neutrons (1991)
R.Vassil'kov et al. ~ 3.5(7) tons nat. U+660 MeV protons (1963-1969) → (1978)
C. Rubbia et al. ~3.6 tons nat.U+0.6-2.7 GeV protons (1995)

Only in three first experiments there was not any moderators and was realized hard neutron spectrum

J. W. Weale et al.1961

dT 14 MeV neutrons + 19.3 tons of natural uranium target

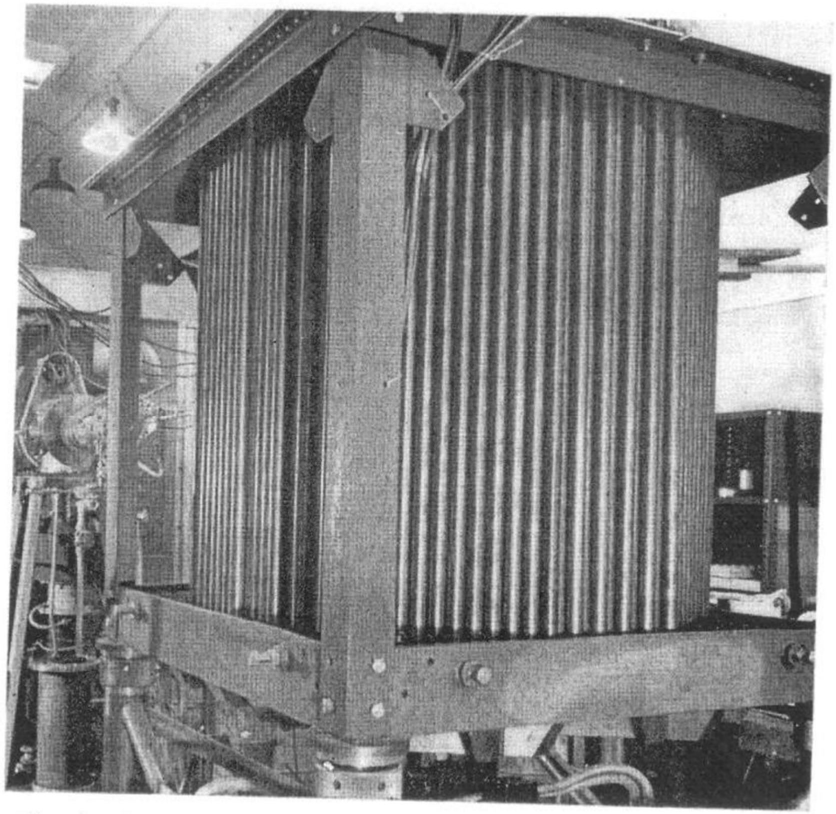


FIG. 2.—General view of the assembly showing the 39 inch diameter pile.

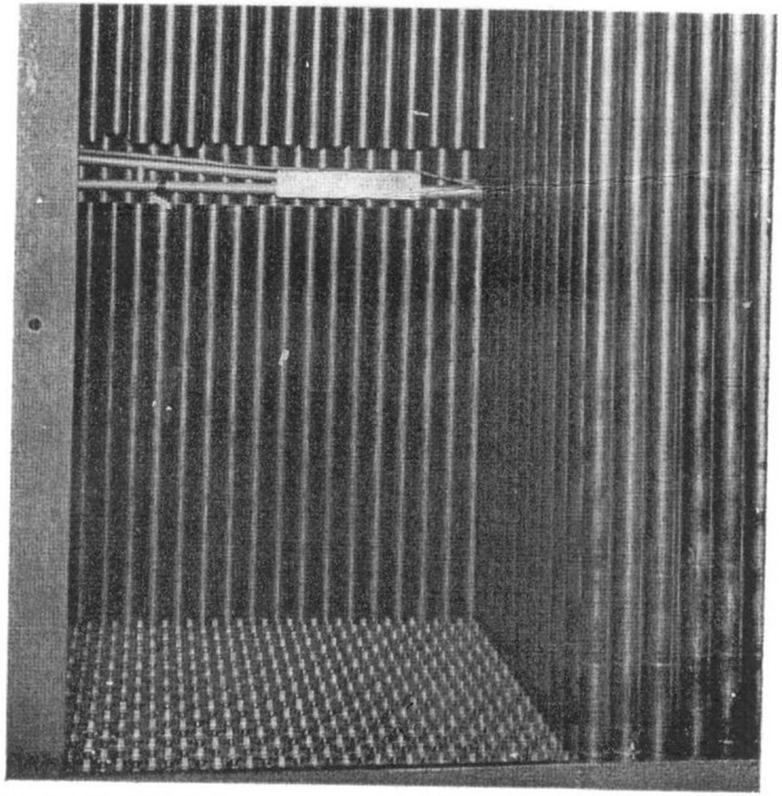
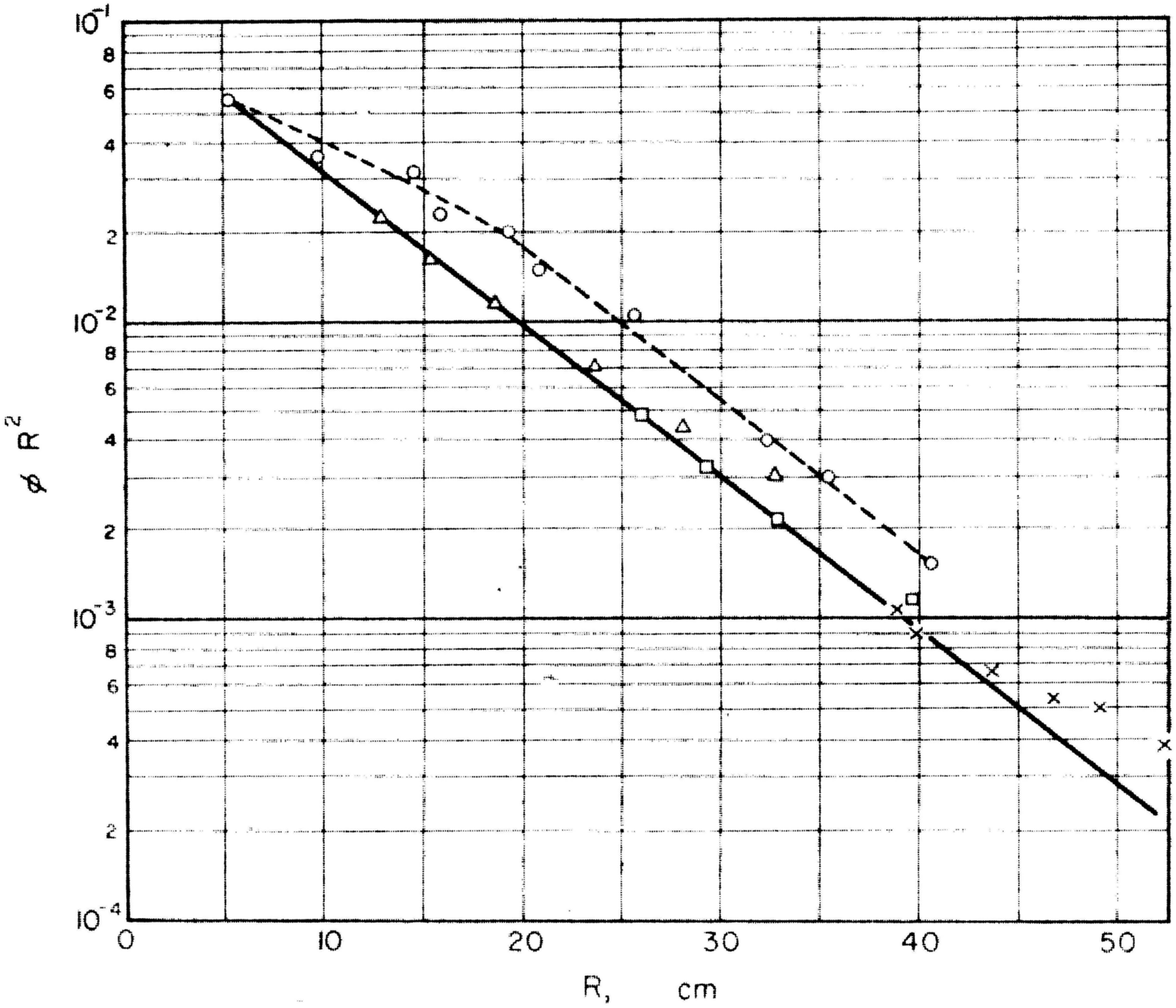


FIG. 3.—The 39 inch diameter pile with one-quarter of the rods removed to show the accelerator drift tube and the target arrangement.

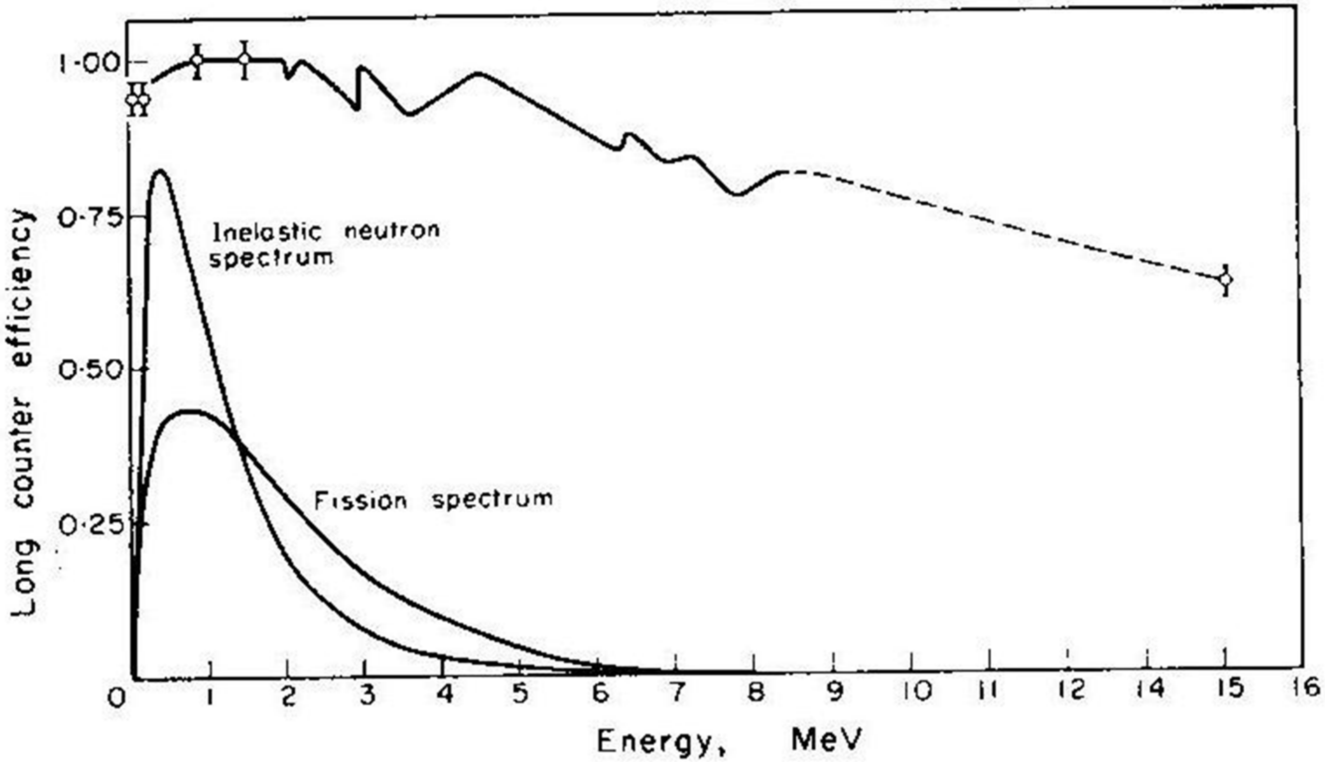
- H = 107 cm, ø = 99 cm, ρ = 16.3 g/cm^3
- Measured integral neutron leakage ~ 40%



Distribution of 14 MeV neutron flux inside of Weale pile

Long neutron counter efficiency as a function of its energy used by J. Weale et al

FIG. 1.—Long counter efficiency as a function of neutron energy. The inelastic neutron spectrum is taken from VASIL'EV *et al.* (1957) and ZAMYATNIN *et al.* (1959) and the fission spectrum from CRANBERG *et al.* (1956).



From the paper of K.W. Allen et al (1961)

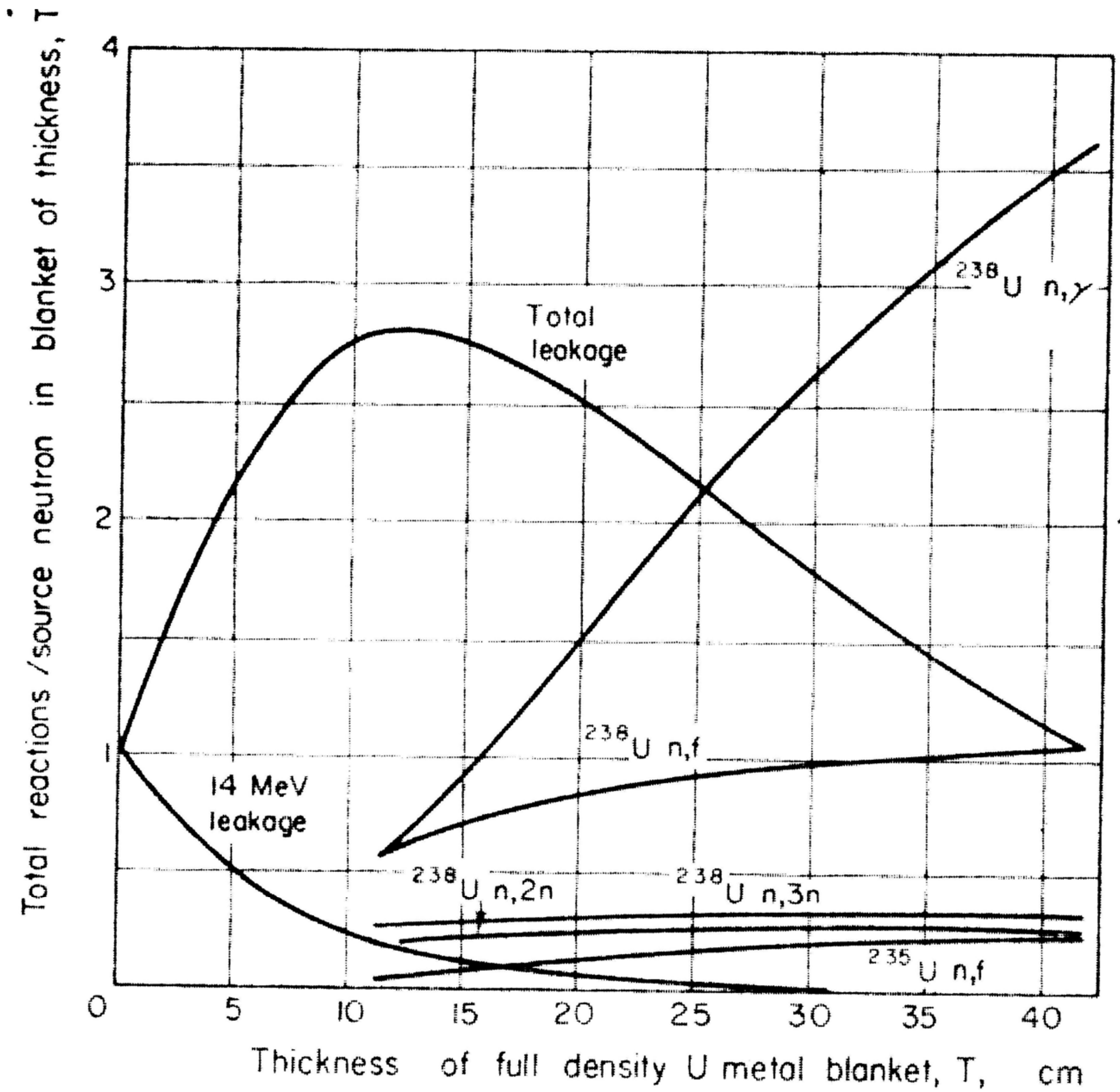


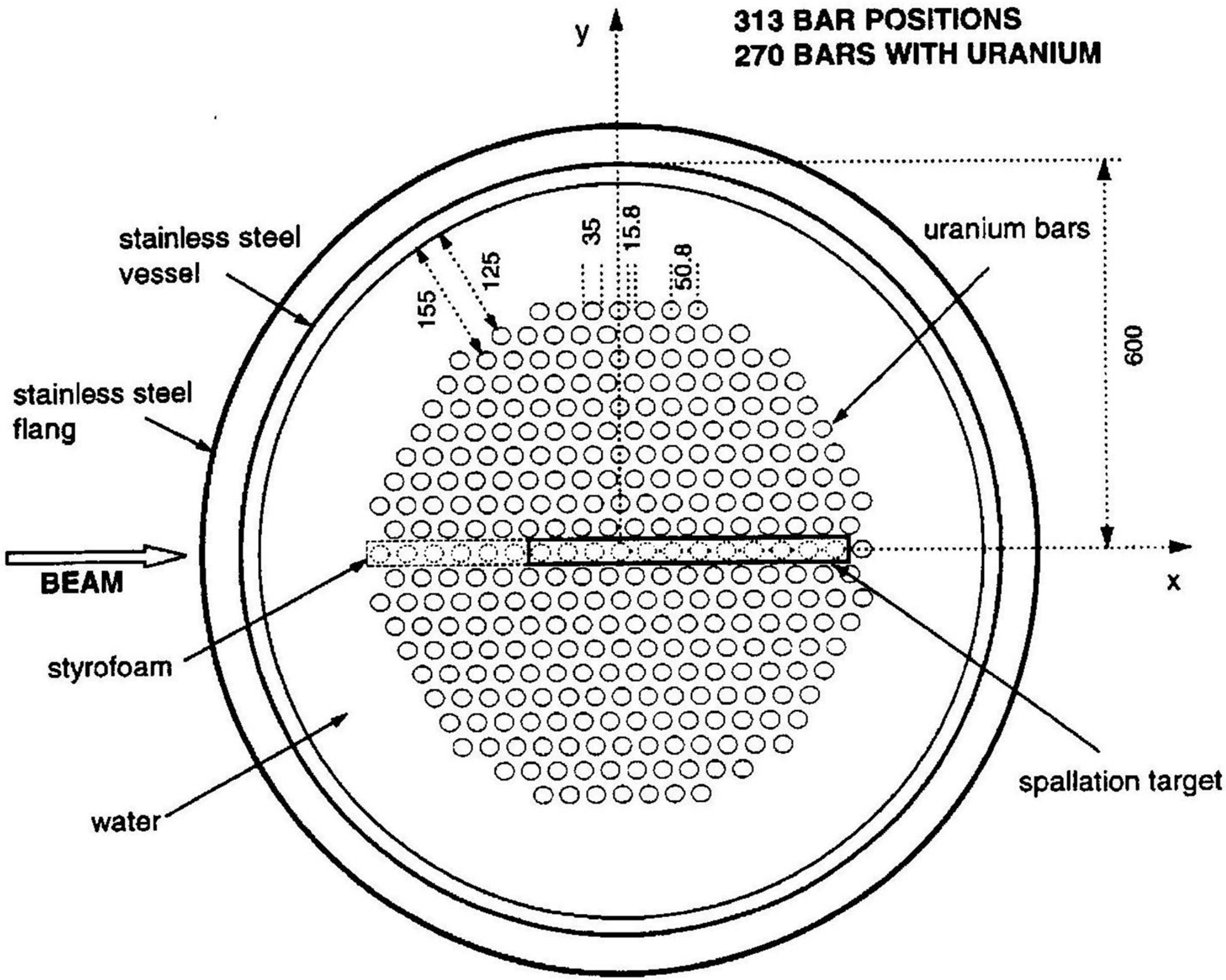
FIG. 11.—Total integrated neutron reactions and leakage per source neutron as a function of uranium blanket thickness. (Point 14 MeV central source and spherical geometry.) The curves for uranium thickness up to 11·3 cm are derived from (I).

By J.W. Weale et al
14 MeV neutrons central source surrounded by spherical uranium blankets thickness T

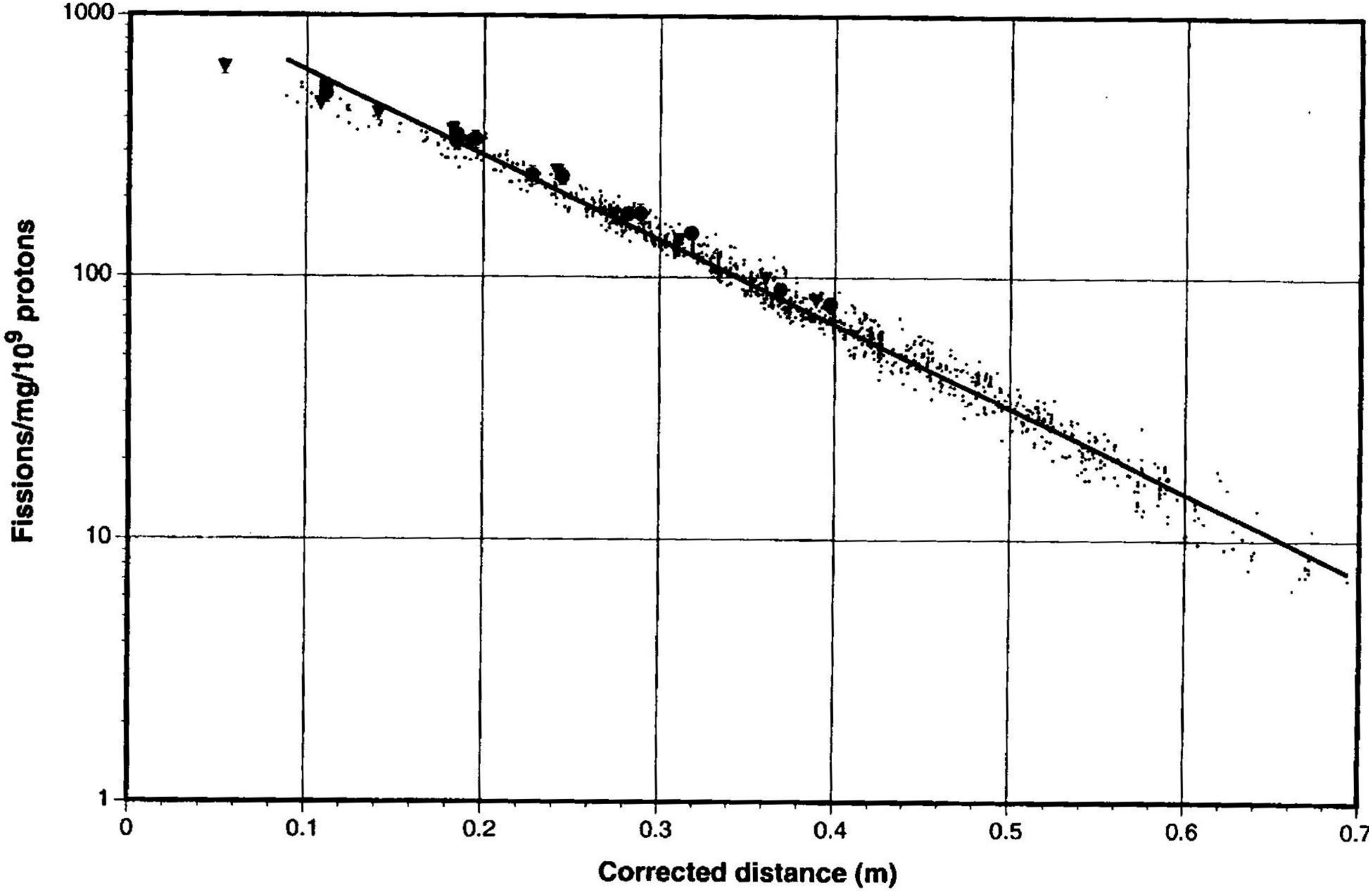
Total neutron leakage (41 ± 2) % (?!)

- Experiment of **Wang Dalun** (1991) has repeated a measurements of only N_f and N_{Pu} of J.W.Weale et al without description of any methodical details

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



Numbers of fissions as a function of distance to the source



Beam power gain (BPG) from FEAT experiment
(S. Andriamonje et al.,CERN/AT/94-45(ET))

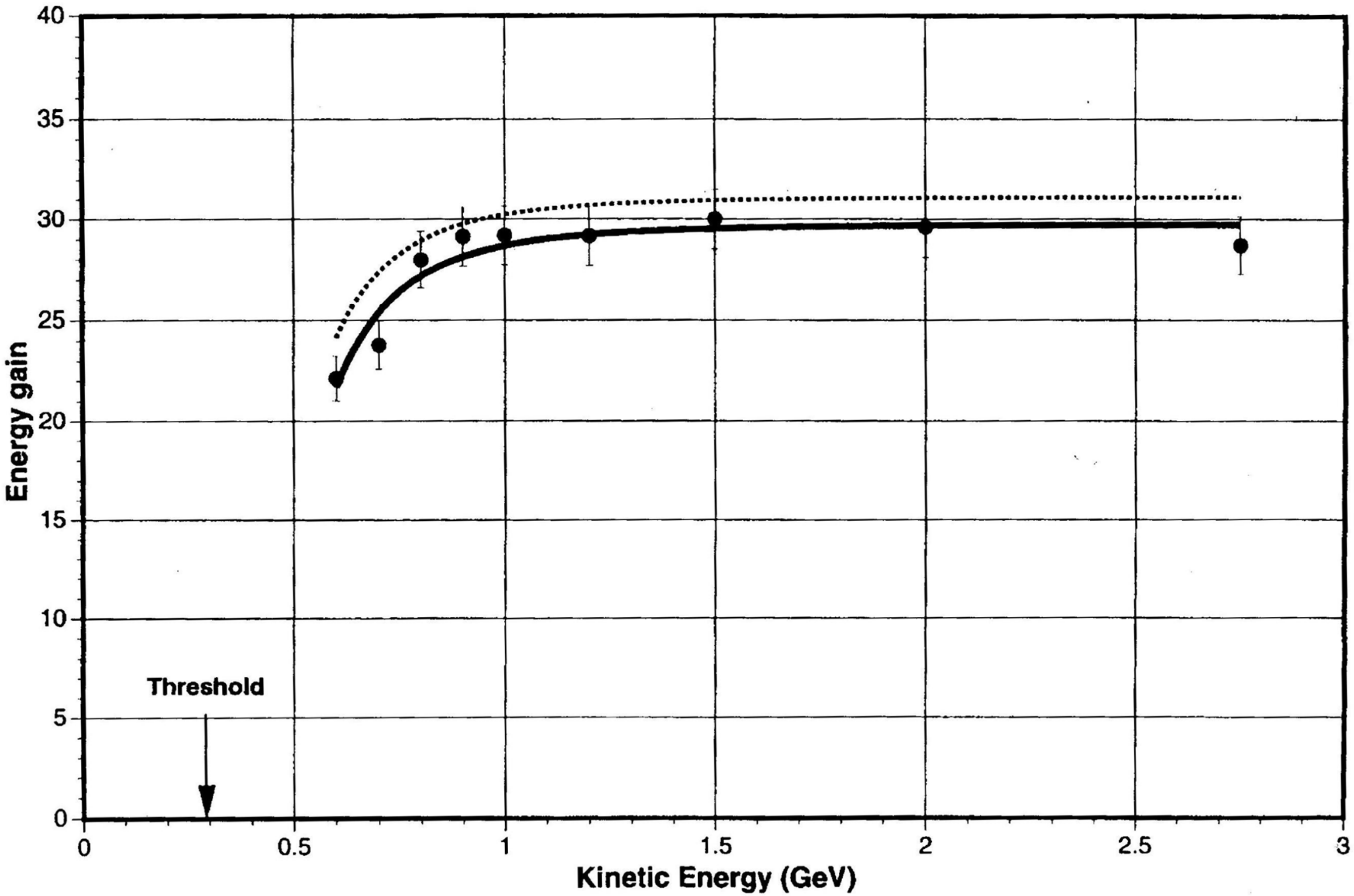


Figure 7

Problems of natural uranium and thorium use

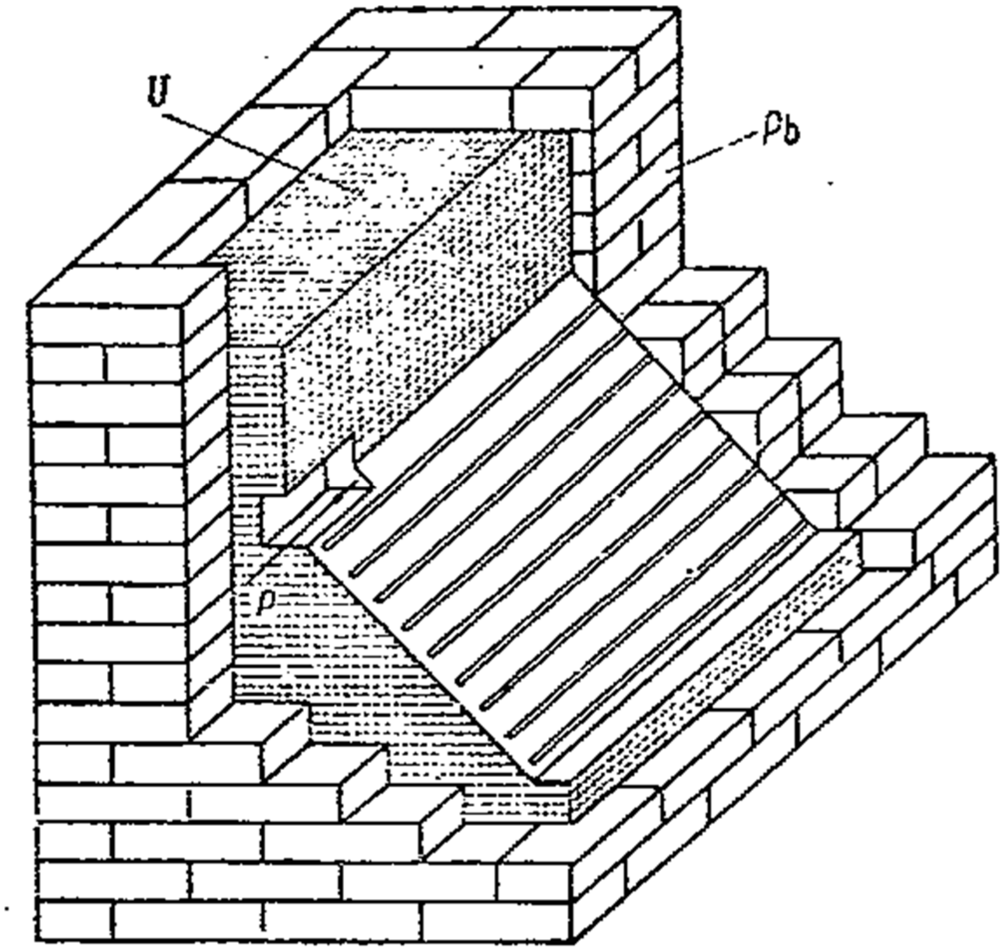
- In this experiment due to light water moderator within AC the neutron spectrum was practically fully thermalized and neutron multiplicity coefficient $\boldsymbol{k_{eff}}$ this system was near 0.9.

- In these circumstances in spite of rather promising BPG~30 it is difficult to implement "burning" of the base core material and any added minor actinides because of their high fission thresholds.

3500 kg 238U → 25 kg 235U → BPG !!!

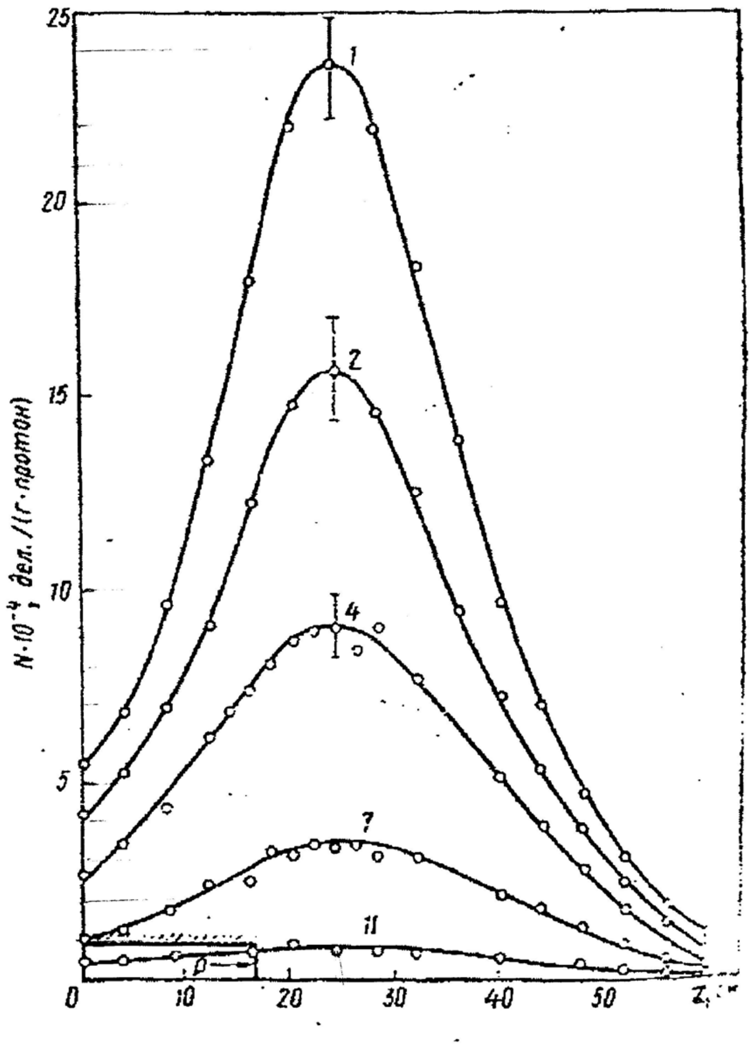
- And actually Energy Amplifier with liquid lead coolant proposed by K. Rubbia must move on to the enriched fuel !?

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***



Natural U

N_f = 18.5 ± 1.7   **BPG = 7.4 ± 0.7**

N_{Pu}= 46 ± 4

Depleted U

Nf = 13.7 ± 1.2   **BPG = 5.7 ± 0.5**

N_{Pu}= 38 ± 1.2

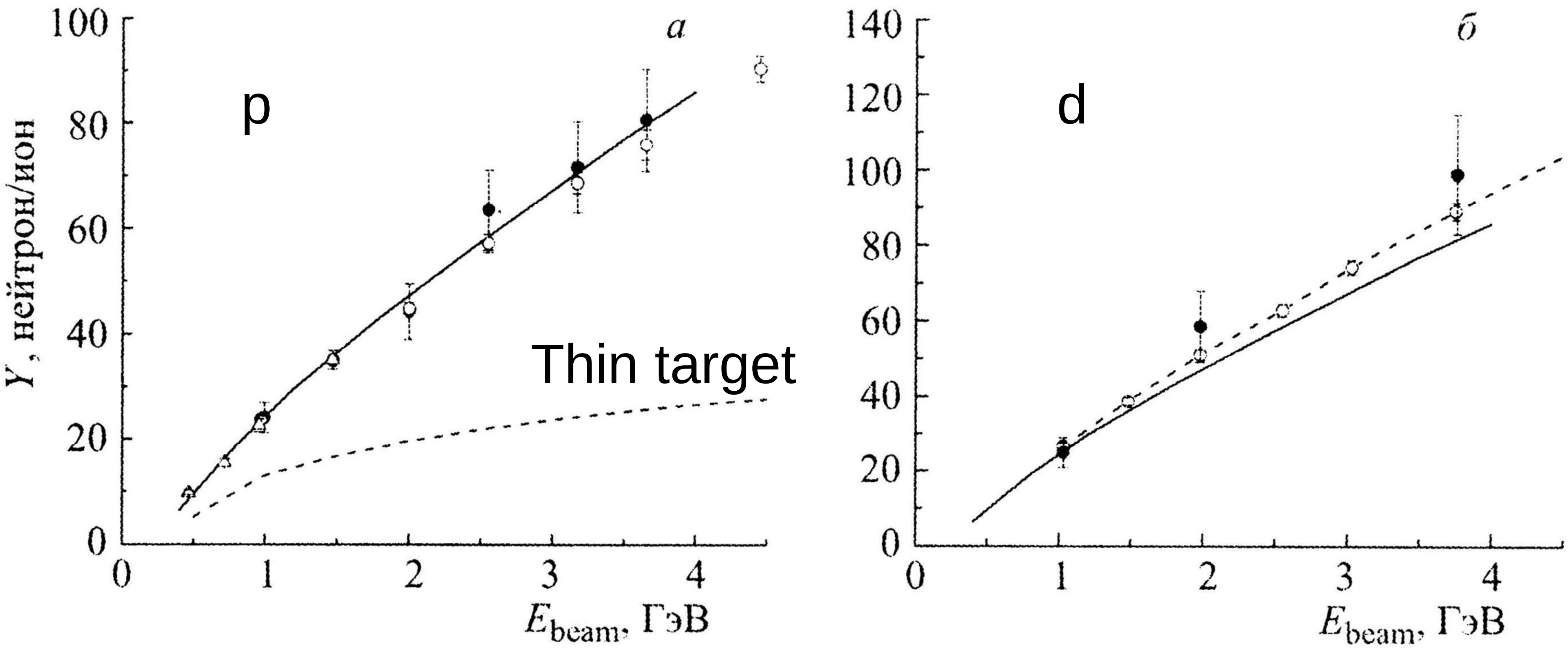
Ep = 0.66 GeV

N_f axial distributions for different radial channels

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

- ***What is known from irradiation of massive target for 1 to 5-10 GeV?***

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



For ø20x60cm lead target irradiated by GeV protons

from V. Yurevich et al (2006)

Eb GeV	<*En*> *MeV*	*En kin MeV*	*En kin/Eb* %	*W MeV*	*W/Eb* %
0.99	8.82	213	21.3	382	38.2
2.0	11.6	513	25.6	822	41.1
3.65	13.7	116	30.3	1670	45.6

W/Eb is a share of beam energy Eb expended in the formation of neutrons

For ø20x60cm lead target irradiated by GeV deuterons

from V. Yurevich et al (2006)

Eb GeV	*<En>* MeV	*En kin* MeV	*En kin/Eb* %	*W* MeV	*W/Eb* %
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

***W/Eb* is a share of beam energy *Eb* expended in the formation of neutrons**

Even for thick (ø20x60cm) lead target

- Mean neutron multiplicity grows at least linearly with the incident energy
- At the same time mean neutron energy $<En>$ and total kinetic energy neutron irradiation $E_{n\ kin}$ increase with growth of incident energy.
- Share W of this energy coming to neutron production approaches to 50% as well.
- *So for multiplicative (uranium or thorium) quasi-infinite target* it is plausible to expect a growth of total neutron multiplicity (and integral fission yield) with increase of incident energy .

- **The new approach to ADS outlined above is called the Relativistic Nuclear Technology (RNT) for energy production and utilization of spent nuclear fuel.**

- **All RNT engineering problems including creation of necessary accelerator have to be discussed practically after detailed study and verification of basic physics ideas of the proposed approach.**

- ***This is just an aim of JINR project "Energy and Transmutation Radioactive Waiste" ("E&T RAW") adopted for realization during 2011-2013 on the basis deuteron and proton beams of JINR NUCLOTRON in incident energy range 1- 10 GeV***

JINR PROJECT “Energy&Transmutation RAW”

- **For realization of “E&T RAW” project there are two target assemblies at JINR :**

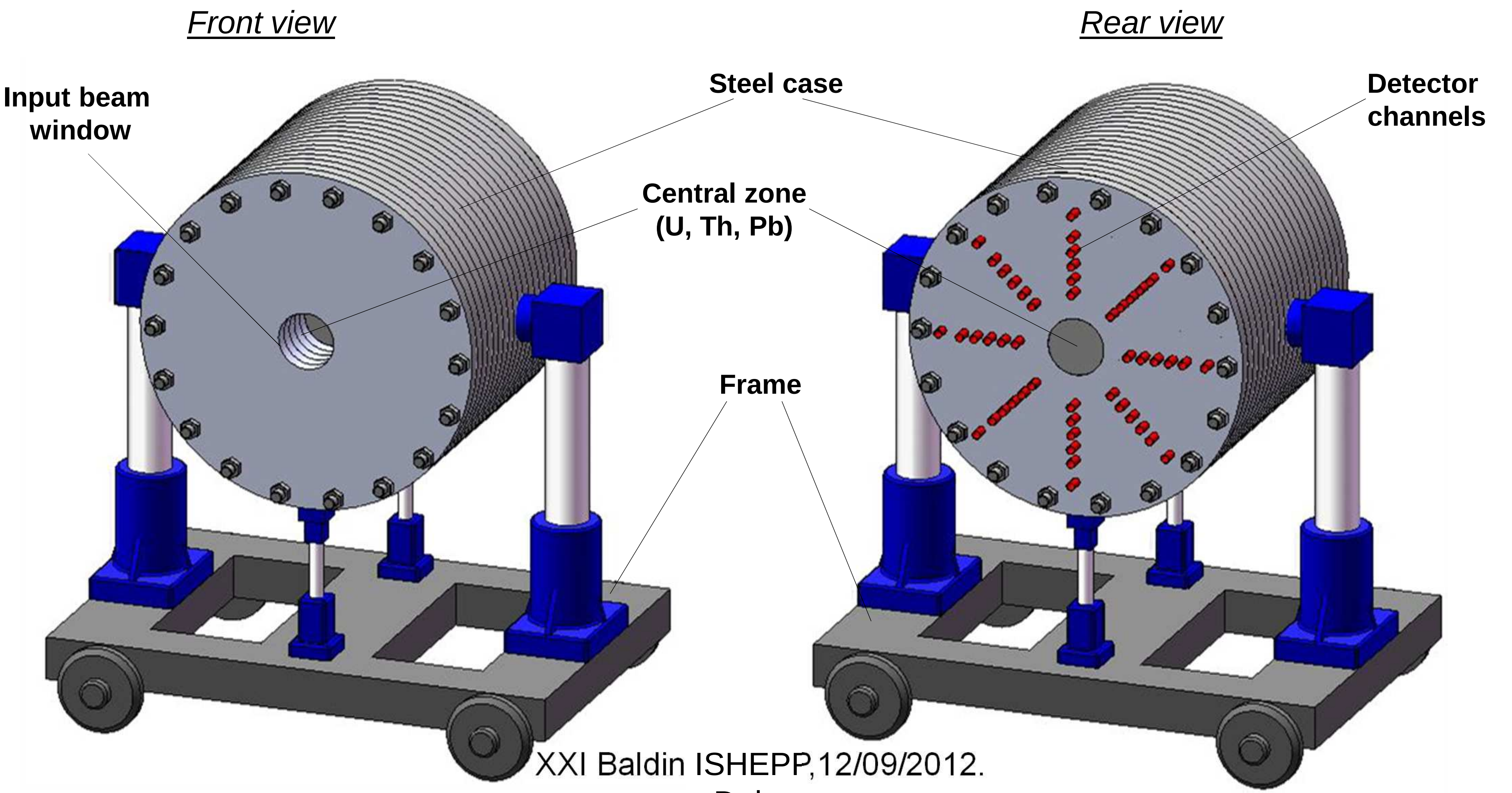
- semi-infinite (~20 tones) depleted uranium AC BURAN

- and smaller (~500kg) nat. U target assembly QUINTA modeling the central part of the larger AC

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

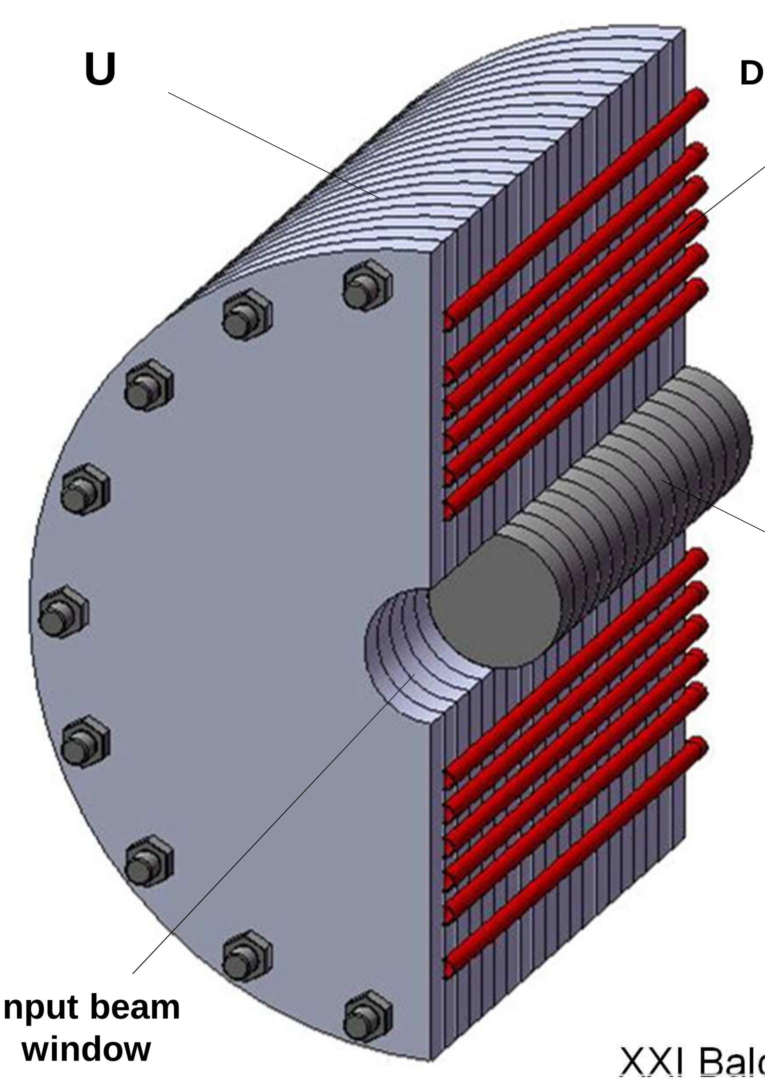
Mass of uranium – 19.5 т.
Diameter – 1,2 м.
Length – 1 м.

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

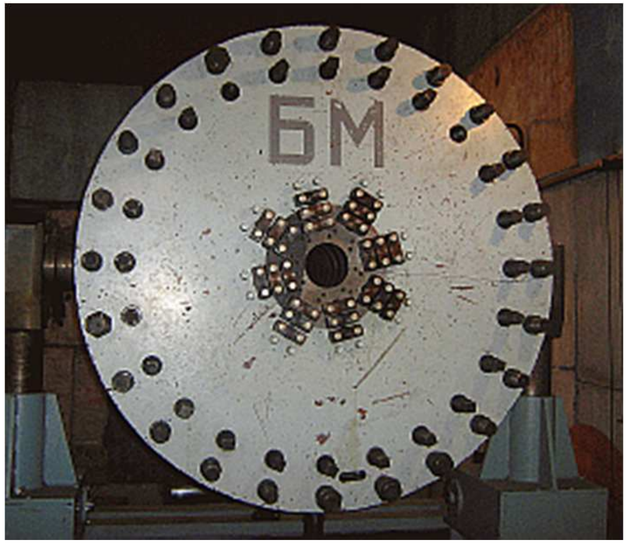


Quasi-infinite depleted uranium target (BURAN) with replacement central zone

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

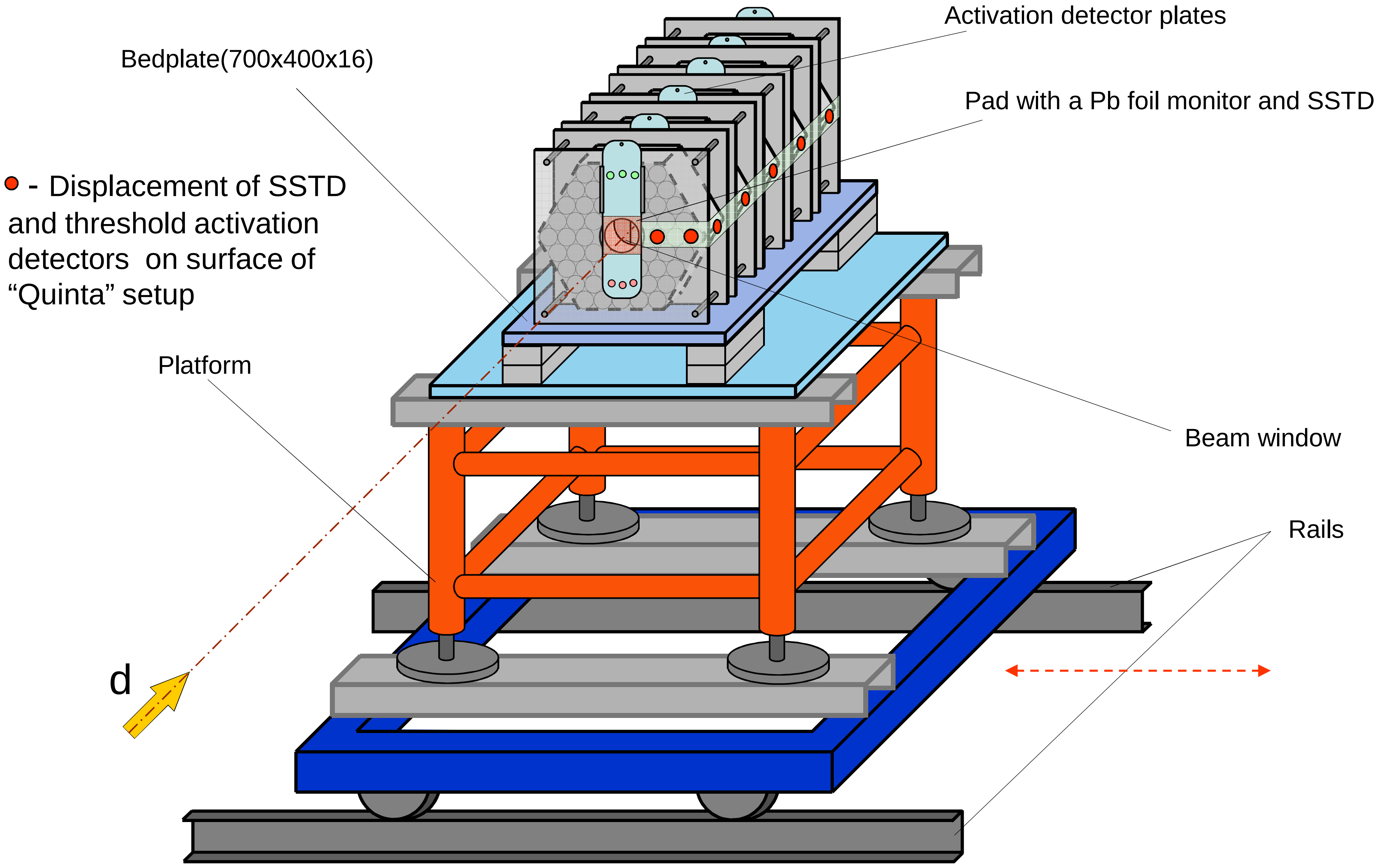


Front view photo

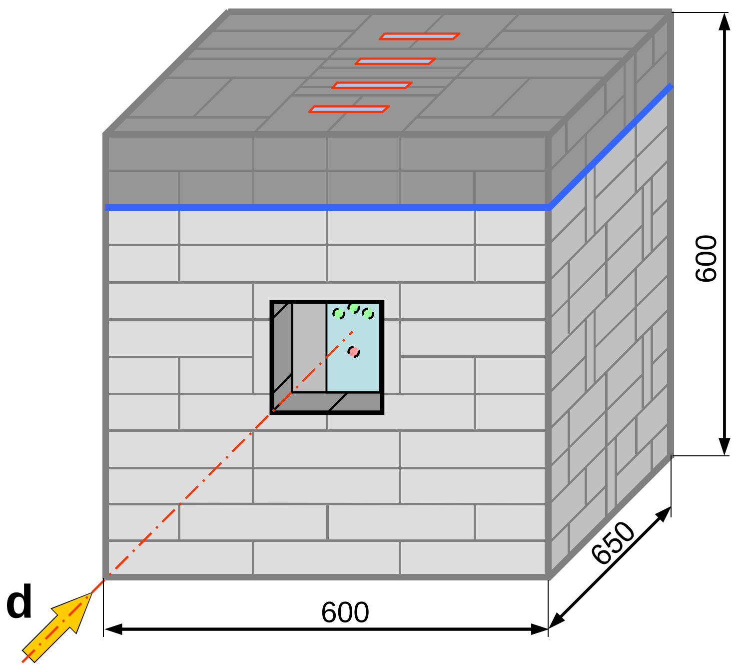


Rear view photo

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



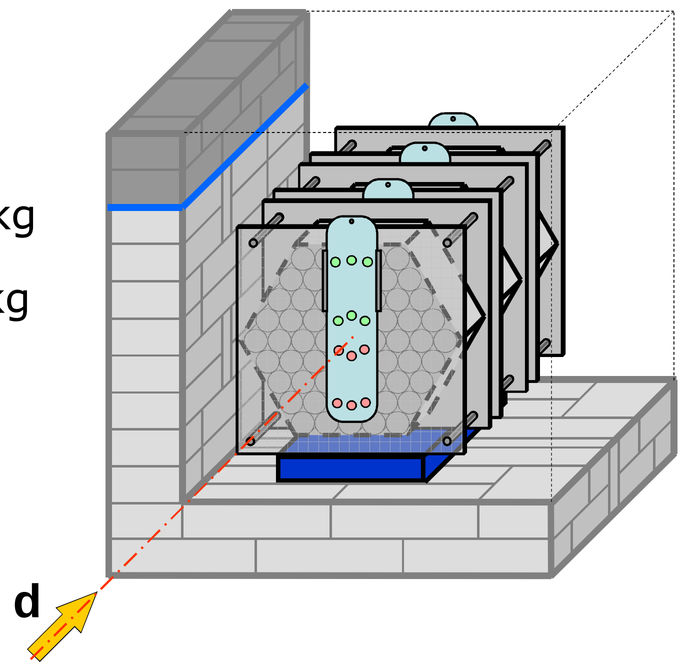
“Quinta” at the irradiation position (June 2009, December 2011, March 2012)



$m_U = 315$ kg

$m_{Pb} = 1780$ kg

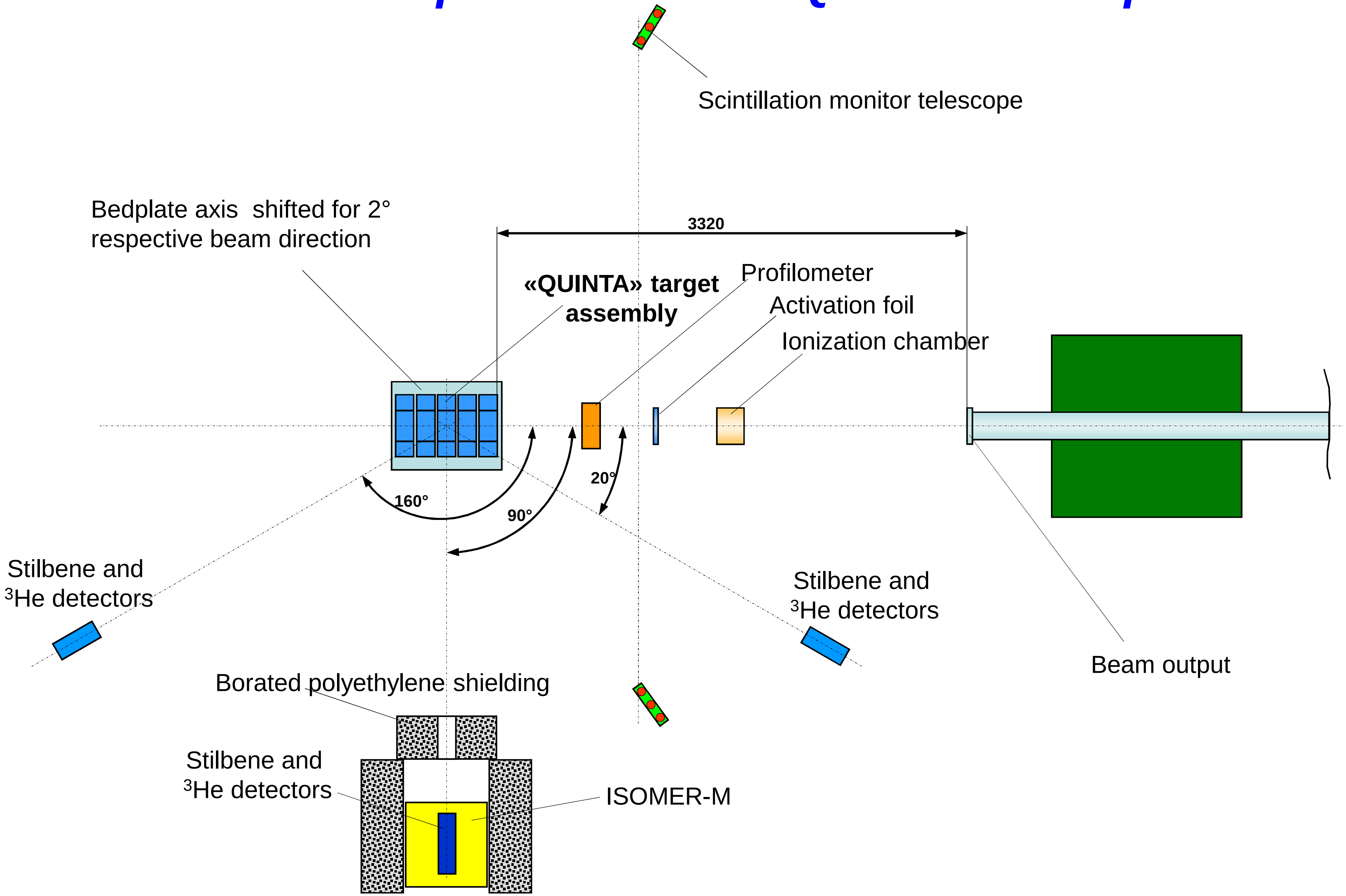
$m_\Sigma = 2125$ kg



Results of experiments 2011-2012

- **In present talk it'll be discussed shortly main results of experiments carried out with target assembly QUINTA during 2011-2012 aimed at study basic physics of RNT**
- **Detailed presentation of these results will be done in following talks of collaboration groups**
- ***It should be pointed out that all experiments with QUINTA setup are preparatory stage to future measurements with quasi-infinite AC BURAN planned for 2013-2015***

Scheme of experiment with “QUINTA” setup



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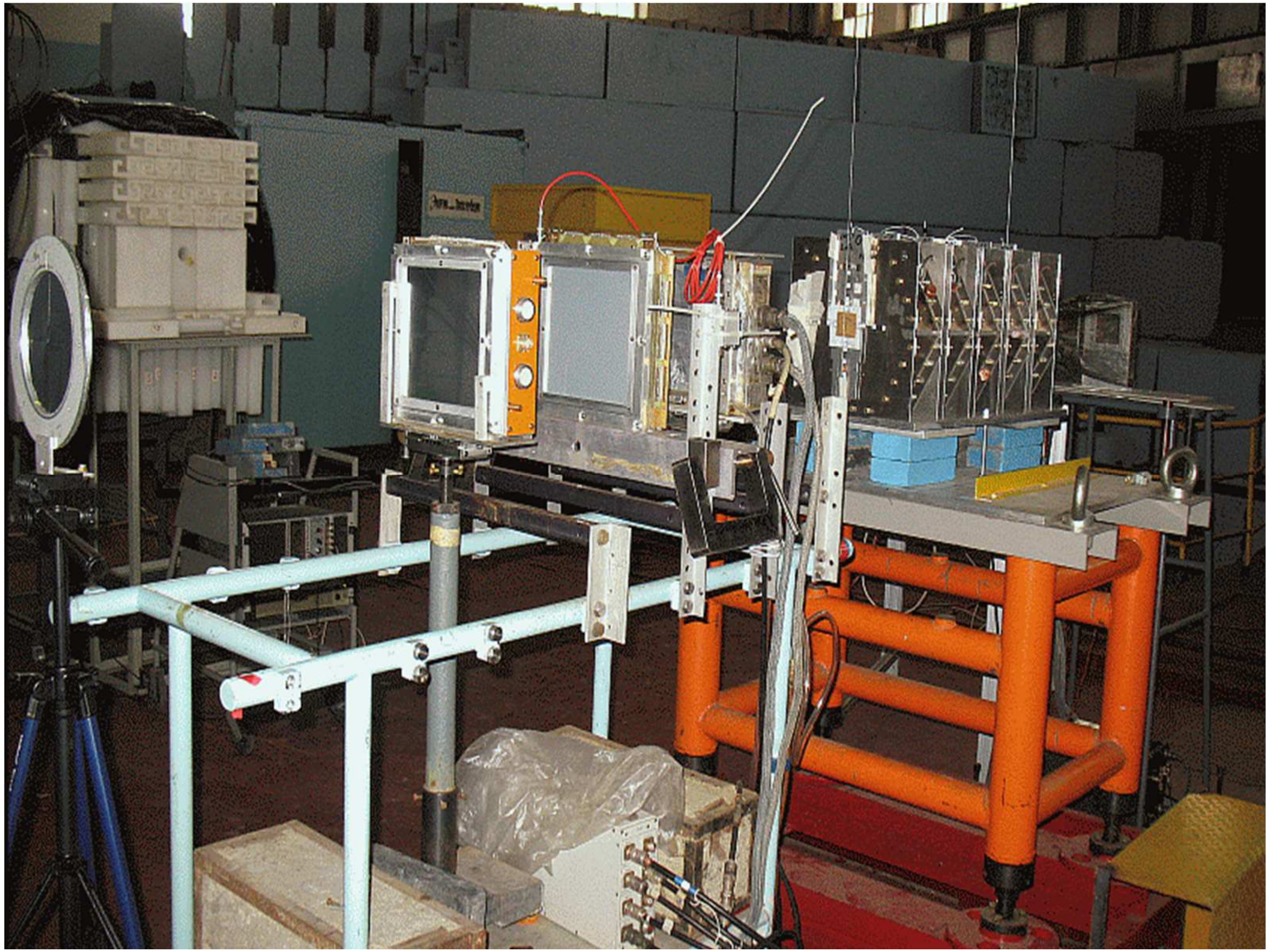
General view of the experimental setup



Deuteron beam dynamical and integral monitoring

- Al and Cu foils + ionization chambers
- Achieved accuracy is ~ 12-15%
- The problems of monitoring of incident beam intensity will be discussed in talks of V. Wagner, V. Voronko and I. Kudashkin later today

More details in V.Voronko talk

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 γ-line from ^{239}Np

γ- 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.

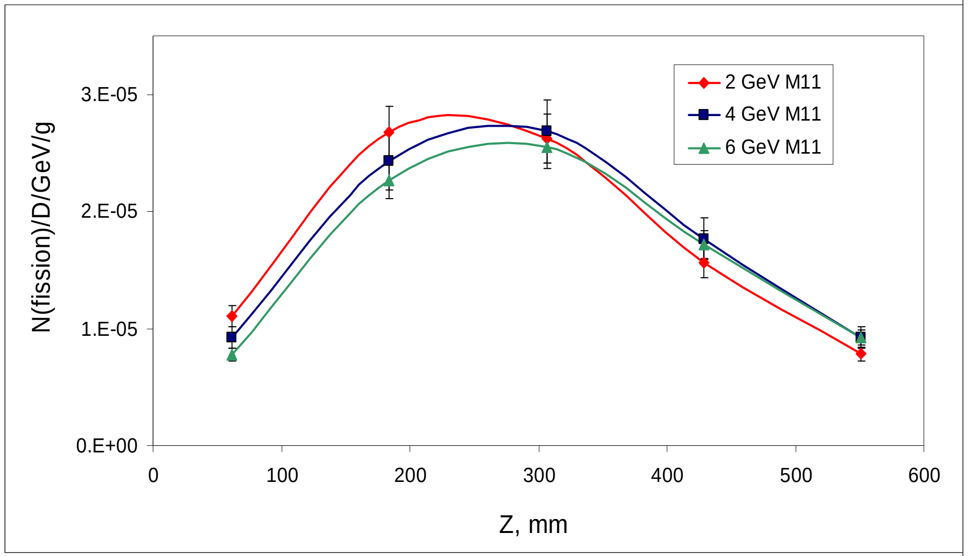
Number of fissions defines by averaging of following fission product yields:

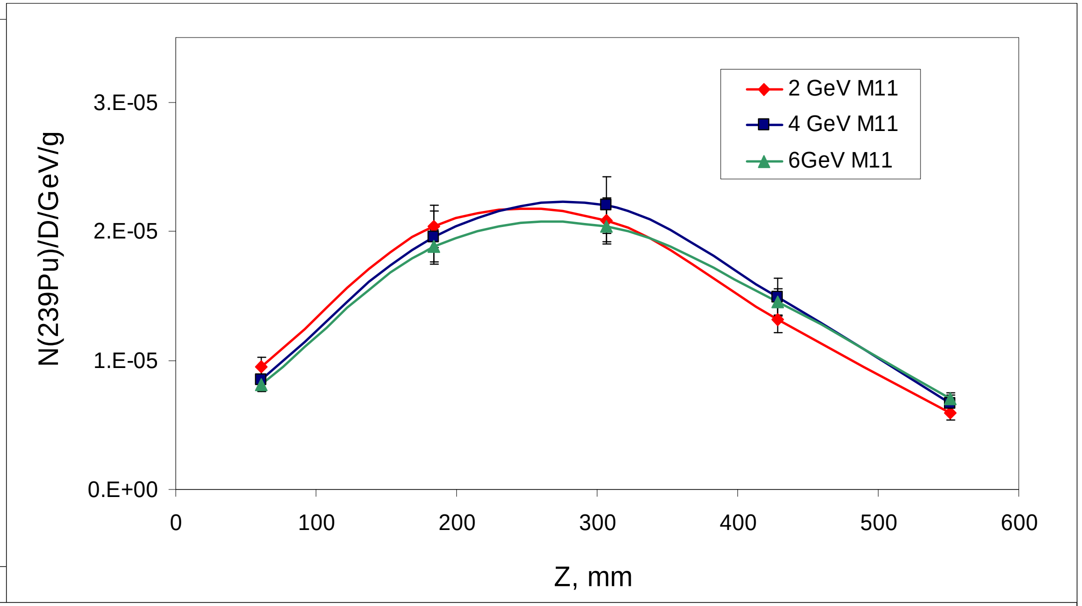
^{97}Zr (5.42%), ^{131}I (3.64%), ^{133}I (6.39%), ^{143}Ce (4.26%)

In brackets there are mean cumulative FP yields

- ^{239}Pu production rate is very important characteristics of present ADS
- In process of long term operation of the deep subcritical AC ^{239}Pu nuclei continuously generate and burn through fission and radiative capture up to reaching its equilibrium concentration.
- As theoretical calculations show at this concentration near (6-8)% BPG could essentially increase (up to an order of magnitude)

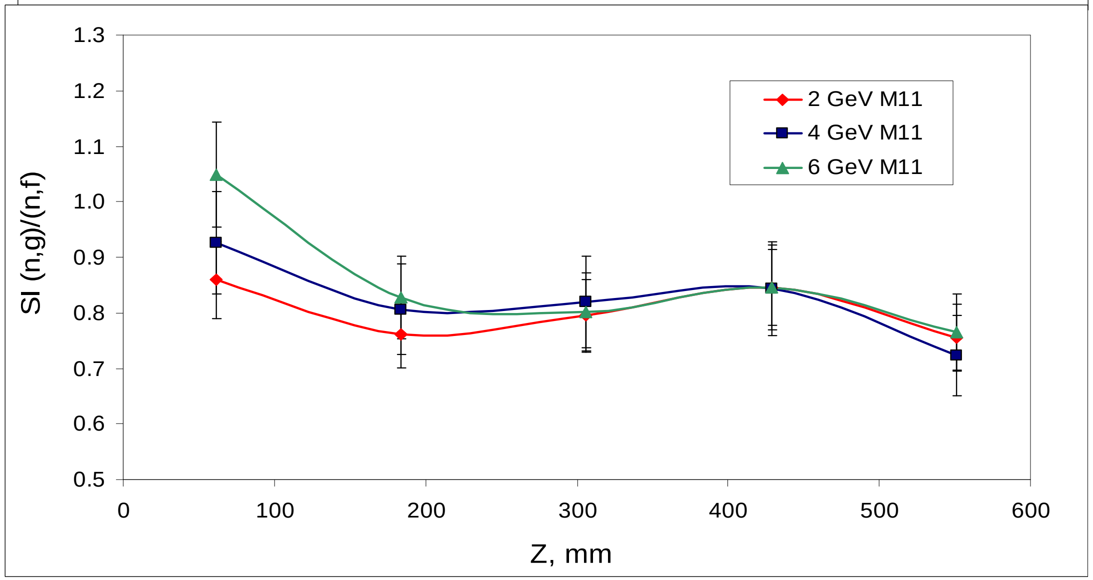
Average spatial distributions of (n,f), (n,γ) reactions and spectral index



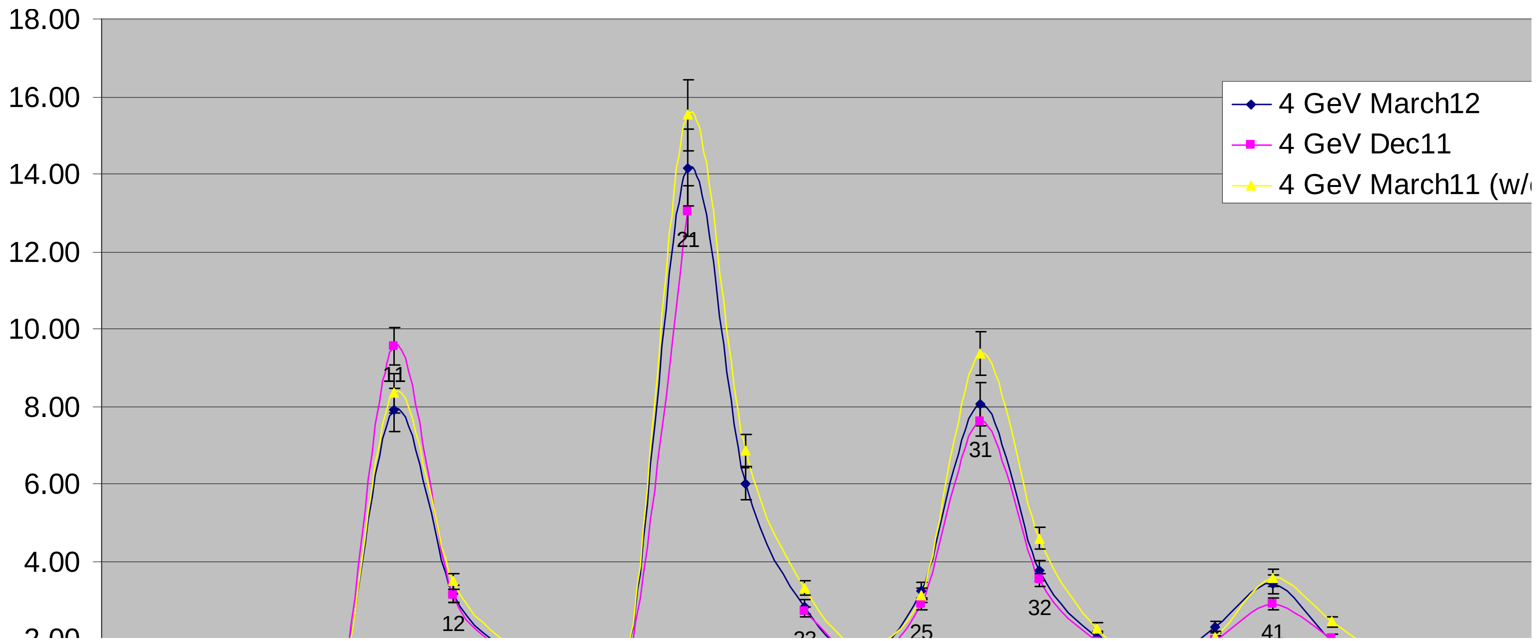


Axial densities of N_f, N_{Pu} and spectral indices averaged over its radial distributions inside of QUINTA.

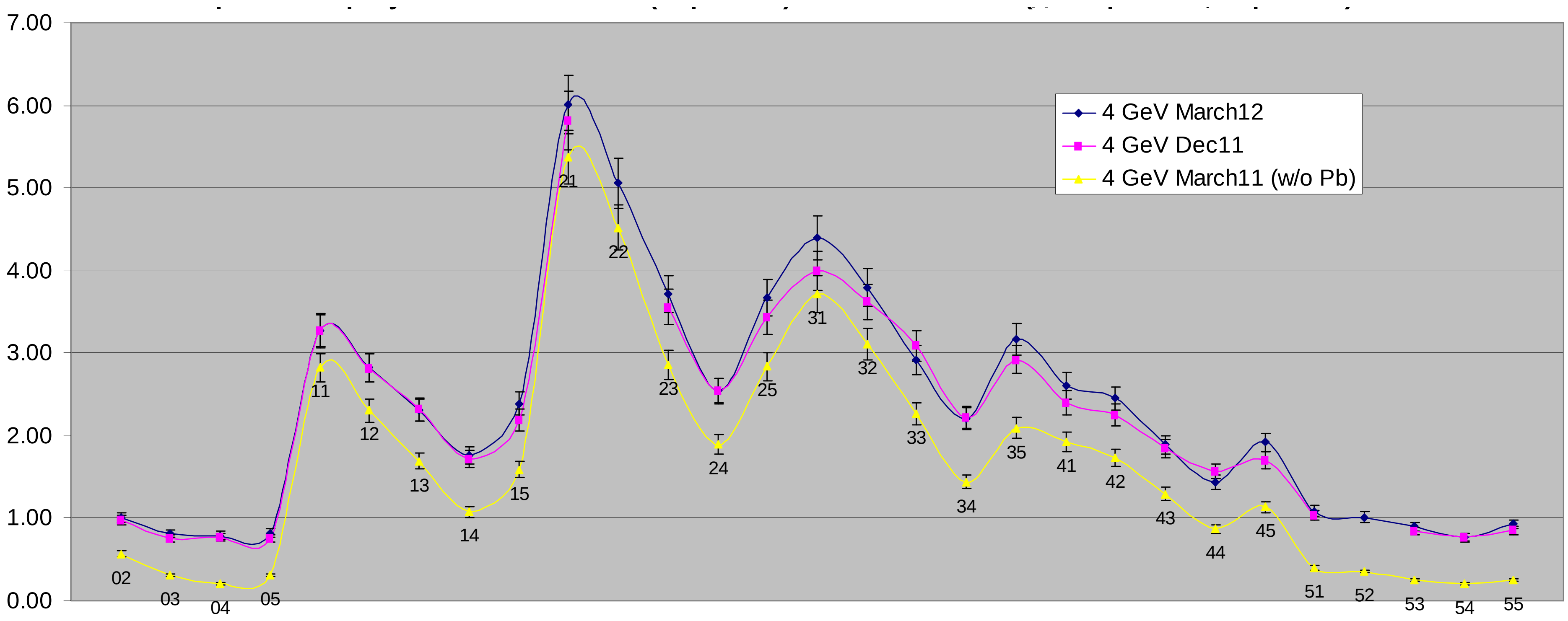
Similar axial distributions of N_f, N_{Pu} and N_{Pu}/N_f are for all deuterons energies from 1 to 8 GeV.



Distribution of N_f density over QUINTA volume
N_f x 10^{-6} /gr. /one deuteron/ 1 GeV



Distribution of N_{Pu} density over QUINTA volume
N_{Pu} x 10^{-6} /gr. /one deuteron/ 1 GeV

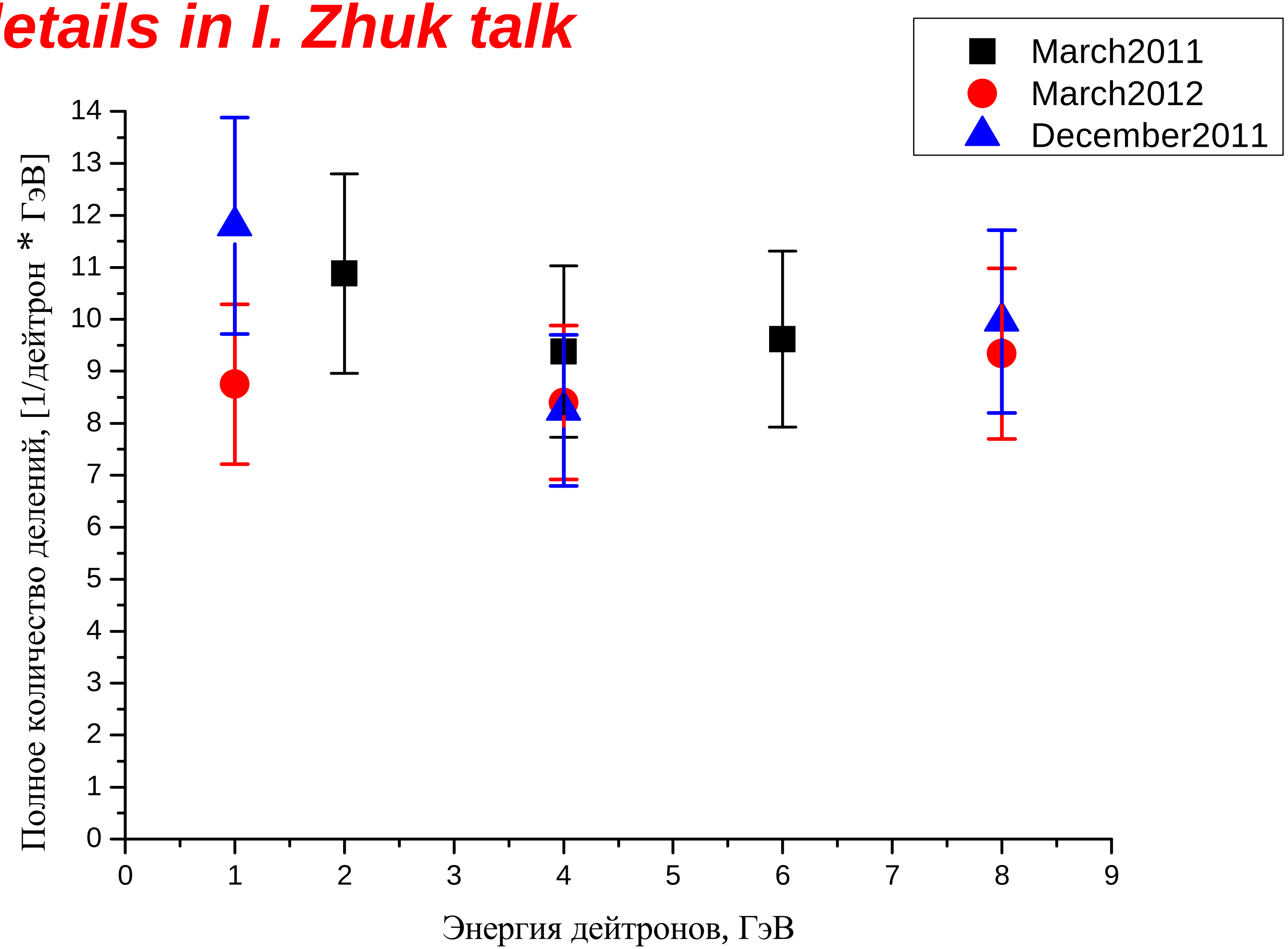


Total numbers (per 1 GeV) of produced nuclei ^{239}Pu (N_{Pu}) and fissions (N_f) of ^{nat}U into target assembly QUINTA

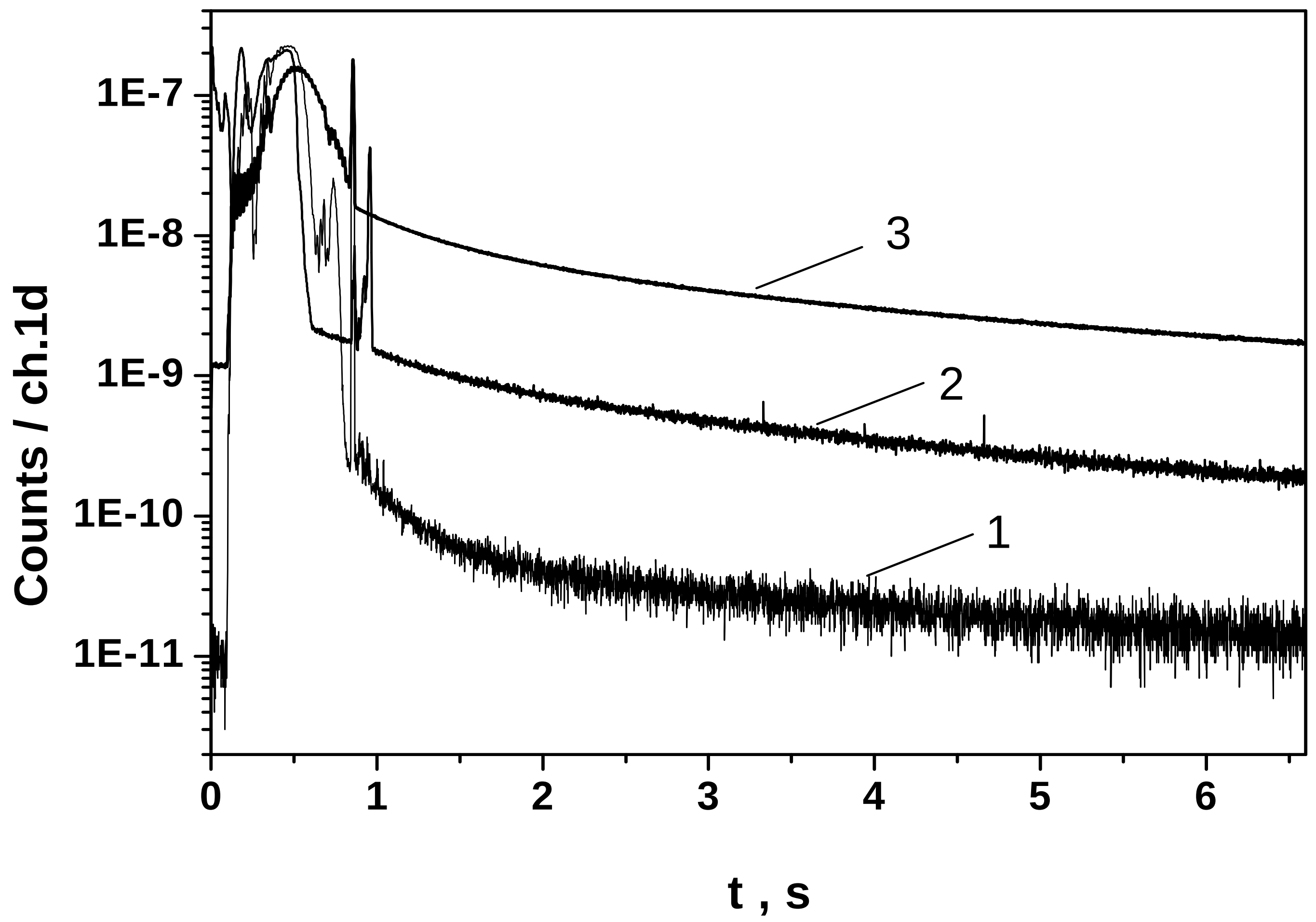
Run	*E_d GeV*	*N_{Pu}*	*N_f*
March 2011	**2**	**(7.0 ± 0.3) ± 0.8**	**(8.8± 0.4) ± 1.0**
(without	**4**	**(7.2± 0.4) ± 0.8**	**(8.8± 0.4) ± 1.0**
Pb blanket)	**6**	**(6.9± 0.3) ± 0.7**	**(8.3± 0.4) ± 0.9**
Dec. 2011	**1**	**(11.8± 0.6) ± 1.2**	**(10.6± 0.5) ± 1.1**
(with	**4**	**(10.8± 0.5) ± 1.1**	**(8.5± 0.4) ± 1.0**
Pb blanket)			
March 2012	**1**	**(11.6± 0.6) ± 1.2**	**(10.5± 0.5) ± 1.1**
(with	**4**	**(11.3± 0.5) ± 1.1**	**(9.7± 0.4) ± 1.0**
Pb blanket)	**8**	**(10.5± 0.5) ± 1.1**	**(9.7± 0.5) ± 1.1**

Total numbers of fissions from QUINTA per one deuteron and 1 GeV from SSTD

More details in I. Zhuk talk



Neutron time spectra measured in June 2009



The time dependence of the neutron yield from the geometrically identical lead and uranium targets. 1 - (Pb+d) for E_d = 4 GeV; 2 and 3 (U+d) for E_d = 1 and 4 GeV.

Comparison of DN and fission data QUINTA with lead blanket

Run	1 Gev	2 GeV	4 GeV	6 GeV	8 GeV
SSTD					
03.12	**8.8 ±1.5**		**8.4 ±1.5**		**9.3 ±1.6**
03.11		**11 ±1.9**	**9.4 ±1.7**	**9.6 ±1.7**	
Activation method					
03.12	**(10.5 ±0.5) ±1.1**		**(9.7 ± 0.4) ±1.0**		**(9.7 ± 0.5) ±1.1**
DN	***Normalized***				
03.12	**9.1 ±0.6**		**8.9 ±0.6**		**11.5 ±0.9**

V.G ~ 30

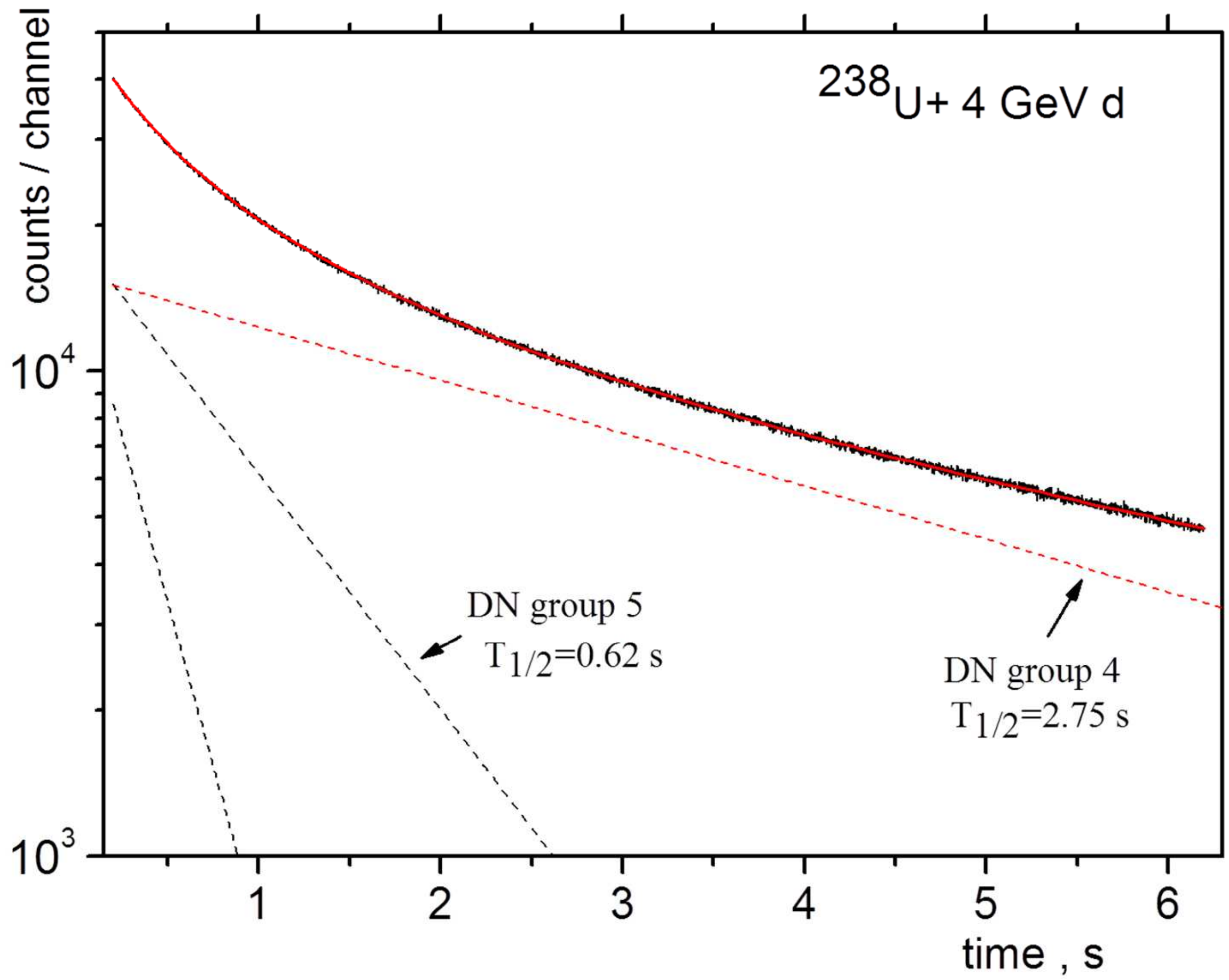
Comparison of DN and fission data

- ***The DN time spectra were obtained for three target assemblies irradiated by deuterons with energy 1, 2, 4 ,6 and 8 GeV.***

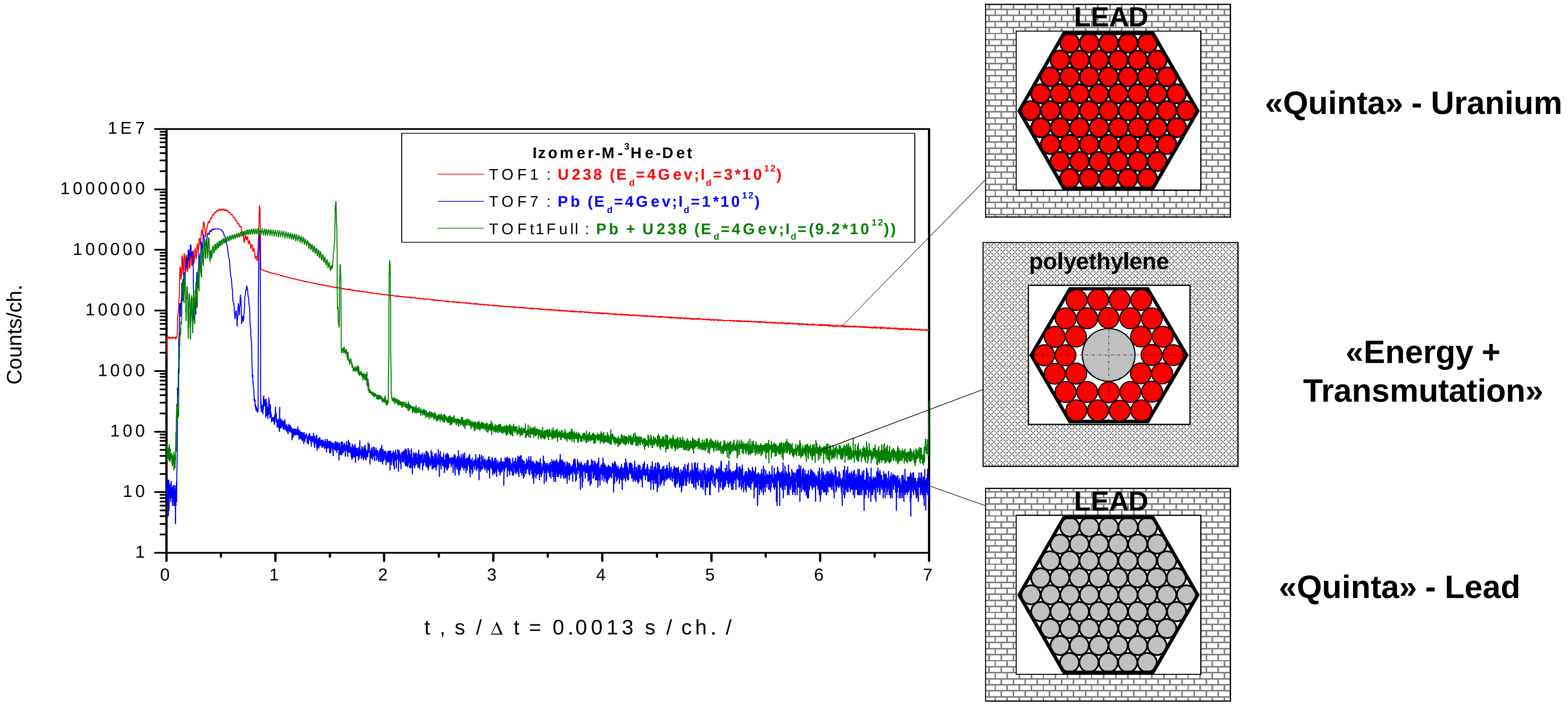
- ***In the last (most reliable) measurements relative DN total yields (per one deuteron and 1 Gev) are constant in limit of experimental errors for deuteron energy range (1 – 8) GeV in agreement with similar behavior of total fission numbers***

- ***An essential increase of these yields observed in June 2009 could be caused by deficiency in monitoring of deuteron fluxes as it became clear now.***

Group analysis of DN time spectra

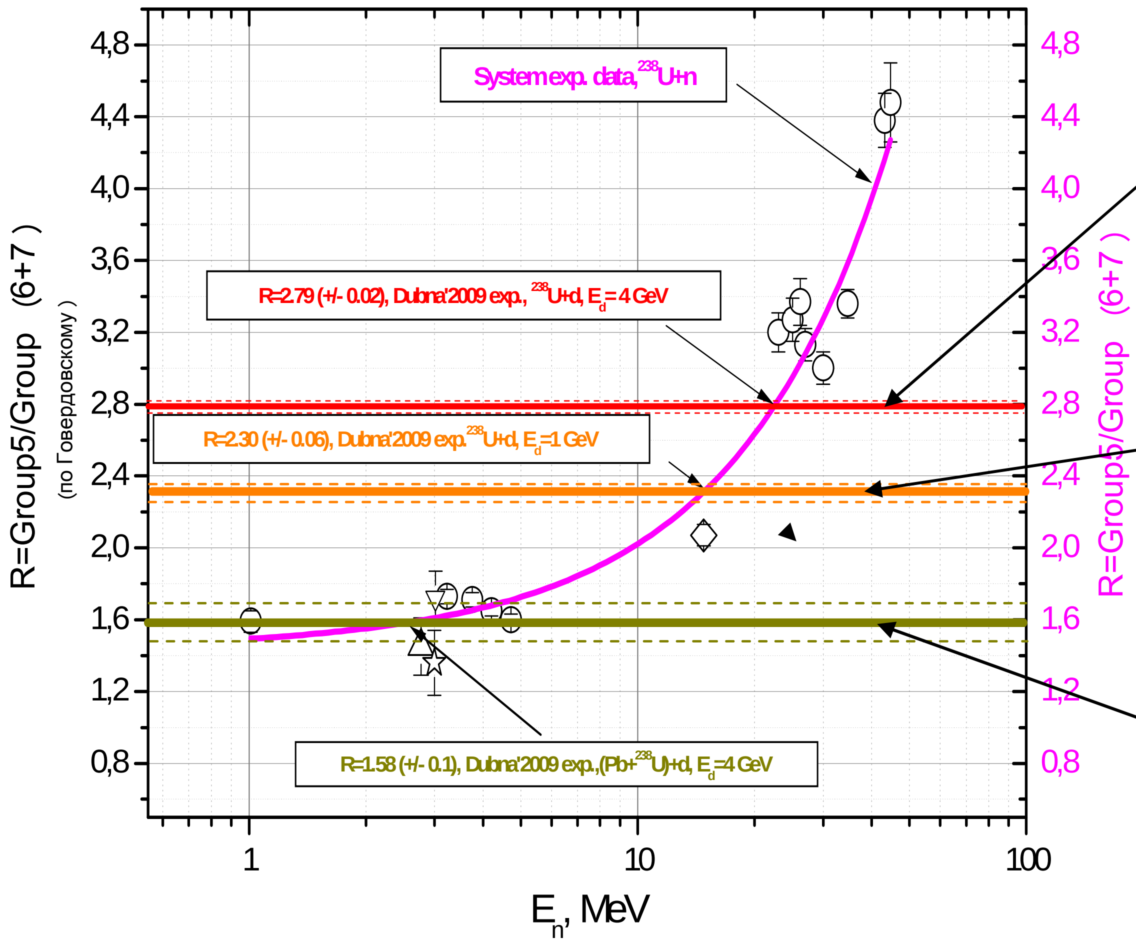


DN time spectra measured in June and November 2009



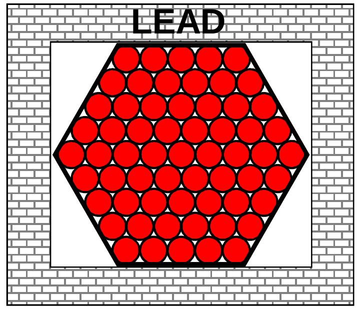
The time dependence of neutron yields from different target assemblies for E_d = 4 GeV.

Results of DN spectra analysis of June and November 2009 data



«Quinta» - Uranium

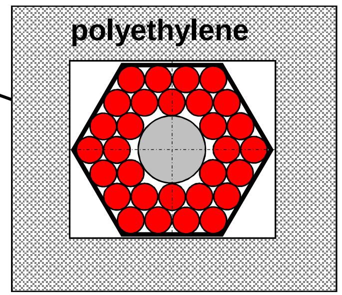
E_d = 4 GeV → $E_{n,f}$ ~ 25 MeV



E_d = 1 GeV → $E_{n,f}$ ~ 15 MeV

«Energy + Transmutation»

E_d = 4 GeV → $E_{n,f}$ ~ 3 MeV

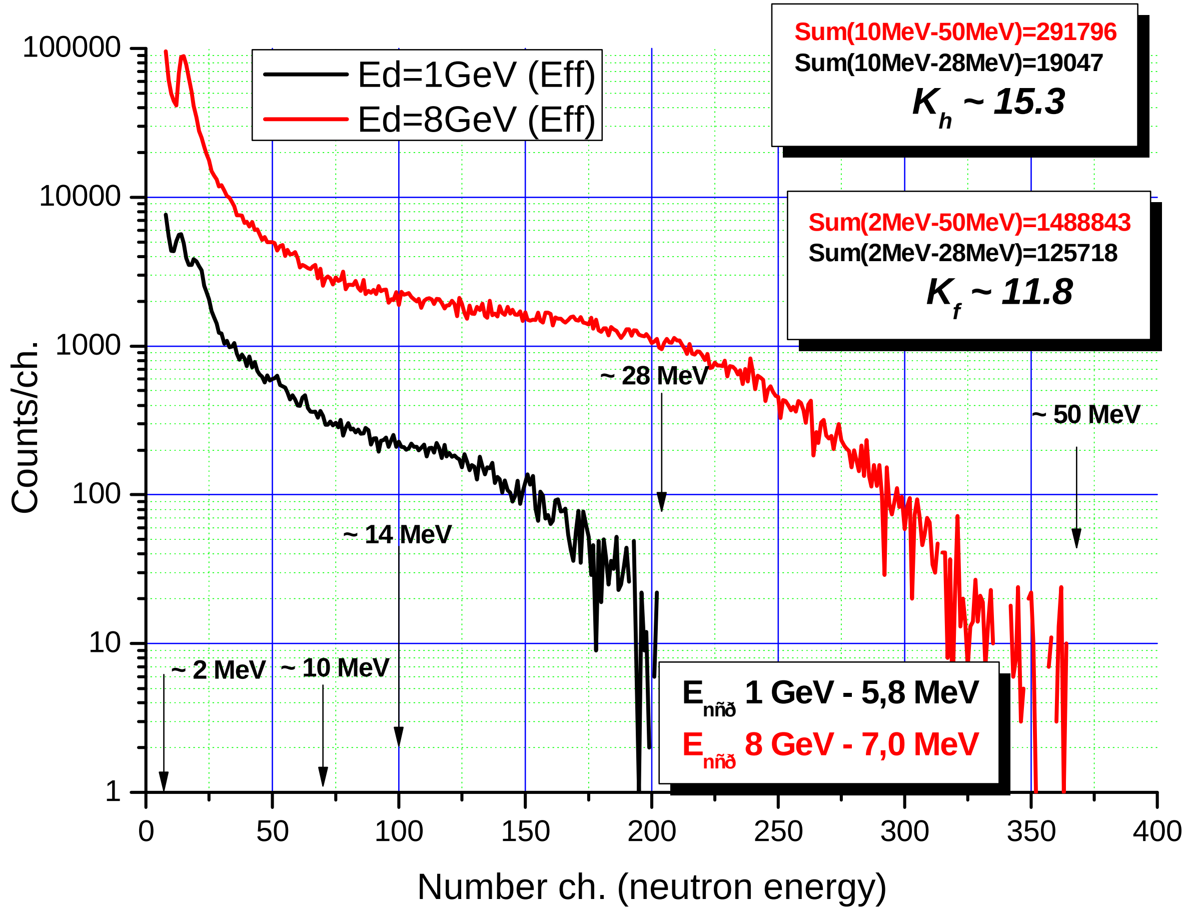


Comparison of neutron energy dependence of the weight ratios of 5-th to (4-th) DN groups from ^{238}U(n,f)-reaction and similar values extracted from DN time spectra measured in present work.

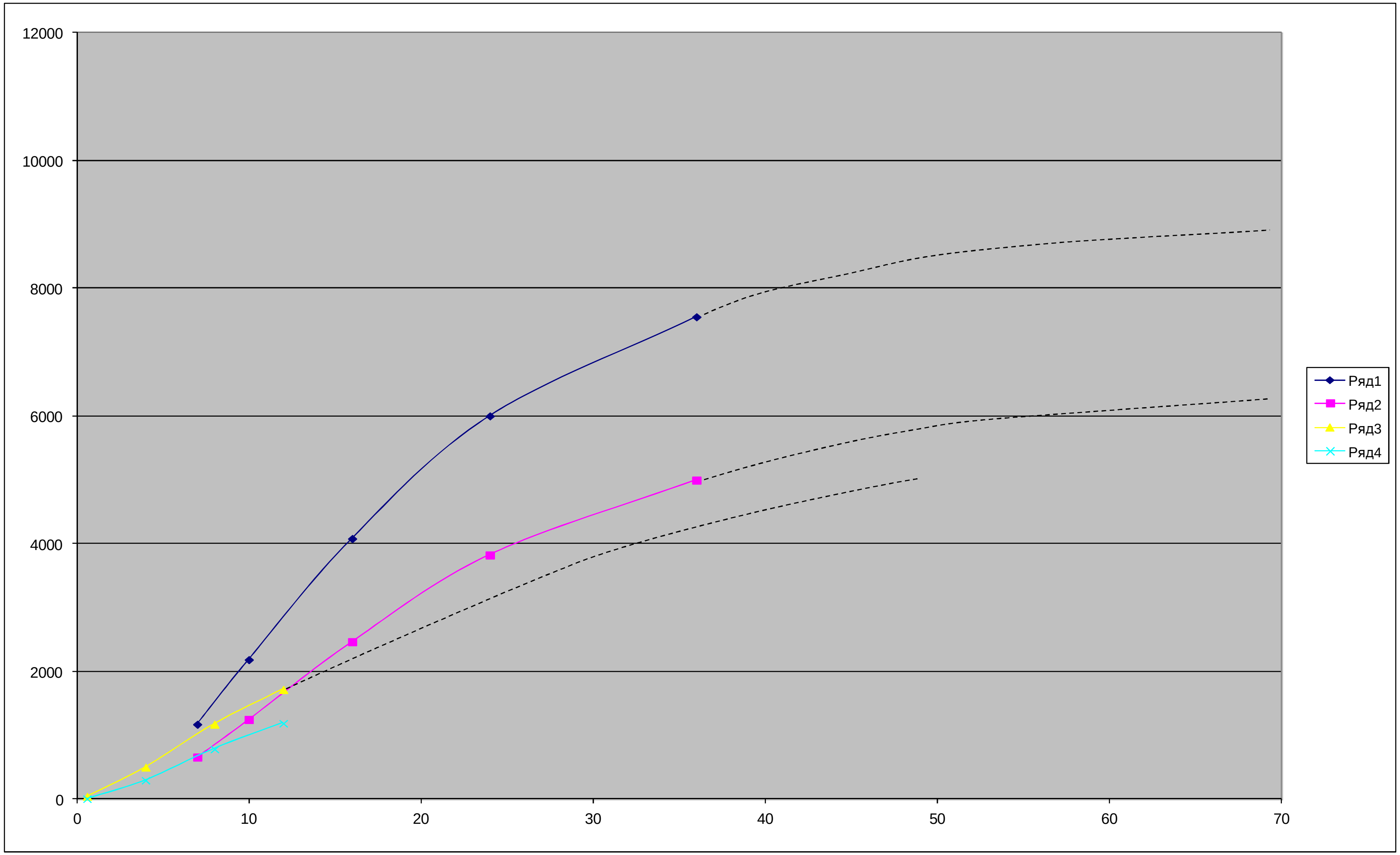
Spatial and energy distributions of neutrons in AC

- Main problem in our experiments is in obtaining of a reliable information on neutron field inside the target assembly
- There were used the technique of threshold activation detectors but up to now we have no reliable data on neutron spectra and its variation inside of QUINTA volume
- *More details in talk of B. Martsynkevich and as new approach to the problem a talk of S. Zhdanov*
- *Very important results on measurements and analysis of the relevant reaction cross-sections will be presented in talks of V. Wagner and L. Zavorka*

Leaked neutron amplitude spectra measured by DEMON detector for Ed = 1 and 4 GeV (March 2012, preliminary)



Discussion of results



Comparison of radial distributions of fission rates in quasi-infinite AC (V. Vassil'kov et al) and QUINTA

- From analysis of radial dependence of integral fission yield obtained from R. Vassil’kov et al data one could estimate a neutron leakage at R=15cm as ~ (55-60)%
- Accounting for difference in uranium densities of QUINTA and Vassilkov’s target assembly we can estimate leakage of neutrons from QUINTA

  as ~ (80-90)%
- *But this conclusion has to be tested by direct measurements planned for next run*

From paper of K.W. Allen et al (1961)

Inner radius cm	Outer radius cm	Thickness cm	T	M	η
11.93	12.70	0.77	0.905	1.224	3.36
5.85	7.88	2.03	0.752	1.568	3.29
5.85	9.91	4.06	0.557	1.990	3.24
4.44	15.75	11.31	0.171	2.809	3.18

$$\eta = 1 + \frac{(\nu - 1)\sigma_f + \sigma_{n,2n} + 2\sigma_{n,3n}}{\sigma_i}$$

$$\sigma_i = \sigma_f + \sigma_{n,n'} + \sigma_{n,2n} + \sigma_{n,3n}$$

$$M = T + \eta(1 - T)$$

Nf = 0.6 per one 14 MeV neutron

η - number of neutrons released per inelastic collision of one 14 MeV neutron

M(T) - neutron multi-lication (transmission)

Discussion of results

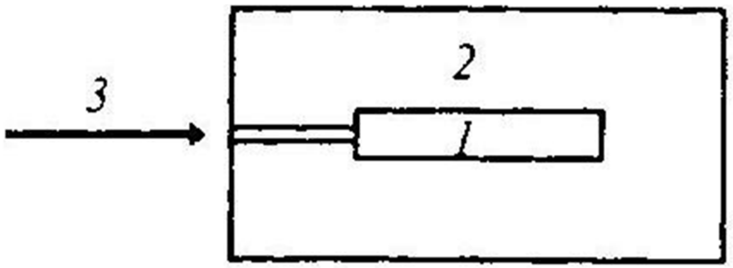
- 1 GeV deuteron energy correspond approximately to 0.5 GeV proton energy

- So taking into account a linear dependence of total fission yield from QUINTA on incident energy in measured range we can obtain as low limit for BPG of quasi-infinite AC the value ~ 8

- It is appropriate to compare our data with calculations of V. Batyaev et al (2008)

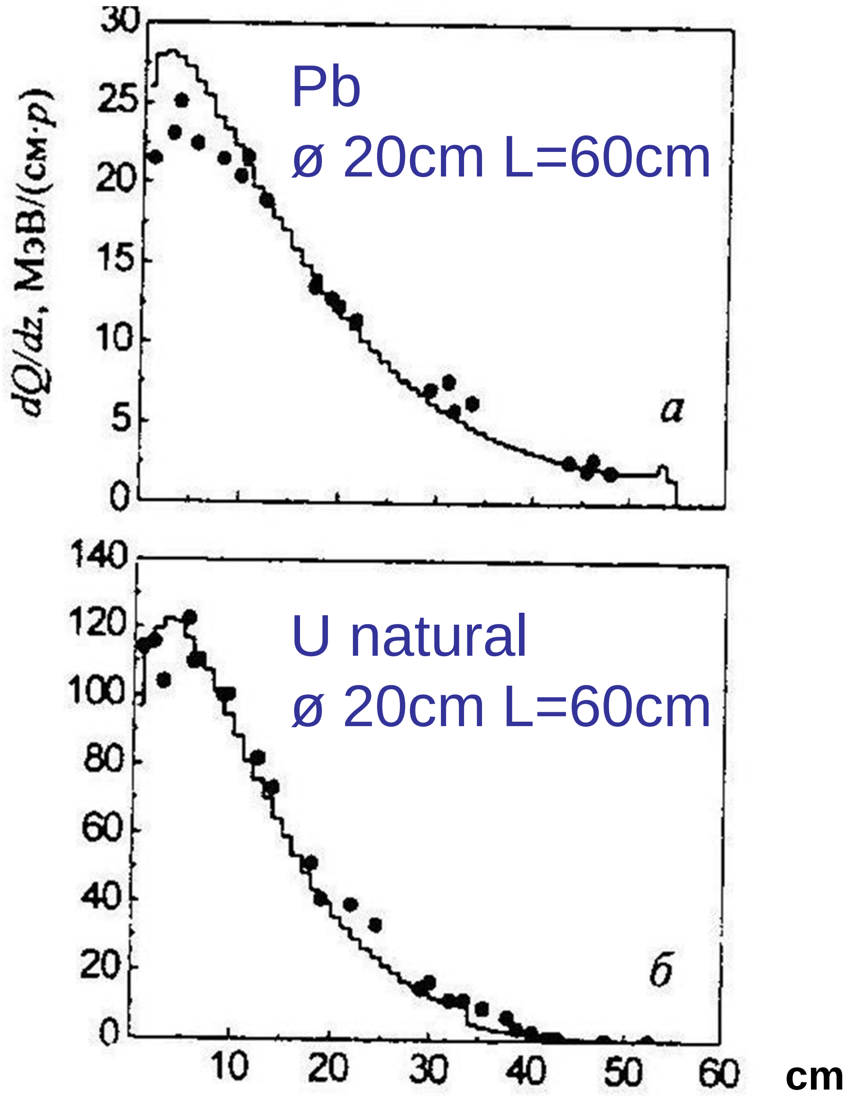
- Authors of this paper attempted to assess the possibilities of RNT and concluded that given approach can not achieve its goals

Calculation by V. Batyaev et al (2008)



Model ADS setup
1 – target ø 20cm, L= 100cm
2 - blanket ø 100cm, L= 200cm
3 - proton beam
Target mass ~ 30 tons

Experiment (E_p = 1 GeV)
V. Belyakov-Bodin et al, 1990 →



Results of calculations of V. Batyaev et al. (2008)

Ep, GeV	Q, GeV	Beam Power Gain	N_{Pu}	N_f En>14MeV	N_f En<14MeV	M_n	<En>	M_{nesc}	<E_{nesc}>
Uranium (depleted) target + Uranium(depleted) blanket									
1	3.27	3.27	68.6	2.93	12.7	44.6	3.05	2.40	0.37
10	29.8	2.98	609	26.5	113	403	3.05	25.2	0.4
40	102	2.54	1930	83.9	358	1260	3.01	77	0.4
Lead target +Lead blanket									
1	0.64	0.64		0.029	171	34.4	3.02	33.7	0.38
10	6.27	0.63		0.12	1550	292	3.06	295	0.40
40	27.3	0.68		0.44.1	5000	934	3.03	930	0.40

But N_f obtained by R.Vassil'kov is ~ 17 at Ep = 0.66 GeV and BPG ≈ 6

Beside the calculated mean energy of neutron spectra seems to be too small in comparison of V. Yurevich et al data

For ø20x60cm lead target irradiated by GeV protons

from V. Yurevich et al (2006)

Eb *GeV*	*<En>* *MeV*	*En kin* *MeV*	*En kin/Eb* *%*	*W* *MeV*	*W/Eb* *%*
0.99	8.82	213	21.3	382	38.2
2.0	11.6	513	25.6	822	41.1
3.65	13.7	116	30.3	1670	45.6

W/Eb is a share of beam energy Eb expended in the formation of neutrons

Conclusion

- ***So only new experiments with quasi-infinite BURAN AC establish reliably a feasibility of RNT approach***
- In next two NUCLOTRON runs (the end of 2012 and March 2013) the program of measurements with QUINTA should be completed
- It aims to prepare and test all experimental techniques planned to use in experiments with BURAN setup

- There is planned test new systems with silicon detectors for on-line registration of spatial distribution inside of AC of fission rates and (may be neutron spectra) because number of experimental channels in BURAN setup so much to use only SSTD and activation techniques
- New method proposed by Kazakstan’s group of our collaboration will be used for measurement of an effective energy provoked fission of 238U inside of AC
- Study of probe samples of fissile isotopes with different thresholds gives information on neutron fluxes in wide intervals of its energy

- It should be completed methodical stage of DEMON detector use and obtained a reliable information on leakage neutrons
- The experiments with long lived minor actinides and fission products as well as with massive thorium samples are planned.
- Upgraded beam monitoring systems will be tested aiming to improve a reliability of its operation
- There will be continued new experiment with transmission of deuterons and high energy neutrons throughout QUINTA
- ***E&T and RAW collaboration is open for new proposals and new participants***

Table 2.- Summary of detector parameters.

Detector	Active Element	Uranium converter	Clustering	Location	Number of units
Gas Ionisation Chamber 4 ata Argon	Circular window, Φ=15 mm vertical plane	1 mg/cm^2 deposit	1 array of 6, spaced 10 cm vertically . 2 arrays of 8, spaced 12.3 cm vertically	in water, between U bars	22
Polycrystalline Si Diode thickness 300μm	Rectangular. window 9.6 x 11.6 mm^2 vertical plane	1 mg/cm^2 deposit	10 arrays of 16 counters, spaced 6.4 cm vertically	in water, between U bars	160
Polycrystalline Si Diode thickness 300μm	90° sector r= 14 mm horizontal plane	1 mg/cm^2 deposit	4 clusters of 6 counters spaced 21.3 cm vertically	in fuel bar, between U cartridges	24
Thermistors in metallic probes	U Cylinders Φ=8 mm Pb Cylinder Φ=10 mm	~ 55 g U	3 thermometers with two U probes	in water, between U bars	6
Lexan foils track detectors	Equilateral triangles r=37mm Circles Φ=32 mm Rectangle 10x25 mm^2 Vertical plane	~ 1 mg/cm^2	5 vertical sets of 2 detectors	in water, between U bars	10

Conclusion

- *Obtained results provide solid grounds for planning of future experiments with quasi-infinite (20 tons) target of depleted uranium available for the "Energy and Transmutation RAW" collaboration at JINR.*

Thanks for your attention