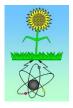
New ADS scheme with deep subcritical multiplying core for energy production and transmutation of radioactive wastes. First experimental results and perspectives.

> W. Furman on the behalf of *"Energy and transmutation RAW" collaboration,* Joint institute for nuclear research, 141980 Dubna, Russia, <u>furman@dubna.ru</u>

"Energy and transmutation RAW" collaboration

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Introduction

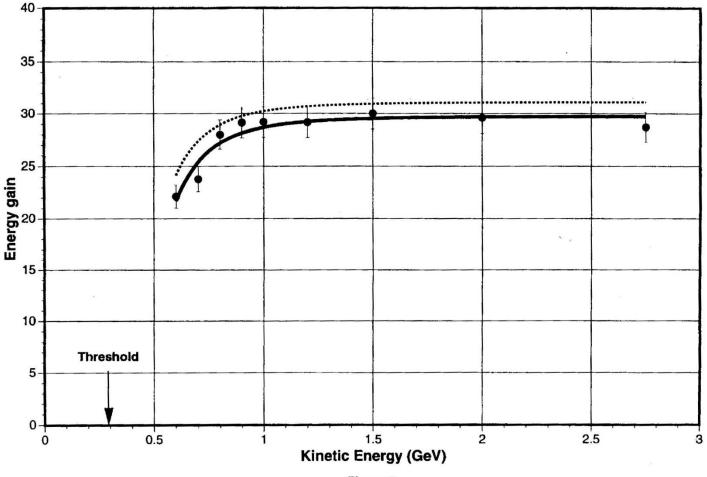
- The problems of rapidly growing energy consumption in the world can not be solved without the use of nuclear energy.
- The key issue here is the availability of an adequate supply of nuclear fuel. In the long term aspect, the use of such materials as enriched ²³⁵U or artificial ²³⁹Pu can not solve the problem of global energy.
- Indeed, a receive of ²³⁵U or ²³⁹Pu is very energy intensive, and the total value 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.

Problems of natural uranium and thorium use

- In the last decade of last century on the initiative of Carlo Rubia was carried out extensive work on exploring the possibilities of so-called energy amplifiers (EA) based on accelerator driven subcritical systems (ADS).
- Were investigated up to sketchy engineering work options such systems, as fuels to be used natural (depleted) uranium or thorium.
- 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 (GBP)* 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 Gain of the Beam Power (GBP)

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





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 multiplicity coefficient \mathbf{k}_{eff} this system was near 0.9.
- In these circumstances in spite of rather promising GBP~30 it is difficult to implement "burning" of the base core material (natural uranium) because of their high fission threshold.

 $3500 \text{ kg } 238U \rightarrow 25 \text{ kg } 235U \rightarrow \text{GBP } \text{!!!}$

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

New is a forgotten old ...

- About 50 years ago at Dubna by Vasil'kov-Goldansky group it was obtained GBP ~ 7 with only 0.66 GeV protons and 3.5 tones of natural uranium target.
- They did not use any moderator and applied the special geometry so their target was equivalent ~ 7 tones setup with rather small (<10%) neutron leakage.
- In this case of "quasi-infinite" active core maximally hard neutron spectrum has been realized with rather low
 *k*_{eff} ~ 0.4
- So it is very attractive to investigate GBP of such type of ADS active core for higher incident energy

The JINR project "Energy&Transmutation RAW"

- Recently as direct continuation of Vasil'kov-Goldansky experiment the new project to study the basic properties of ADS with large natural uranium core driven by proton and deuteron Nuclotron beams with energy up to 10 Gev was proposed and adopted for implementation at JINR during 2011-2013.
- The project has name "Energy&Transmutation RAW" and is aimed at study of basic features of such AD systems (so called Relativistic Nuclear Technology (RNT)) for energy production and utilization of spent nuclear fuel.

Basis features of RNT

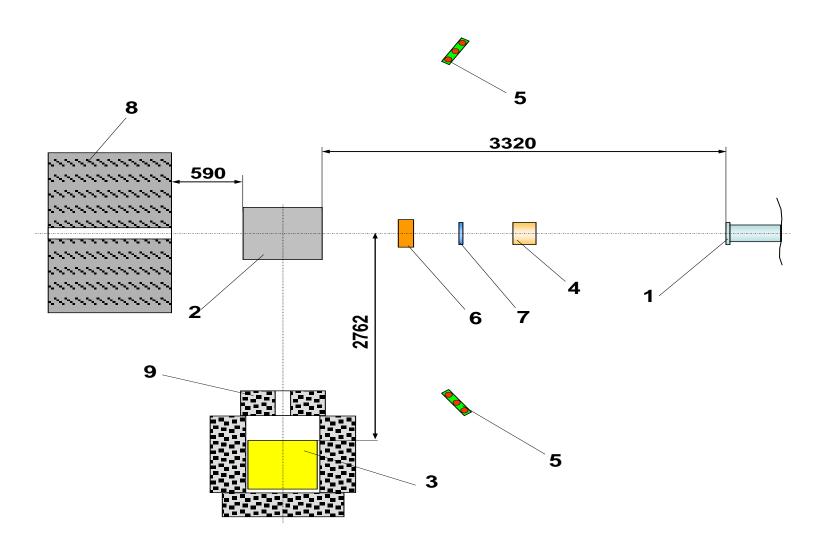
- Using the deep subcritical active core (AC) of natural (depleted) uranium or thorium the size of which provides minimal leakage of neutrons
- An increase in energy of incident particles up to ~ 10 GeV instead of 1 GeV as in the traditional ADS schemes
- Using as a target for incident beam the material of AC
- 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
- Using the technology of high temperature helium coolant for primary circuit

 For realization of "E&T RAW" project there are two targets at JINR :

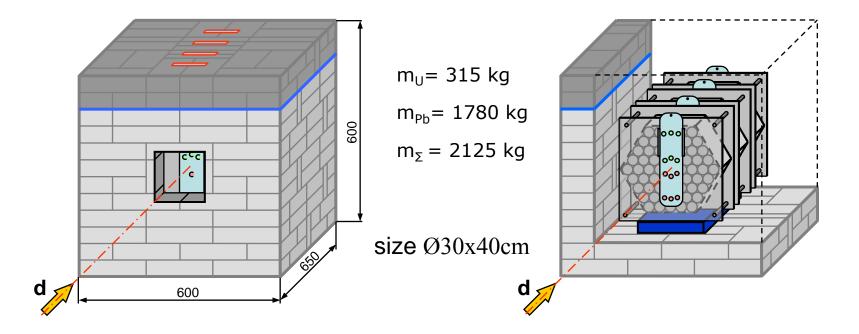
 semi-infinite (~22 tones) depleted uranium AC and smaller (~500kg) target modeling the central part of this AC

- In present talk there is discussed the results of experiments carried out with smaller targets during 2009-2011 aimed at study first three aspects of RNT
- These experiments are preparatory stage to future measurements with quasi-infinite AC planned for 2013

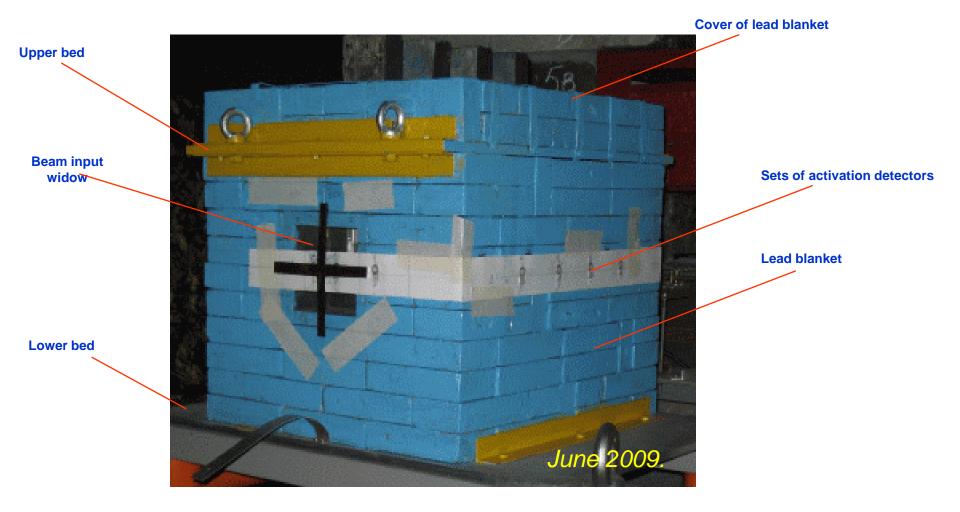
Layout of experiment performed in June 2009

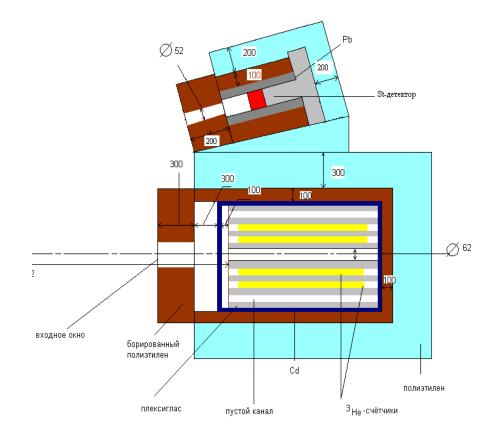


Target assembly "Quinta" (June 2009)



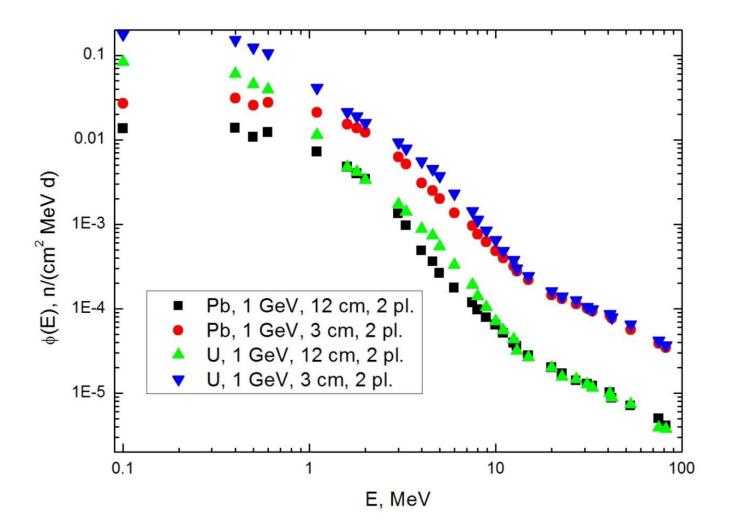
"Quinta" at the irradiation position (June 2009)

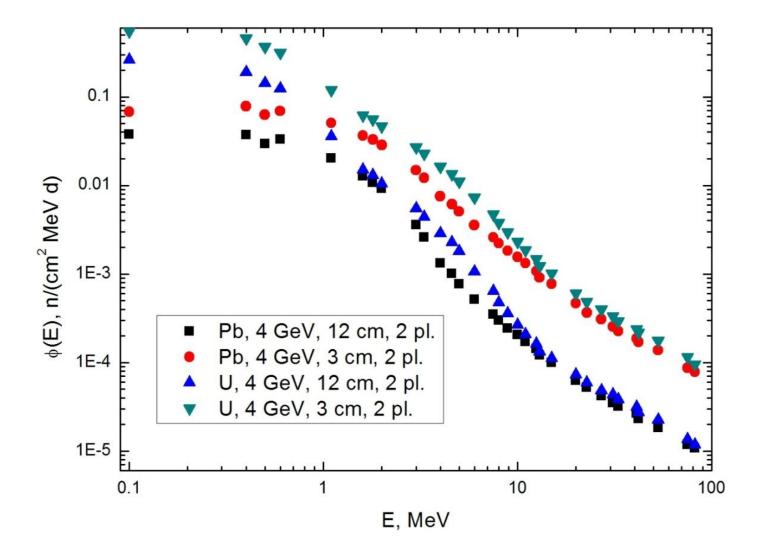


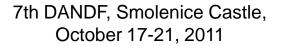


For $E_d = I$ and 4 GeV it was studied :

- the neutron spectra inside the uranium target (and lead target for comparison)
- the time dependence of the neutron yield, including its delayed component (DN)
- Study of DN time spectra expected to be a very sensitive test for basic mechanisms of the fission process inside of massive target

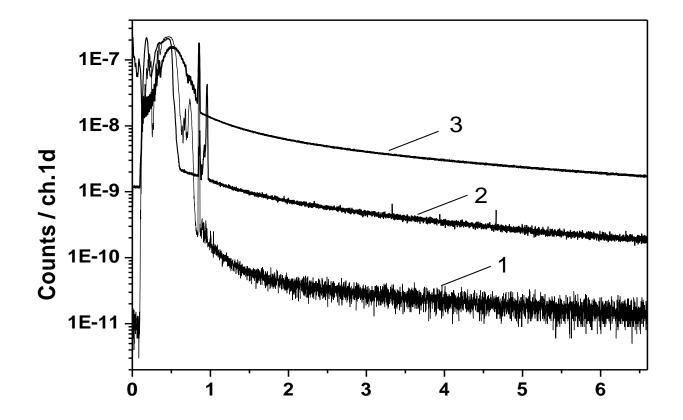




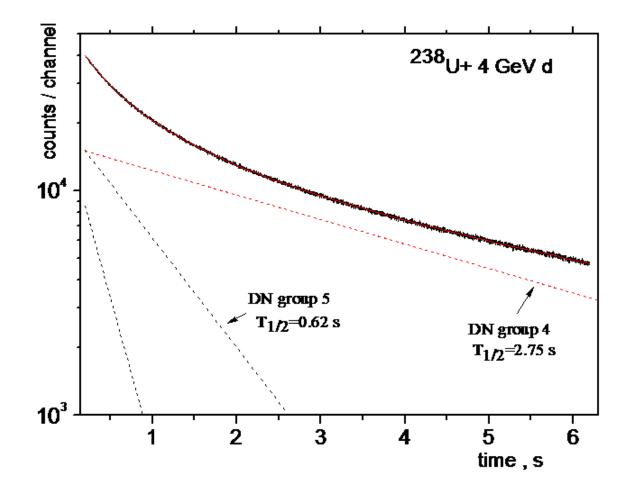


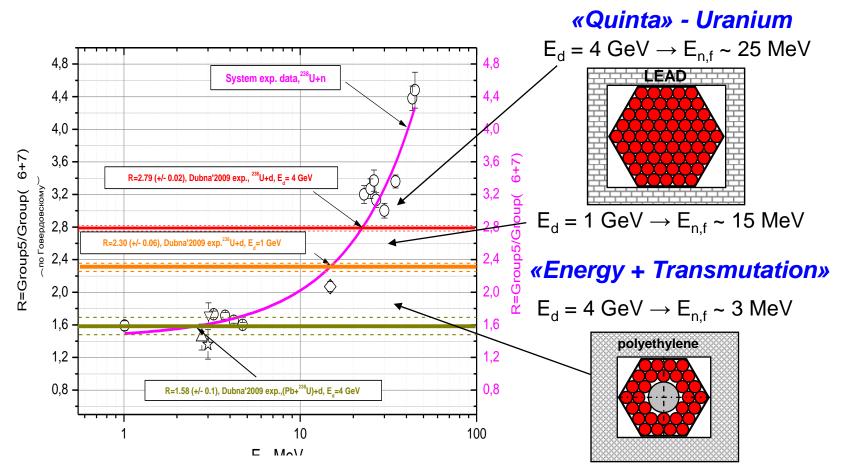
- The comparison of prompt neutron energy spectra from Pb and U targets demonstrates the pronounced contribution of prompt fission neutrons for the uranium target in the energy range (1 – 10) MeV
- Role of these fission neutrons is more important for the central zone of the U target than for its peripheral regions
- This effect becomes more pronounced with increasing incident deuteron energy

Results measured by IZOMER-M in June 2009



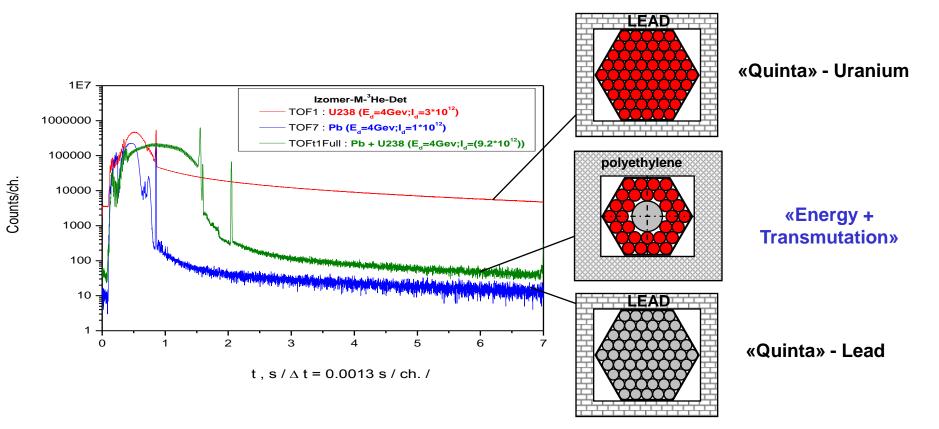
The time dependence of the neutron yield from the geometrically identical lead and uranium targets. 1 - (Pb+d) for $E_d = 4$ GeV; 2 and 3 (U+d) for $E_d = 1$ and 4 GeV.





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

Results measured in November 2009

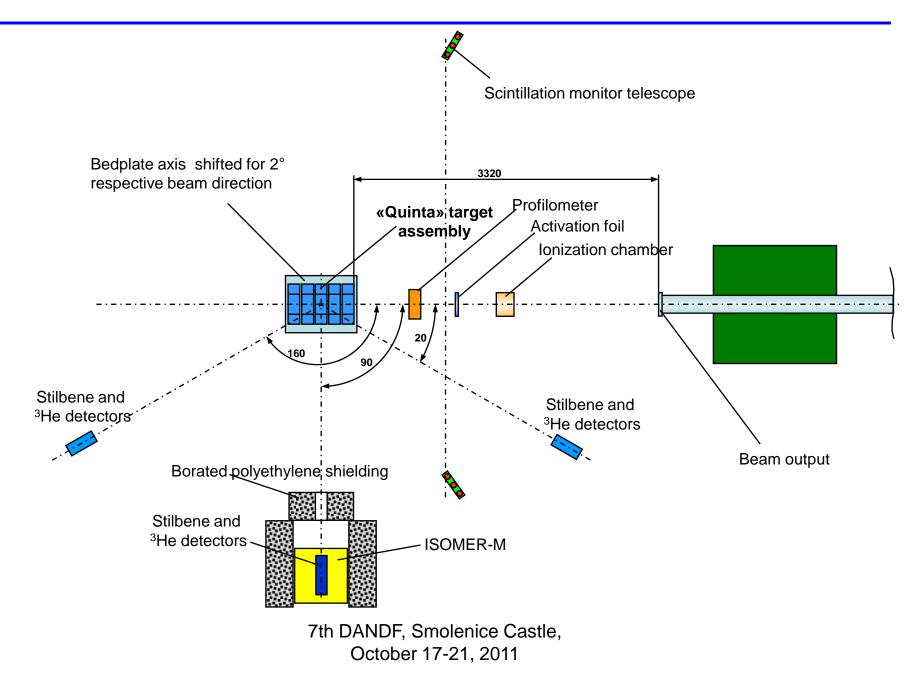


The time dependence of neutron yields from different target assemblies for $E_d = 4$ GeV.

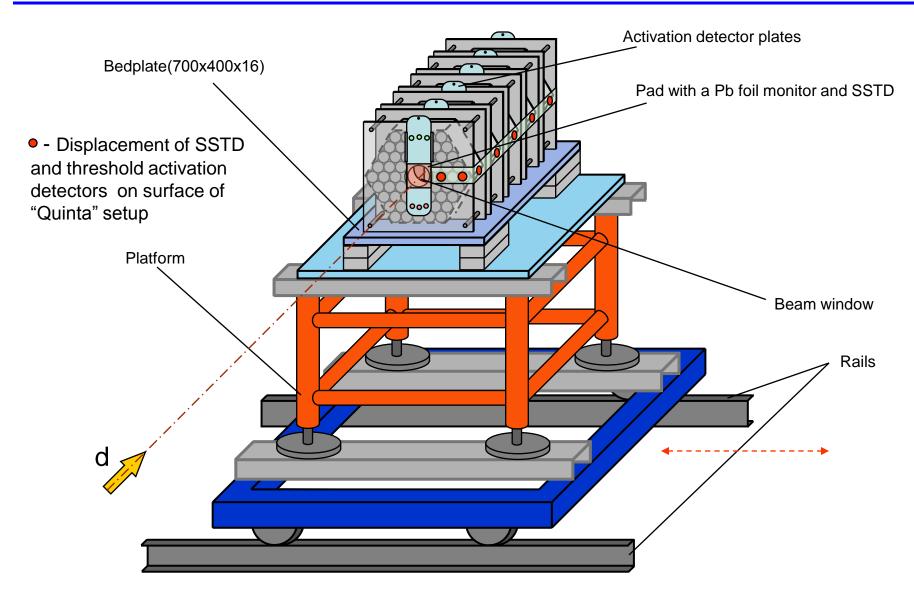
Conclusions from DN measurements by June 2009

- A comparison of the relative DN yields for deuteron energy 1 and 4 GeV has led to very challenging conclusion:
- Relative DN yield normalized to one incident deuteron for uranium target increases in about 8 times for this range of incident energy!
- If DN yield is proportional to integral number of fissions in the target assembly so beam power gain increases with its energy?!
- This result must be verified by direct measurement of total number of fission events within target for range of deuteron energy

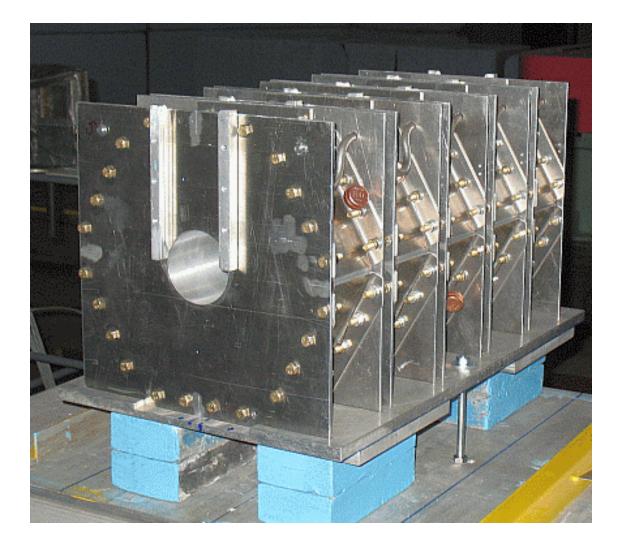
Scheme of experiment with upgraded "Quinta" setup, March 2011



Layout of upgraded target assembly "Quinta" at the irradiation position



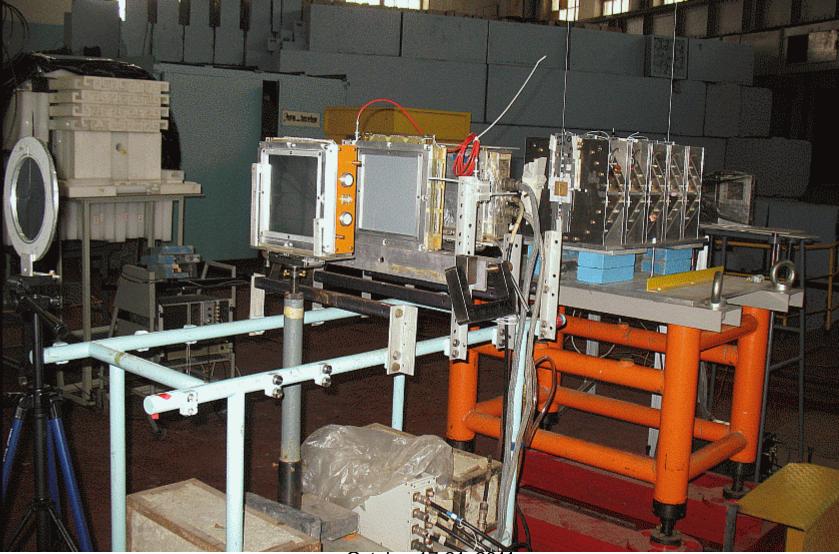
Upgraded target assembly "Quinta" (front view)



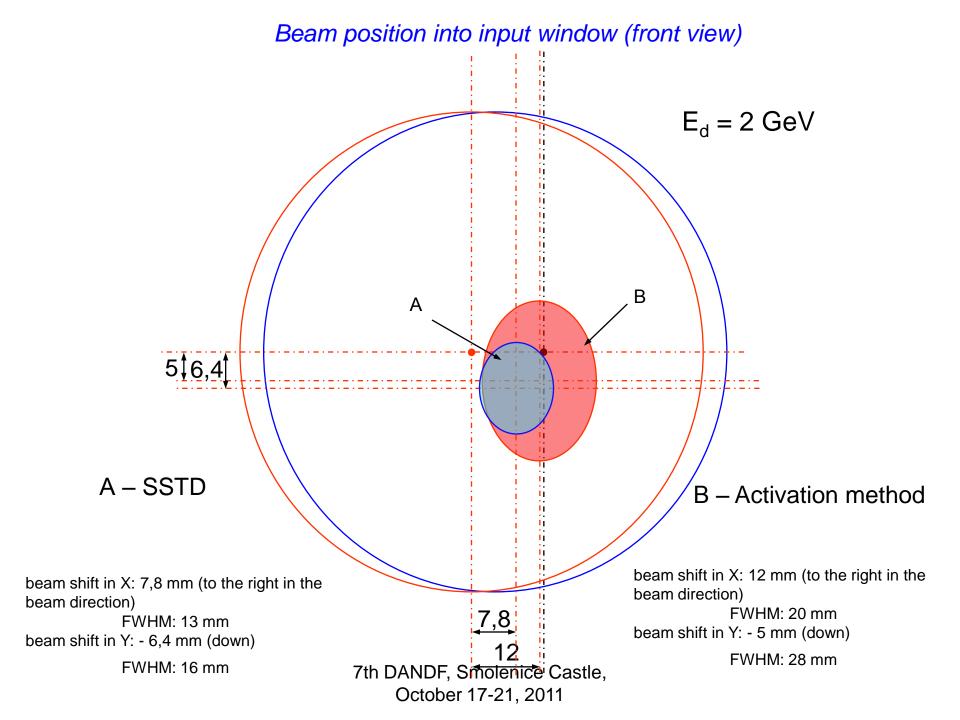
"Quinta" at the irradiation position (front view)



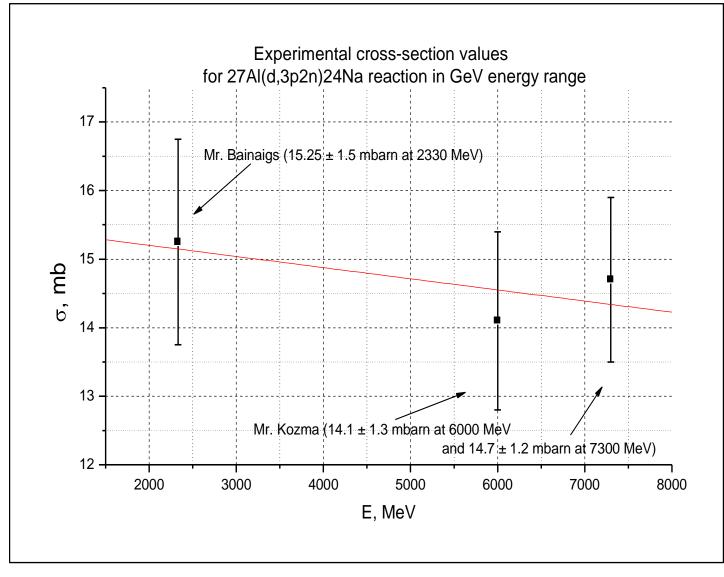
General view of the experimental setup



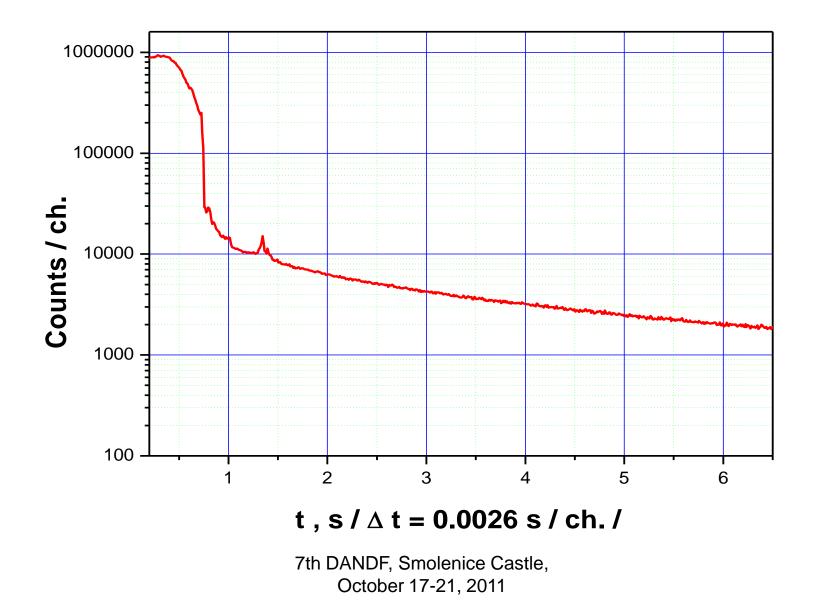
October 17-21, 2011



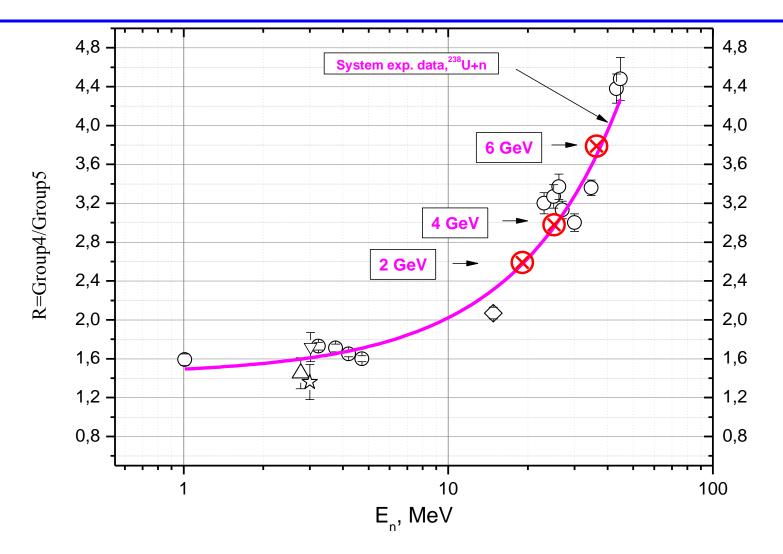
Calibration of monitors



<u>Neutron time spectrum at Ed= 6 GeV (March 2011)</u>

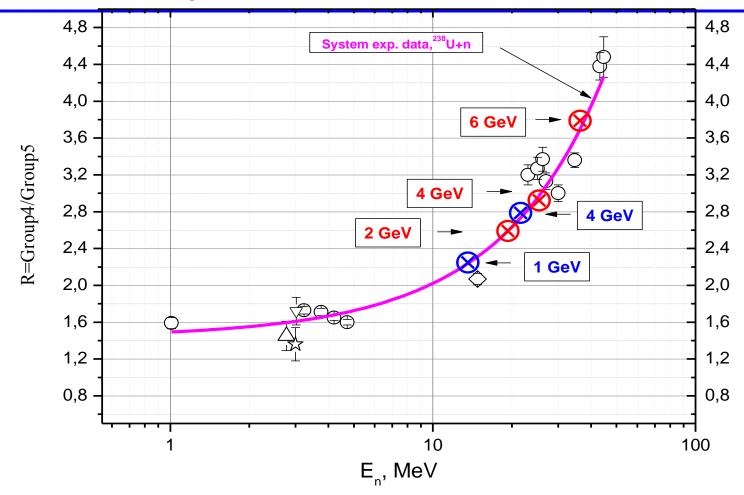


Analysis of DN time spectra measured by March 2011



Comparison of neutron energy dependence of the weight ratios of 5-th to 4-th DN groups from ²³⁸U(n,f)-reaction and similar values extracted from the DN time spectra measured in March 2011

Analysis of DN time spectra from June 2009 and March 2011 runs



Comparison of neutron energy dependence of the weight ratios of 5-th to 4-th DN groups from ²³⁸U(n,f)-reaction and similar values extracted from the DN time spectra measured in June 2009 and March 2011

So "mean neutron energy" <*En*> inducing fission of target nuclei ²³⁸U

increases from $\langle E_n \rangle = (13 \quad 3) \text{ MeV}$

up to
$$< E_n > = (37 \pm 4) MeV$$

with growth of incident deuteron energy

from $E_d = 1 \text{ GeV}$ to $E_d = 6 \text{ GeV}$

Ed	2 GeV	4 GeV	6 GeV
DN relative yield/ per	20 ± 1.4	69 ± 4.9	120 ± 8.4
one deuteron (arbitrary units)			

Gamma-activity method in March 2011 run Plutonium production

²³⁸U(n,γ)²³⁹U (23,54 min) β- →²³⁹Np (2,36 days) β- →²³⁹Pu 277,6 keV γ-line from ²³⁹Np

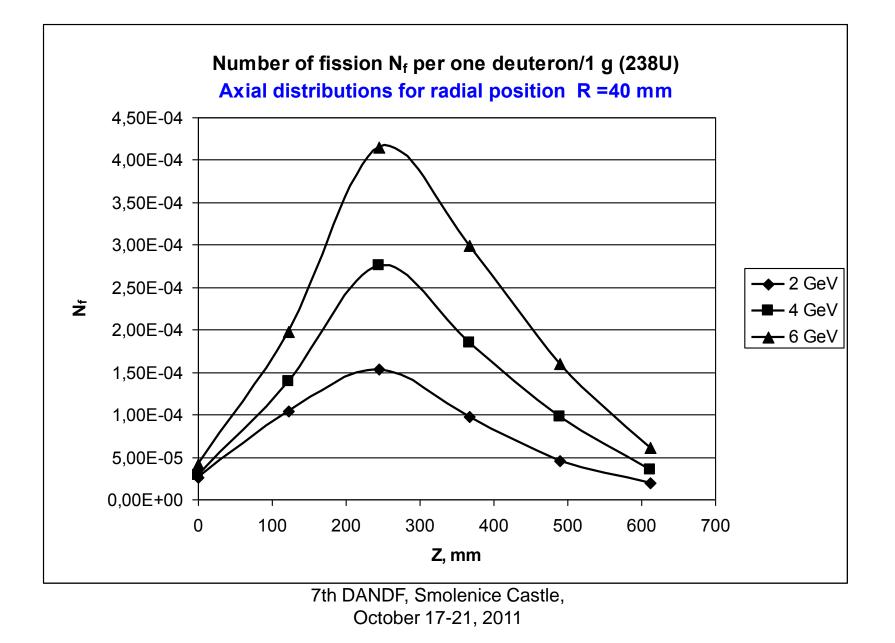
γ- detector calibrated with ⁶⁰Co, ⁵⁴Mn, ⁵⁷Co, ⁸⁸Y, ¹⁰⁹Cd, ¹¹³Sn, ¹³³Ba, ¹³⁷Cs, ¹³⁹Ce, ¹⁵²Eu, ²²⁸Th, ²²⁶Ra standard sources.

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

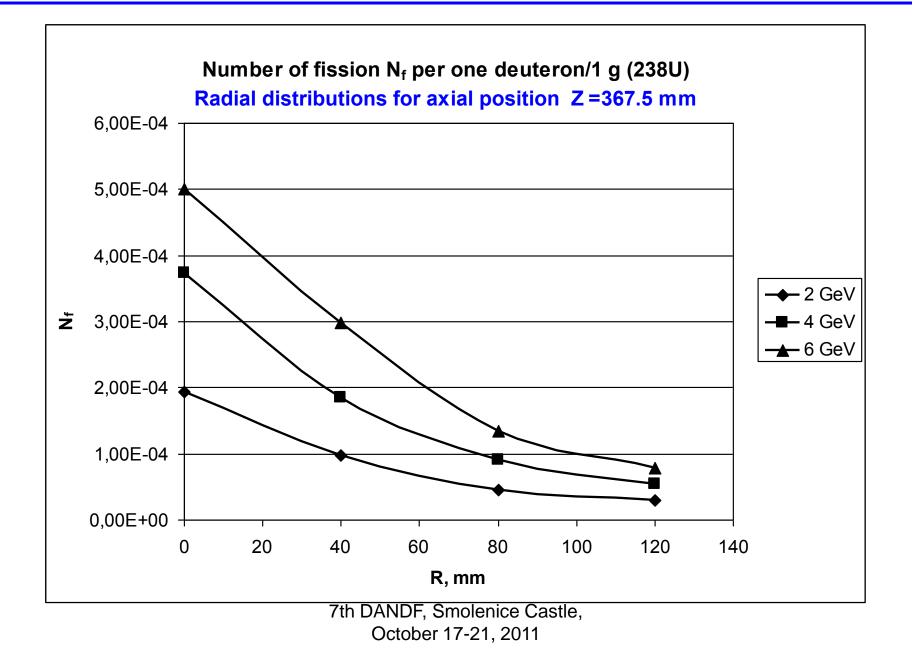
⁹⁷Zr (5.42%), ¹³¹I (3.64%), ¹³³I (6.39%), ¹⁴³Ce (4.26%)

In brackets there are mean cumulative FP yields

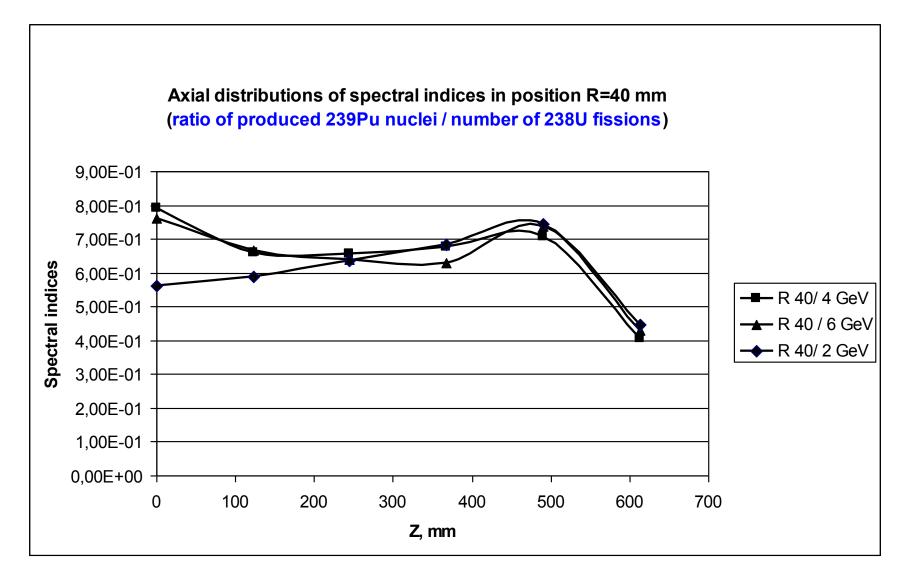
Gamma-activity method in March 2011 run



Gamma-activity method in March 2011 run



Gamma-activity method in March 2011 run

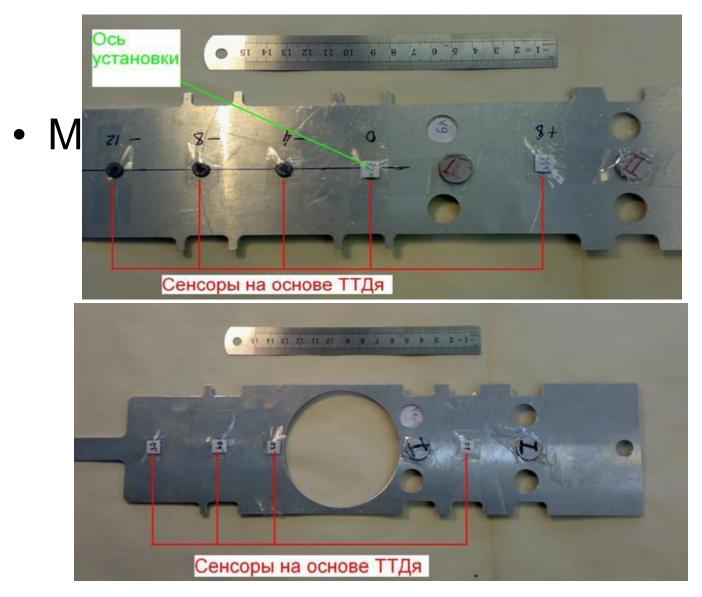


Results&Analysis - Gamma activity method

(numbers per one deuteron)

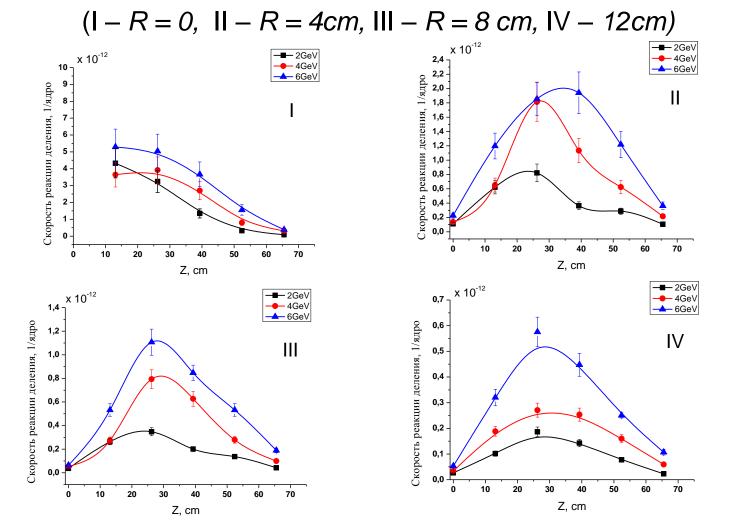
Ed	2 GeV		4 GeV		6 GeV	
²³⁹ Pu production	17	1,7	30	3,0	45	4,5
²³⁸ U(n,f)	21	3,6	36	3,6	54	5,4

Measurements with solid state track detectors (SSTD)

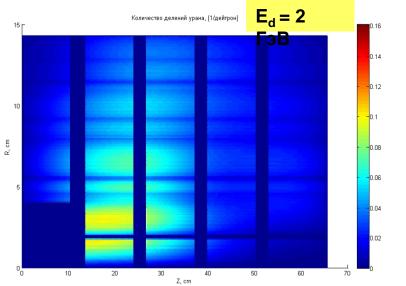


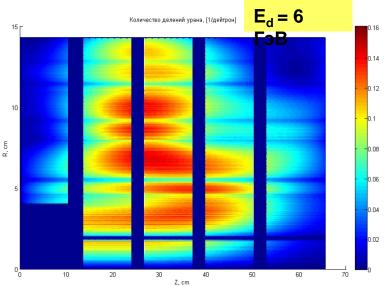
Mica SSTD

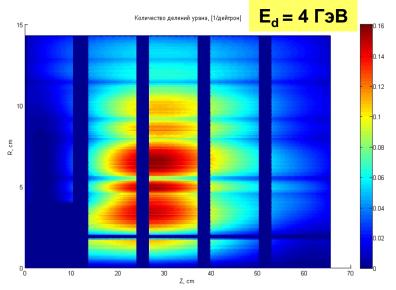
Axial distribution of fission rate per one deuteron measured by SSTD



Preliminary results of March 2011 experiments







Spatial distributions of fission rates (in cm³/one deuteron) for QUINTA setup irradiated by deuteron beam

Measurements by solid state track detectors (SSTD)

Discussion of March 2011 results

Number per one deuteron

Ed	0,66 GeV (Vassil'kov et al)	2 GeV	4 GeV	6 GeV	
²³⁹ Pu production	46 4	17 1,7	30 3,0	45 4,5	
²³⁸ U(n,f)	18,5 1,7	21 3,6	36 3,6	54 5,4	
²³⁸ U(n,f)		20.4 3,8 2	32 6 3.2	44 7 4.4	
DN relative yield normalized to fission data at 2 GeV		20 ± 1.4	69 ± 4.9	120 ± 8.4	

Intermediate conclusion

The preliminary results obtained for upgraded Quinta setup irradiated by deuterons with energy 2, 4 and 6 GeV.

Time spectra of DN was measured and analyzed. Extracted "mean neutron energy" inducing fission of ²³⁸U increases with growth of incident deuteron energy.

Relative DN yield goes up 6 times with increase of E_d from 2 up to 6 GeV in qualitative accordance with our previous (June 2009) measurements.

Total numbers of fission for the same range of E_d increase only ~ 3 times, proportionally to incident energy growth.

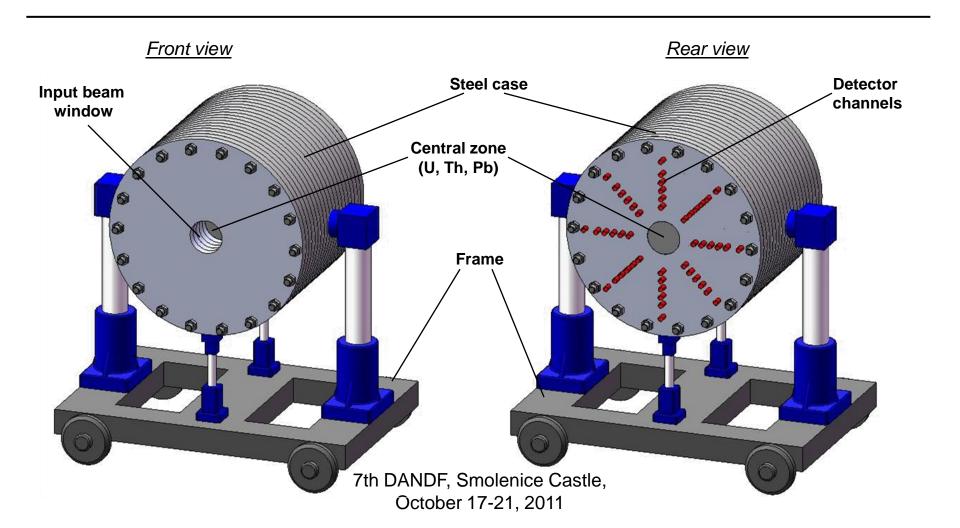
Difference in incident energy dependence of DN relative yields and total fission rate is the subject for further experimental and theoretical study.

Obtained data for spatial distribution of fission rates shows that energy release increases with growth of incident energy and radial size of multiplying target

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

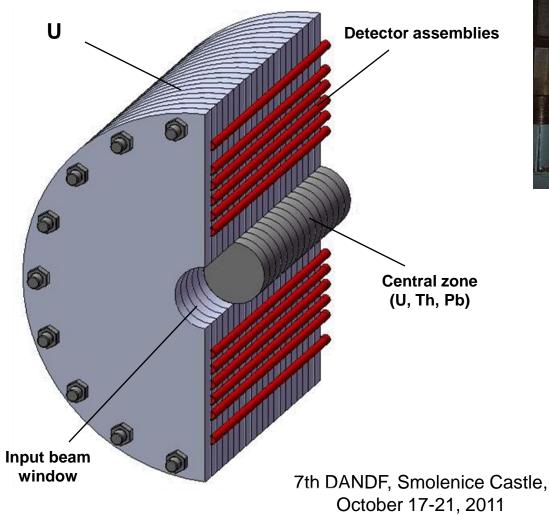
Mass of uranium	– 22 т.
Diameter	– 1,2 м.
Length	– 1 м.

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

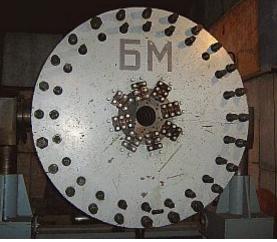


Quasi-infinite depleted uranium target (QIUT) with replacement central zone

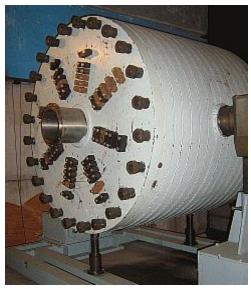
Longitudinal section of the QIUT together with central zone and detector sets



Front view



<u>Rear view</u>



Project "E&T – RAW" - Research program

Measurements will be done with protons (deuterons) in range of incident energy 1 to 10 GeV (0.5 up to 5 GeV/nucleon)

Task 1 . Integral data.

It includes wide set of experiments with the targets **QINTA** and "QIUT":

- study of spatial distributions with and without a graphite reflector (below for different target configurations):
 - of neutron spectra within the target volume and spectra of leakage neutrons;
 - of fission rates and transmutation cross sections of actinide fission products
 - of radiative capture (n, γ) and (n,xn)- reactions in the samples of long-lived isotopes from spent

fuel placed in measurement channels;

- of accumulation and burn-up of ²³⁹Pu aimed at evaluation of main parameters of its "equilibrium"

concentration - the value and a necessary irradiation time to reach it;

- of heat release;

- study of prompt neutron spectra and multiplicity, the delayed neutron time yields for different target configuration as well as beam particle type and energy;
- measurements of GBP in dependence on incident particle type and its energy for different target configurations;
- improvement and optimization of on-line and off-line methods for monitoring intensity, geometric characteristics and position on the target of the Nuclotron beam;
- study of integral decay rates of target irradiated with different doses.

Project "E&T – RAW" - Research program

Task 2 . Nuclear data.

- It includes the series of measurements of fission cross sections for relevant set of target nuclei and delayed neutron yields.
- For reliable simulation of AD systems it is necessary to know the characteristics of corresponding reactions in both thin and thick (≥2000 g/cm2) targets.

Task 3. Simulation.

- It is aimed at an improvements of underlining physical models and the constant databases of the computer codes designed to describe multiple particle production in a quasi-infinite ADS active cores for incident energy up to 10 GeV per nucleon.
- An appropriate account of high energy fission channels is of great importance for calculation of neutron fields and heat release in such systems, because the present options of these codes could not reproduce even qualitatively the respective experimental data obtained up to now.
- An implementation of this task provides a theoretical support of the experimental part of the research project program and helps to improve a planning of subsequent experiments

Project "E&T – RAW" - Research program

Task 4 . Materials

Investigation of relativistic beam impact on structural and fuel materials.

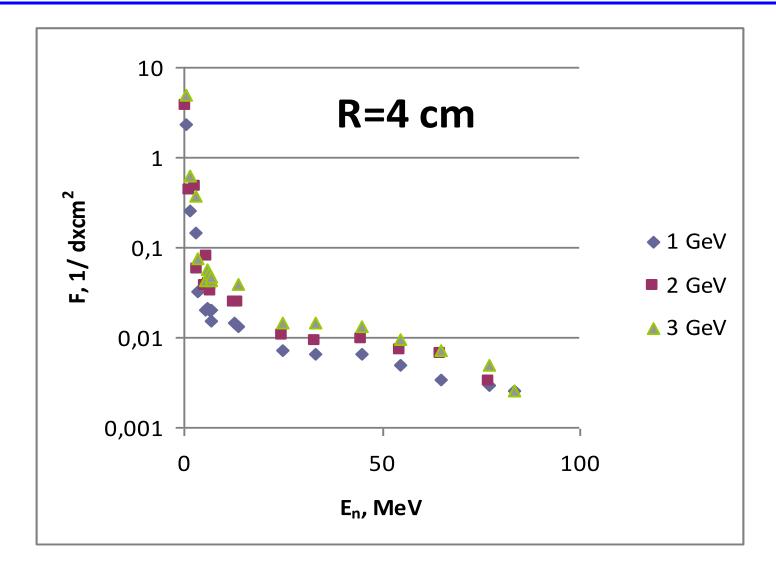
- It is planned to measure of the gas (^{3,4}He) production rates in interaction of relativistic beams and fast neutrons with the construction elements and the fuel.
- Radiation damage depending on the energy and type of primary particles will also be studied.
- For this task it is necessary to form a minimal size of Nuclotron beam on the target.
- Our project is open for all collaborators interested in its main goals

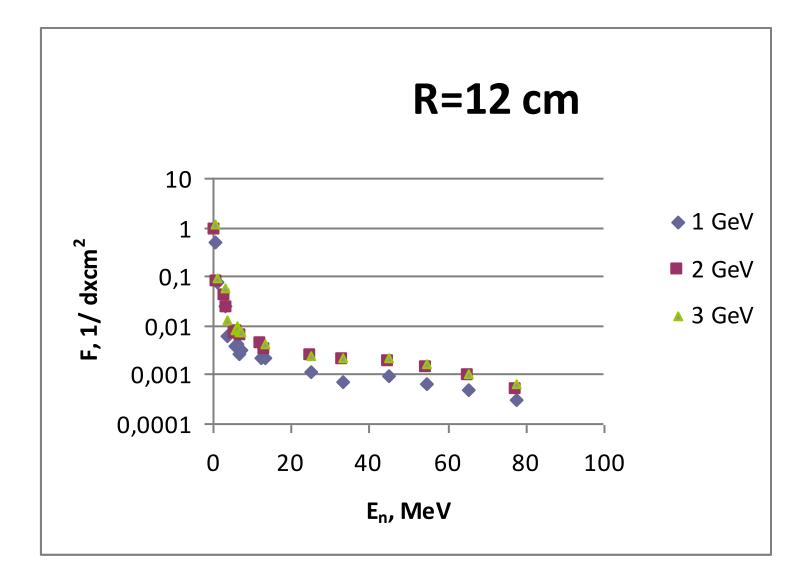
Thanks for your attention



 Our project is open for all collaborators interested in its main goals

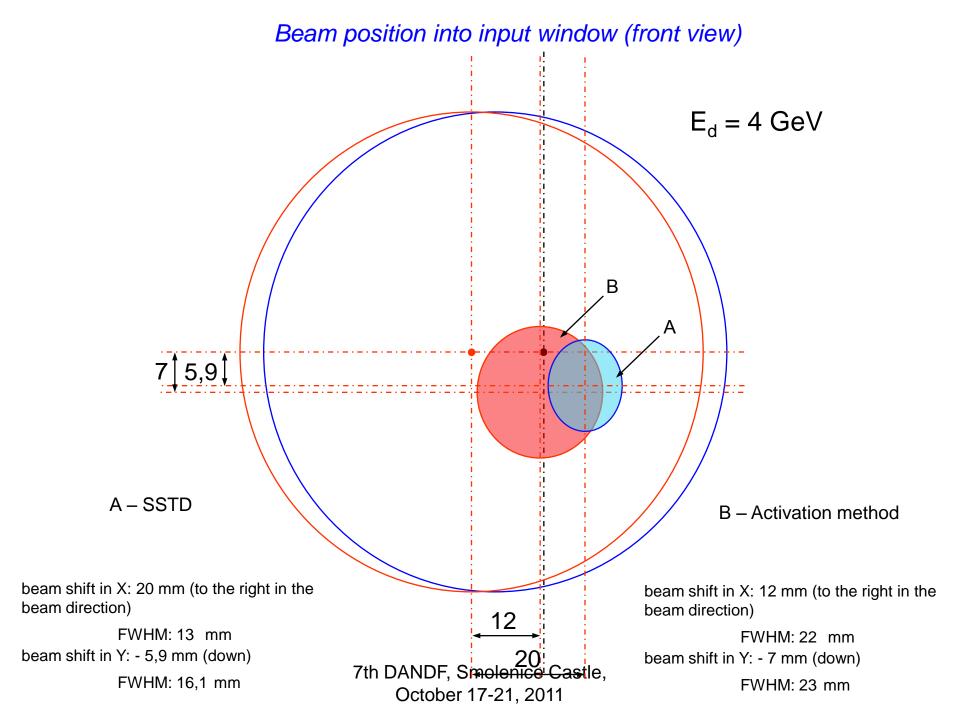
Results from threshold activation detectors (March 2011)

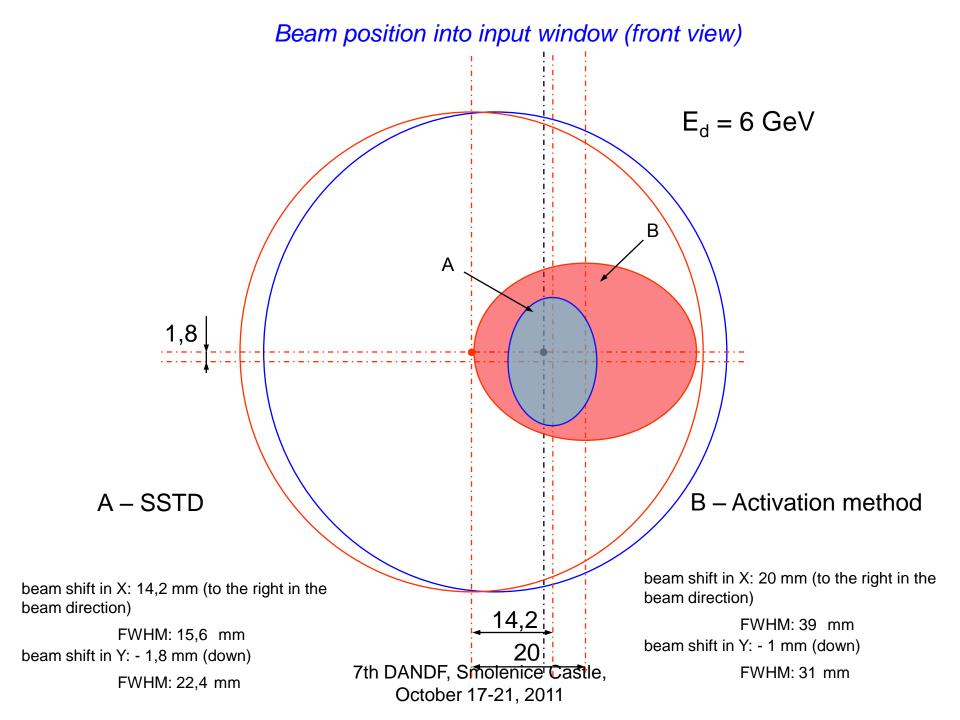


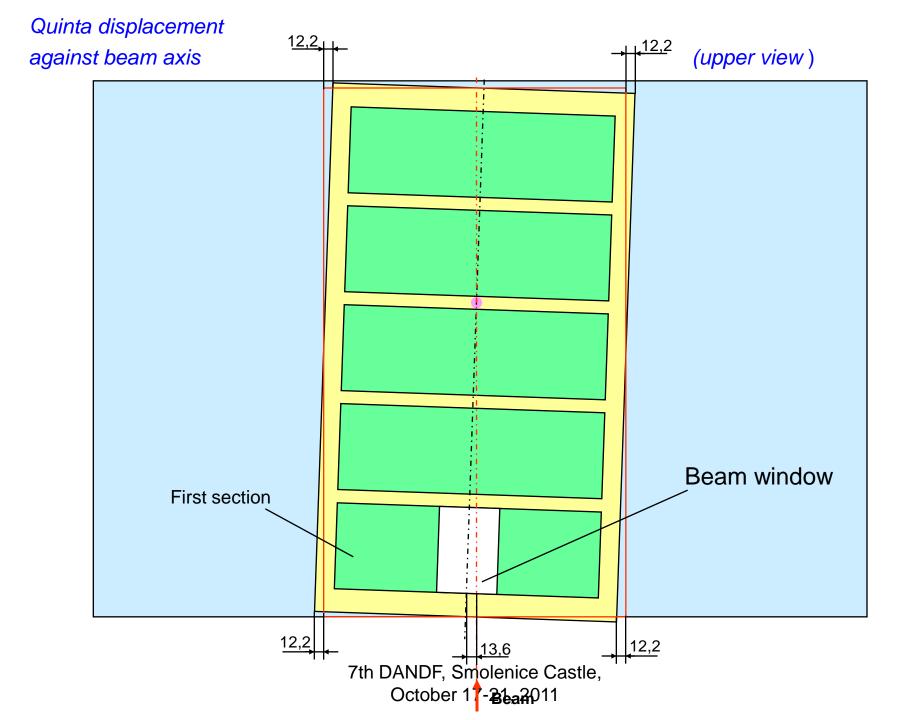


Rates I of 47 Ti(n,p) 47 Sc reaction (at Z = 27 cm, second gap) (March 2011 run)

E _d	R=4 cm		R=12	2 cm	R=17,5 cm (<i>Quinta surface</i>)		
	<i>I</i> , 1/d <i>I</i> , 1/d / GeV		<i>I</i> , 1/d	<i>I</i> , 1/d / GeV	<i>I</i> , 1/d	<i>I,</i> 1/d / GeV	
	•10 ⁻²⁷	• 10 ⁻²⁷	• 10 ⁻²⁷	• 10 ⁻²⁷	• 10 ⁻²⁷	• 10 ⁻²⁷	
2 GeV	16,8	8,40	2,94	1,47	0,79	0,40	
4 GeV	37,4	9,30	5,49	1,37	1,36	0,34	
6 GeV	44,7	7,50	7,16	1,19	2,30	0,38	







Project "E&T – RAW"

 More detailed information on program of future measurement in the framework of the project is presented in

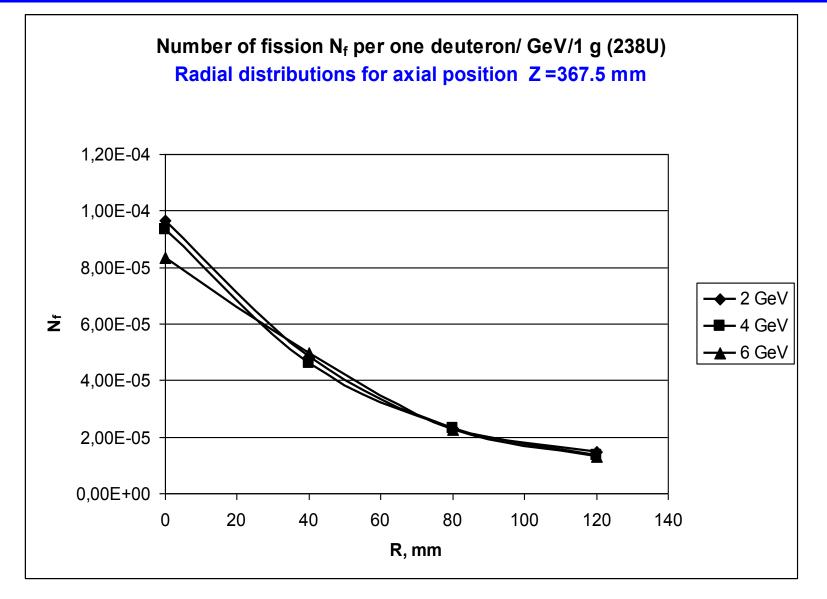
J.Adam et al. "Study of deep subcritical electronuclear systems and feasibility of their application for energy production and radioactive waste transmutation" («E&T – RAW» Collaboration), JINR Communication E1-2010-61.

- Our project is open for all collaborators interested in its main goals
- We hope that this project has serious innovation potential.

Thanks for your attention

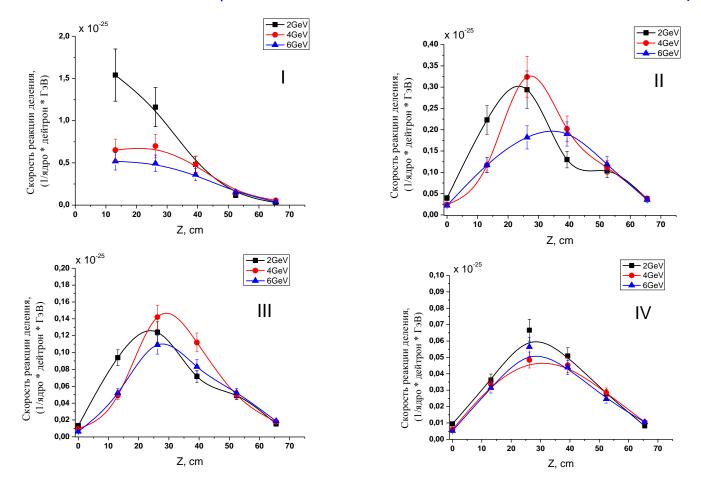
Experimental program of March 2011 run

- 1. Verification of incident energy dependence of total DN yields obtained in June 2009;
- 2. Study the incident energy dependence of the spatial and energy distributions of neutrons (by threshold activation detectors), spatial distributions of numbers of fission and plutonium production (by SSTD (mica) and γ activity of specific fission and (n, γ)- products), as well as the spectra and multiplicity of leakage neutrons;
- 3. Direct determination of total fission numbers within Quinta setup by two independent methods aimed at determination of the dependence of the beam power gain on its energy ;
- 4. Obtaining a wide set of experimental data to proceed with the modification of intra nuclear cascade models and respective MC codes aimed at improvement the reliability of predicting outcomes of future experiments with large (~ 20 t) uranium target under the "E & T –RAW" project.



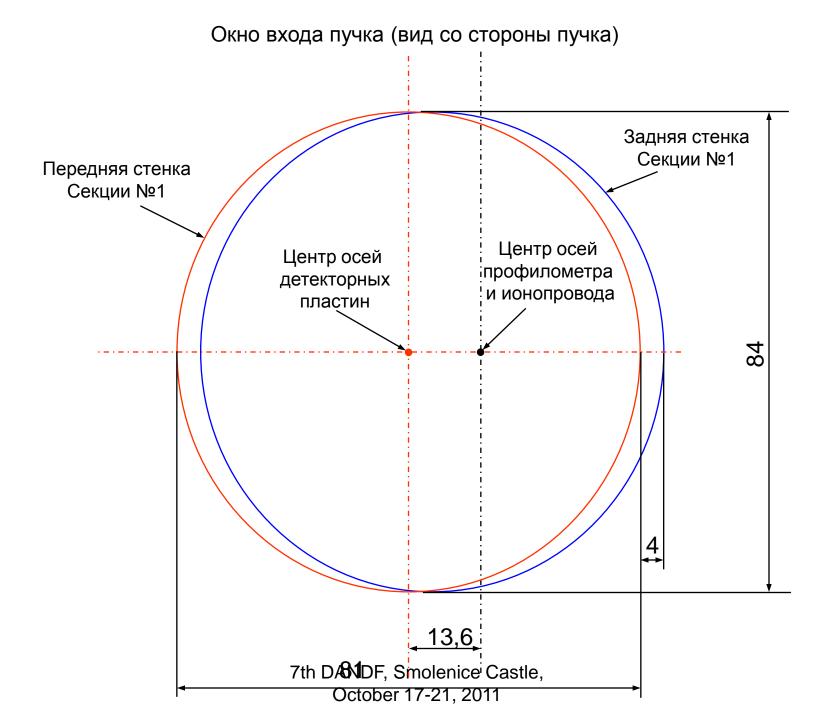
Axial distribution of fission rate per one deuteron per GeV measured by SSTD

(I - R = 0, II - R = 4cm, III - R = 8 cm, IV - 12cm)



SST fission detectors (fission numbers per deuteron)

	Ed			Ed			Ed		
	2 GeV			4 GeV			6 GeV		
²³⁸ U(n,f)	20.4	3,8	2	32	6	3.2	44	7	4.4

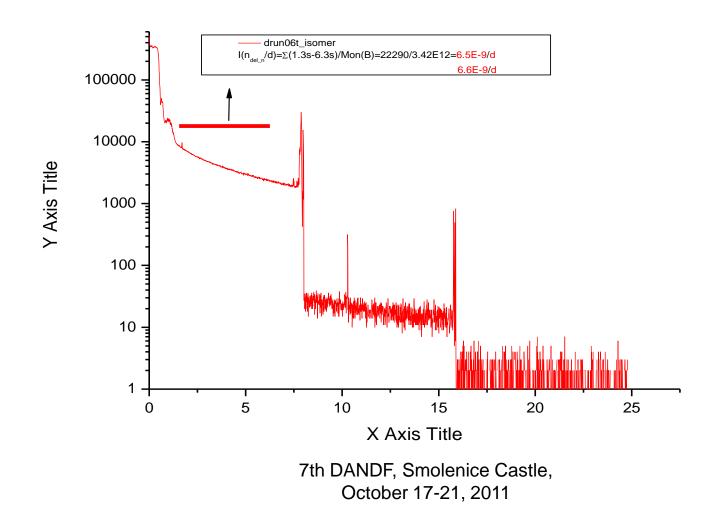


MOTIVATION

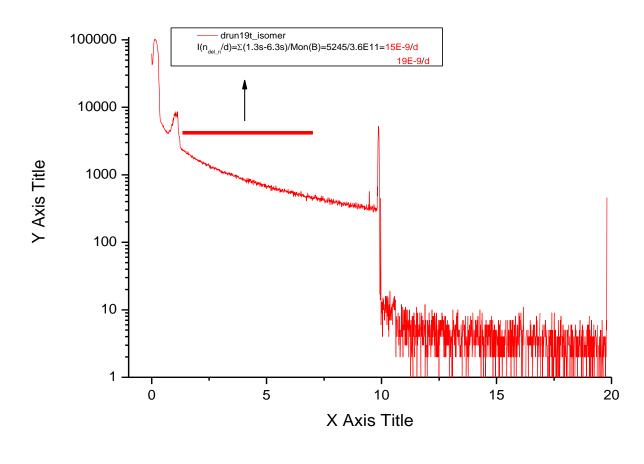
- ADS is considered now as tool for the transmutation of the long-lived components of radioactive wastes (RAW) and in principle for the solution of global energy problems.
- This work is aimed at study of basic physics of the ADS with quasiinfinite size active core (AC) from natural uranium irradiates by deuteron beam of (1-6) GeV energy.
- The long-range goal is a study of the possibilities of such ADS with very hard neutron spectrum inside of AC to realize recently proposed so called Relativistic Nuclear Technology (RNT) for transmutation RAW and energy production due to burning of AC material.
- Important aim of the experimental program is to provide comprehensive benchmark data set for verification and adjustment of INC models and transport codes.

Neutron time spectrum

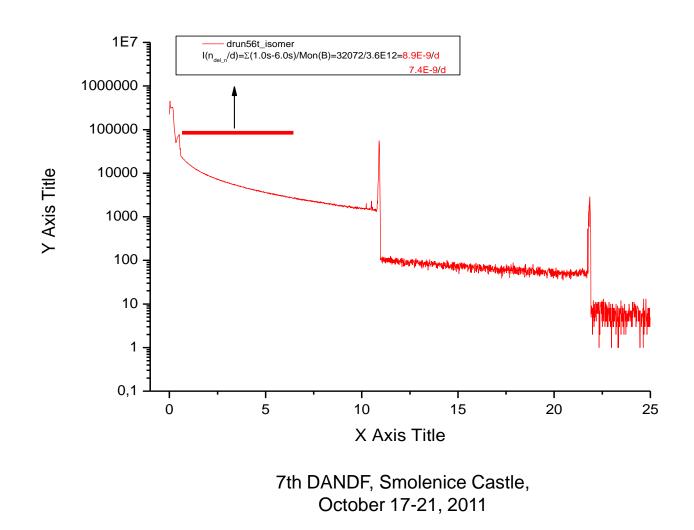
$E_d = 2 \text{ GeV}, (T = 8 \text{ s}, \Delta T = 0.65 \text{ s}), \text{ total DN yield I} = (1.3\pm0.1) \cdot 10^{-9}/d$ March 2011 run



 E_d = 4 GeV, (T = 10 s, ΔT = 0.34 s), total DN yield I = (4.5±0.3) 10⁻⁹/d



Neutron time spectrum $E_d = 6 \text{ GeV}, (T = 11 \text{ s}, \Delta T = 0.25 \text{ s}), \text{ total DN yield I} = (7.8 \pm 0.6) \cdot 10^{-9}/d$



Results&Analysis