# DETERMINATION OF THE NUMBER OF NUCLEAR FISSION $^{238}$U AND THE ACCUMULATION $^{239}$PU IN $^{NAT}$U / PB - ASSEMBLY OF THE ASSEMBLY "ENERGY PLUS TRANSMUTATION", BOMBARDED BY RELATIVISTIC DEUTERONS

# EXPERIMENTAL AND MONTE CARLO STUDIES ON THE CALIBRATION FACTOR OF THE MICA DETECTORS WITH A LEAD RADIATORS IN THE EXPERIMENTS ON «ENERGY PLUS TRANSMUTATION» SETUP


**Presenter: Igor Zhuk** zhuk@sosny.bas-net.by

*Joint Institute for Power and Nuclear Research,*

*220109, Minsk, Belarus*

Co-authors

A.S. Potapenko, A.A. Safronova,

*Joint Institute of Power and Nuclear Research, 220109 Minsk, Belarus.*

S.R. Hashemi-Nezhad,

*School of Physics, A28, University of Sydney, NSW 2006, Australia.*

M.I. Krivopustov,

*Joint Institute of Nuclear Research, 141980 Dubna, Russia.*

V.A Voronko, V.V. Sotnikov

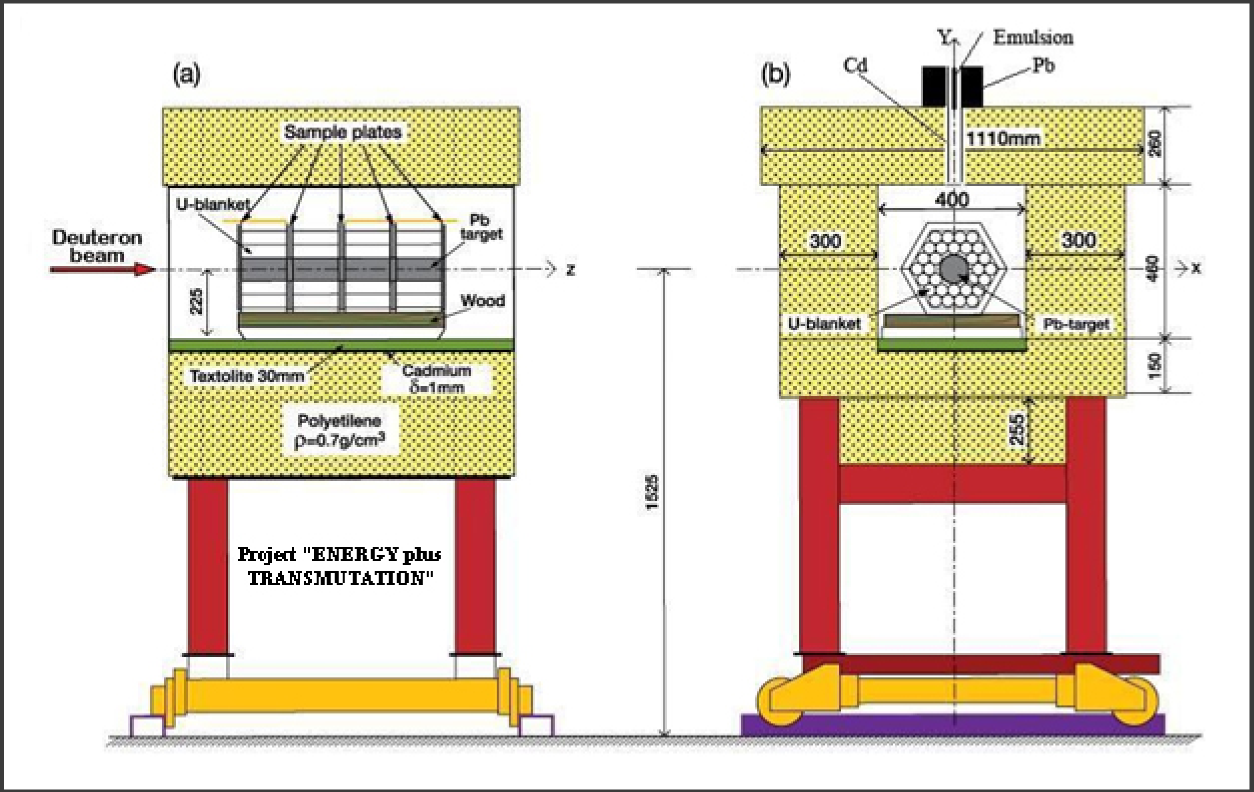
*National Scientific Center Kharkov Institute of Physics and Technology NASU, 61108 Kharkov, Ukraine*

# OUTLINE

- **Experiment : Determination of the calibration factor of the mica detectors with a lead radiators in the experiments on «Energy plus Transmutation» setup**
  - *Calibration factor of "Lead + Mica" sensor*
  - *Applications*
- **Experiment : Neutron generation in the «Energy plus Transmutation» setup**
  - *Experimental techniques*
  - *Fission rates determination*
  - *Capture reaction rates determination*
  - *Comparison with the experiment on the "LEAD BLOCK" target (professor Tolstov et.al.)*
- **Monte-Carlo simulations: codes, comparison with experiments**

EXPERIMENTAL SETUP

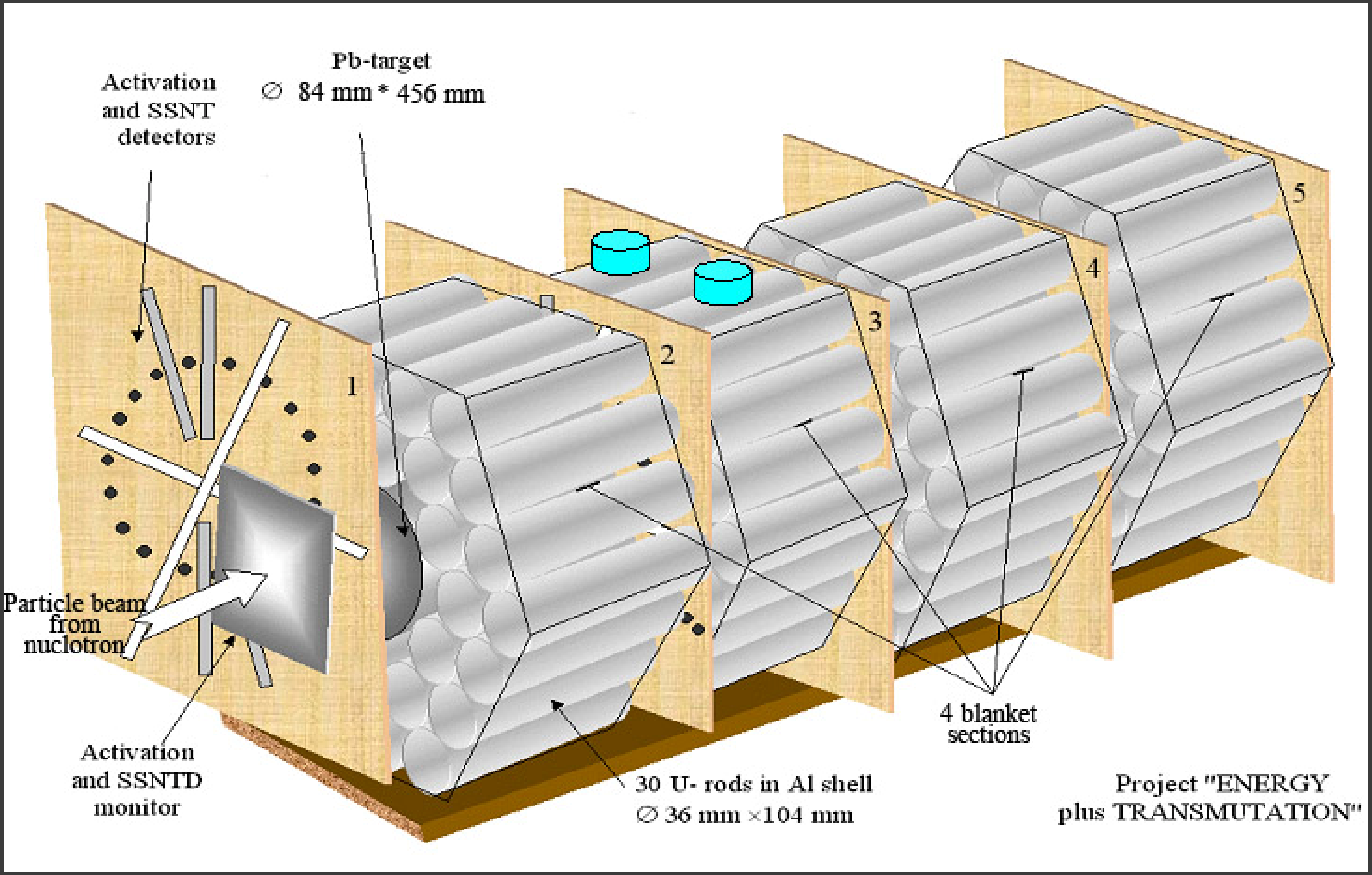
# Experimental setup




1) Cylindrical lead targets with diameter 8.4 cm and length 45.6 cm.
2) A natural uranium blanket surrounds the target.
In the experiment these blanket sections were located one after another; the front of the first blanket section and the front face of the lead target were in the same plane. Blanket sections consist of uranium rods (metal natural uranium packed into aluminum cladding), with diameter 3.6 cm, length 10.4 cm and weight 1720g. Each blanket section contains 30 uranium rods with weight 51.6 kg. Thus the 4 blanket sections contain 120 uraniu rods with total weight 206.4 kg. There is a 0.8 cm gap between the blanket sections to be used for detectors.
3) The whole target-blanket system was placed within a wooden container filled with granulated polyethylene. The inner walls of the container were covered with a Cd foil of thickness 1 mm.

EXPERIMENTAL SETUP

# Target + blanket assembly




Scheme of the four-section "Energy plus Transmutation" setup with a massive lead target and uranium subcritical blanket. The placement of transmutation samples in presented at the surface of the second section of U-banket.

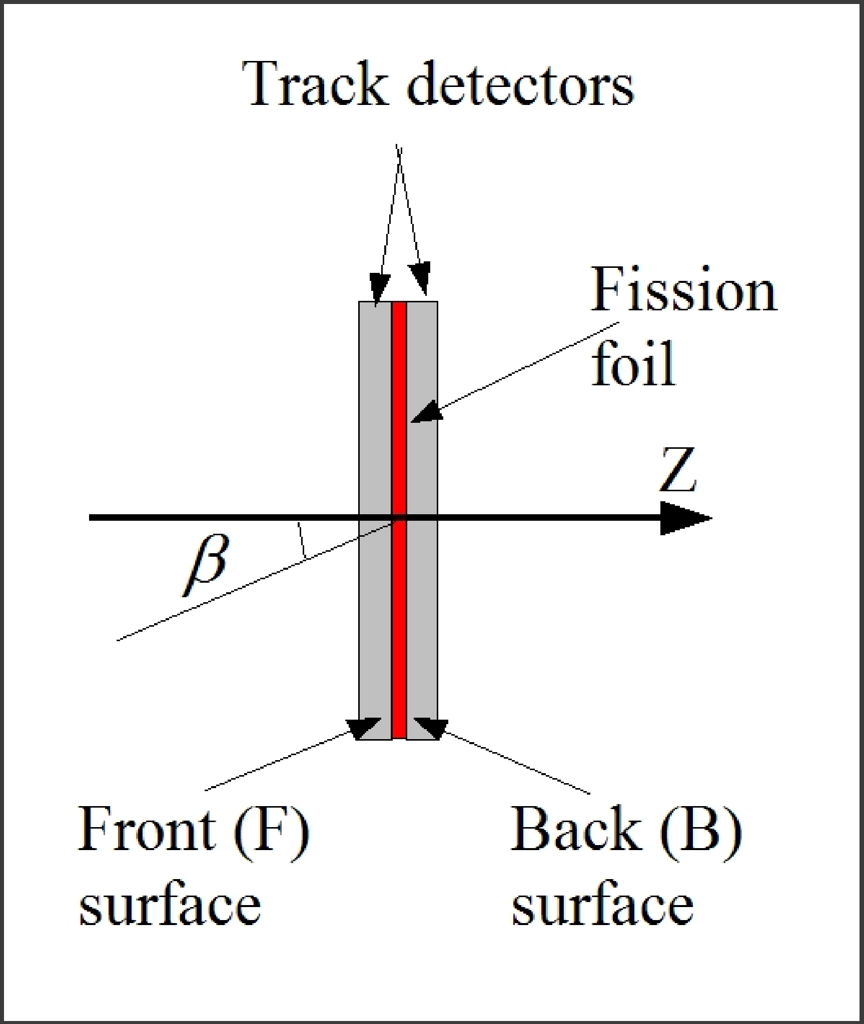
EXPERIMENTAL SETUP

# Haw it's looks

**Subcritical assembly + biological shieldi**

# SSNTD : concept






The basis of fission reaction rates measurement using solid nuclear track detectors (SSNTD) is the parity between the density of tracks formed on a surface of the detector, (irradiated at close contact to a radiator which is a source of fission fragments), and investigated neutron flux.

Reaction rate is proportional to a track density on the detector.

S
S
N
T
D

# SSNTD : how it's looks

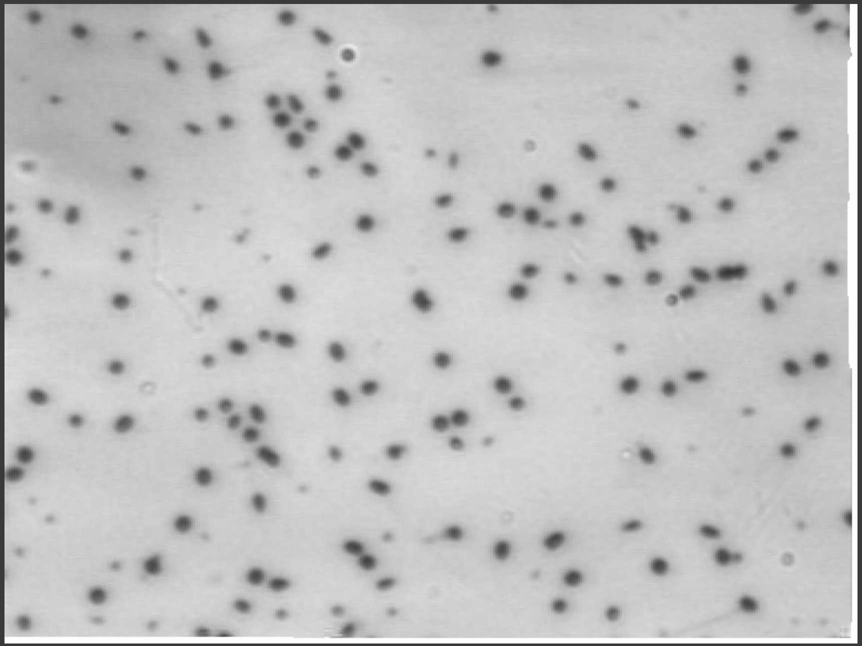
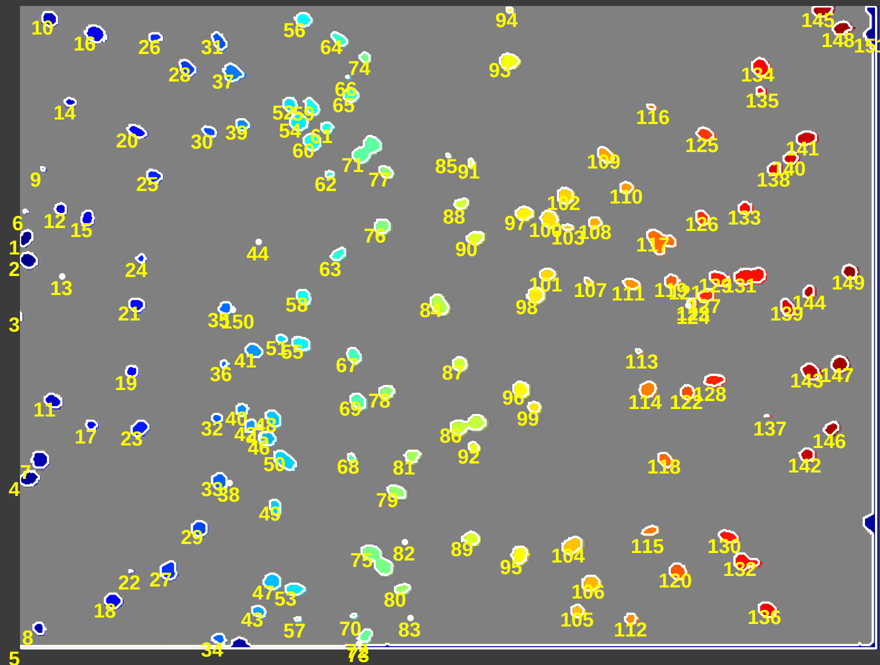


**Image from an optical microscope: surface of irradiated detector after etching**

… and the image after computer processing



SSNTD THEORY

# SSNTD theoretical background

The number of tracks on the detectors is given by the following formula:

$$N_q^i = A^i \mu^i \varepsilon_q d_q \rho_q P \int_0^\infty \sigma_f^i(E) \varphi^P(E) dE$$

$$k_q^{sens} = A^i \mu^i \varepsilon_q d_q \rho_q$$ - Calibration factor

The formula for calibration factor includes:

- number of fission fragments in fission reaction;
- part of charged particles reaching of the detector;
- efficiency of charged particle registration;
- thickness of the foil;
- nuclear density.

"Thick" radiators are characterized by a constant yields of fission fragments, therefore at measurement of fission rates and spectral indexes *using such radiators, individual calibration of each radiator in a standard neutron field is not required.*

$$\mu^i = \begin{cases} \frac{1}{2}\left(1 - \frac{d_q}{2\overline{R}_0}\right) & \text{for } d_q < \overline{R}_0 \\ \frac{1}{4} & \text{for } d_q = \overline{R}_0 \\ \frac{1}{4}\frac{\overline{R}_0}{d_q} & \text{for } d_q > \overline{R}_0 \end{cases}$$

It's the reason to use "thick" radiators:
R<<d

where R is a free length of fission fragment in a foil material and d is a foil thickness

SSNTD THEORY

# SSNTD theoretical background (continue)

$$Q_f^i = \int_0^\infty \sigma_f^i(E)\varphi^P(E)dE$$

Fission reaction rate (reaction integral)

$$Q_f^i = \frac{N_q^i}{k_q^{sens} P}$$

Evaluated formula for reaction rate

Calibration factor can be found knowing :

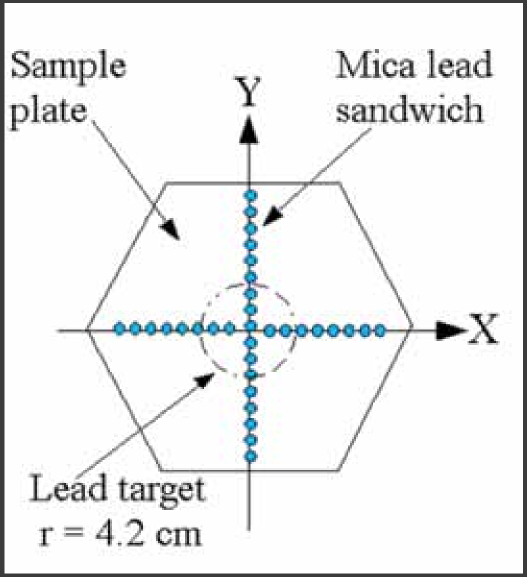
1. P – total particles flux
2. N – track density
3. Q – reaction rate

Problem is that fission cross section for non fissionable material (heavy metals) has energy thresholds in tens of MeV. It's very difficult and expensive to find neutron sources with such parameters.

!!! SOLUTION is to use high energy proton from accelerator

C A L I B R A T I O N

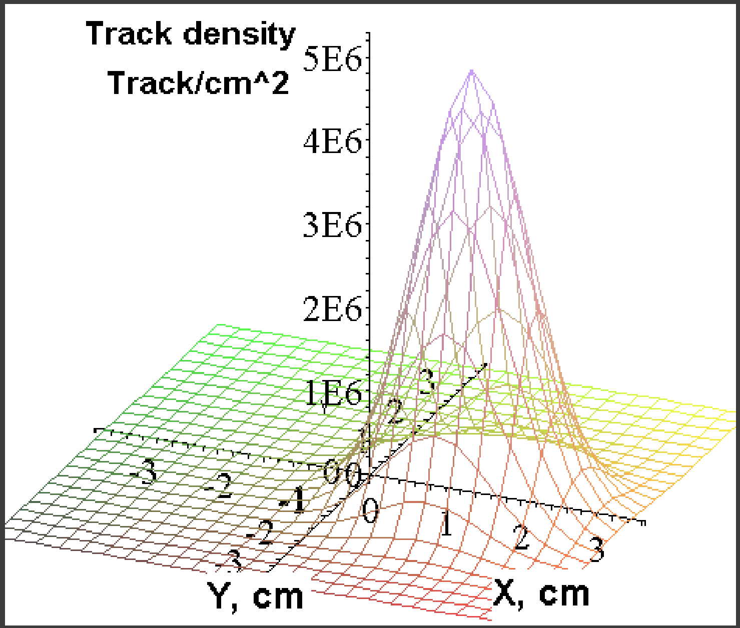
# SSNTD calibration using Nuclotron proton beams




Top of the assembly
And schematic drawing
of the experimental plate

The first experimental plate
with a lines of sensors.
Each sensor contains lead foi
and two track detectors are
from the both sides of the foil.

Gaussian approximation of experimental
dates from SSNTDs.
Track densities on the top of the assembly
is proportional to spatial distribution of
primaries particles



C A L I B R A T I O N

# Calibration factor: experimental and calculated results

**Experimental values of calibration factor for mica track detector and lead radiator obtained using proton beams from Nuclotron**

| E, GeV | Back/Face | $K^{sens}$; $\times 10^{-19}$ | Experimental uncertainty % |
|---|---|---|---|
| 0,7 | 1,31 | 1,35 | 12 |
| 1.0 | 1,40 | 1,21 | 12 |
| 1,50 | 1,49 | 1,34 | 15 |

Note: dimension of $K^{sens}$ is [track×cm$^{-2}$×neutron$^{-1}$]

Note: calibration factor is the average value for "Back" and "Face" sides of the sensor

Calculation of calibration factor for mica track detectors:

| Code | Radiator | $K^{sens}$; $\times 10^{-19}$ | uncertainty % |
|---|---|---|---|
| MCNPX | $^{nat}Pb$ | 1.15 | 13 |
| MCNPX | $^{209}Bi$ | 1.15 | 13 |
| FLUKA | $^{nat}Pb$ | 1.20 | 20 |
| MCNPX | $^{238}U$ | 0.99 | 3 |

see

Calibration of track detectors for fission rate determination: an experimental and theoretical study. // Nuclear Instrument and Methods: A568, p816-825, 2006. S.R. Hashemi-nezhad, I. Zhuk, A. Potapenko and M. Krivopustov

APPLICATION

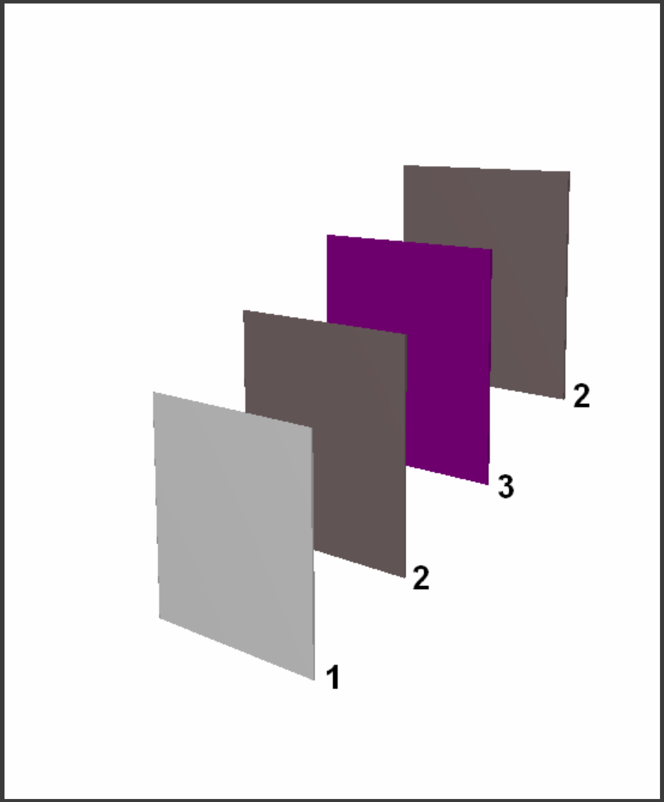
# Beam parameters

The beam profiles obtained using $^{Nat}Pb(p, f)$ reaction and mica track detector.
The proton beam fluence was determined using $^{27}Al(d, 3p2n)^{24}Na$ reaction. There is possibility to measure it using only SSNTD data. (not yet implemented)

| Energy | FWHM (cm) | | Center position (cm) | | Fluens |
|---|---|---|---|---|---|
| | X | Y | Xc | Yc | |
| 2.52 | 1.5±0.1 | 1.6±0.1 | 1.5±0.1 | -0.3±0.1 | $5.9\times10^{12}$ |
| 1.60 | 2.8±0.1 | 1.9±0.1 | -0.6±0.1 | -0.4±0.1 | $2.1*10^{13}$ |

A P P L I C A T I O N

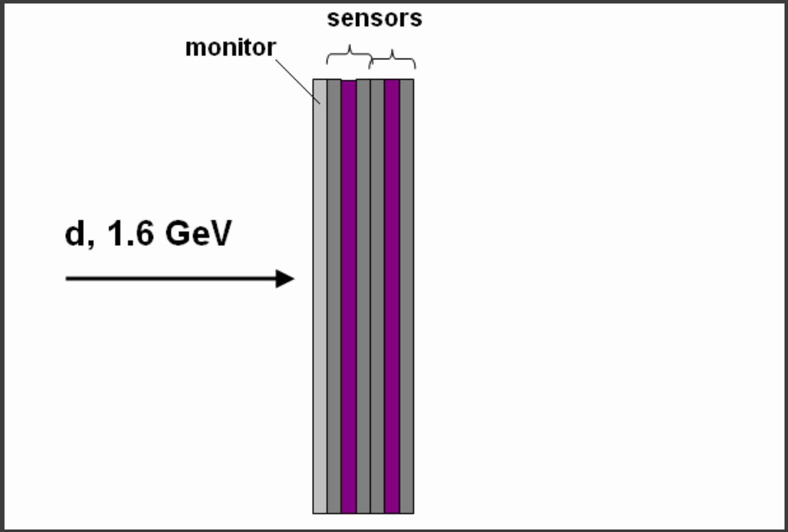
# Cross section measurements




1. Al monitor
2. SSNTD (artificial mica)
3. Radiator (source of fission fragments)

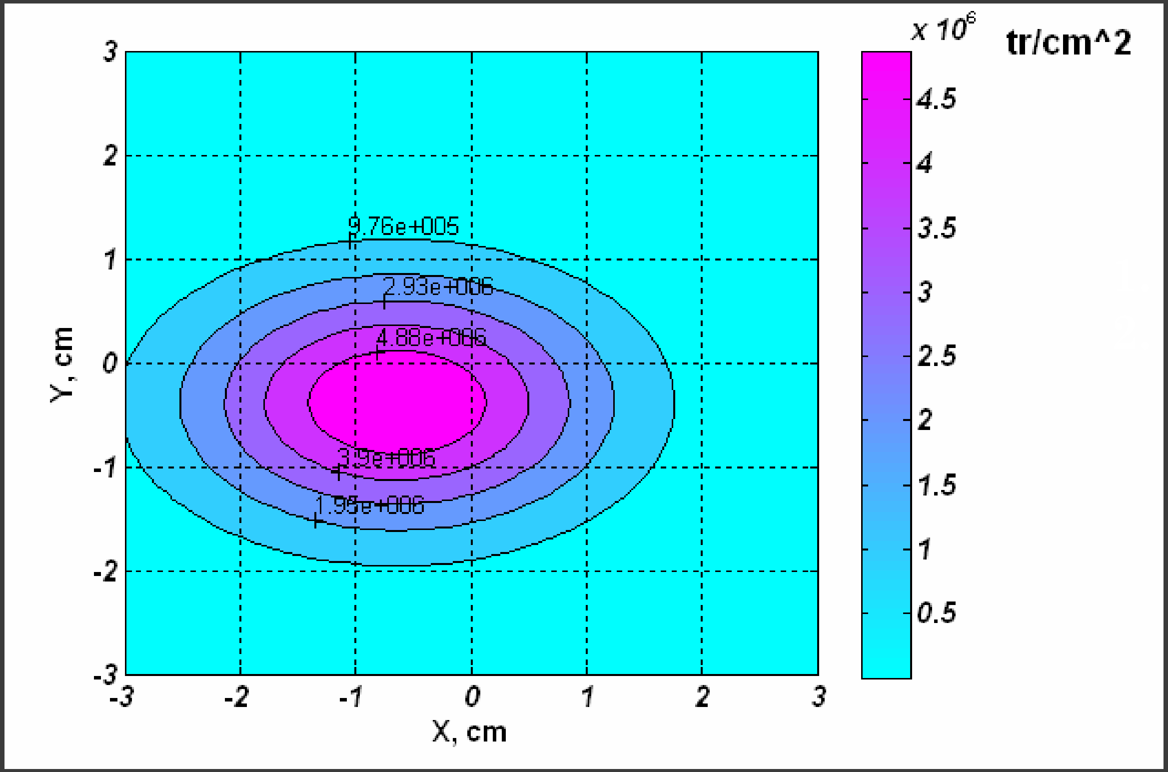
Size of Al foils:
12 x 12 x 0,1 mm

Sensors schematic drawing for
Fission cross section measurement



APPLICATION

# Cross section measurements




**Sensor location on the direct beam at deuteron energy 1.6GeV:**

**(x=0cm, y=0cm)**

**(x=-2cm, y=0cm)**

A structure (profile) of deuteron beam for Energy 1.6 GeV

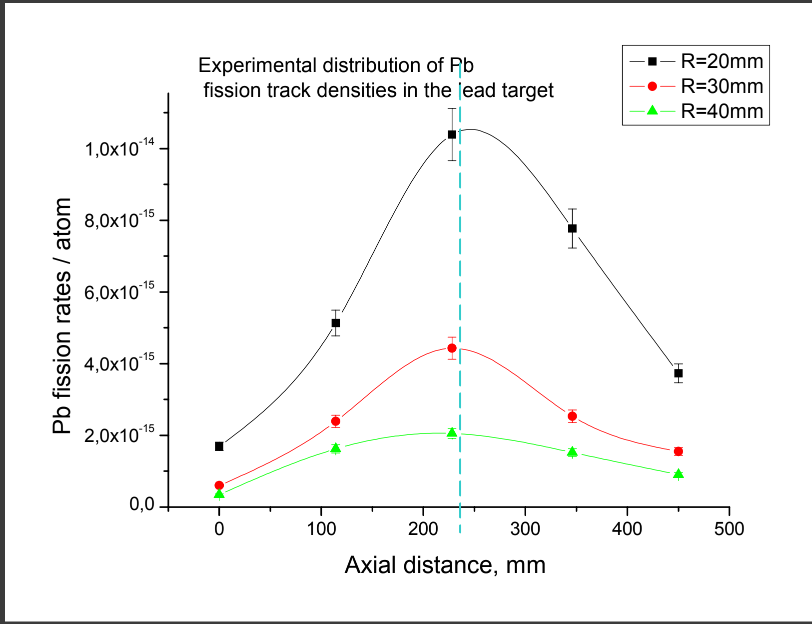
APPLICATION

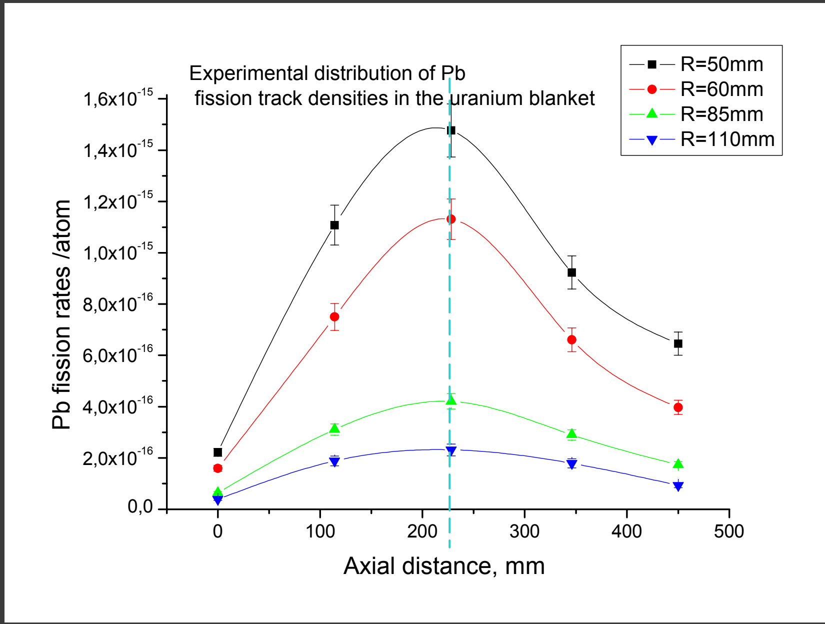
# Results of experimental measurements fission cross-section for deuteron energy 1.6GeV (SSNTD method)

| Samples | Fission cross-section for deuteron energy 1.60GeV **(experiment, present work) mb** | Fission cross-section for deuteron energy 2.1GeV **(experiment, EXFOR library) mb** |
|---|---|---|
| **U-nat** | **1700 ±300** | **1654 ± 340** |
| **Pb-nat** | **200 ± 50** | **182 ± 40** |
| **Bi-209** | **220 ±80** | **323 ± 60** |

APPLICATION

# Experiment: natural lead fission rates (SSNTD technique)




Axial distributions of fission rates for natural lead inside the Pb-target

Axial distributions of fission rates for natural lead in the Uranium blanket

Maximum of natural lead track densities distributions for lead target and uranium blanket is on the third plate (axial distance from the top of the target is 228 mm)

The fission of $^{nat}Pb$ is a threshold reaction - neutron induced fission cross-section becomes significant at neutron energies of greater than ~30 MeV. The fission rate describes ultra-fast part of neutron spectra in the experimental setup.

E X P E R I M E N T T E C H N I Q U E S

# Techniques of fission rates measurement

## Measured value: reaction rates

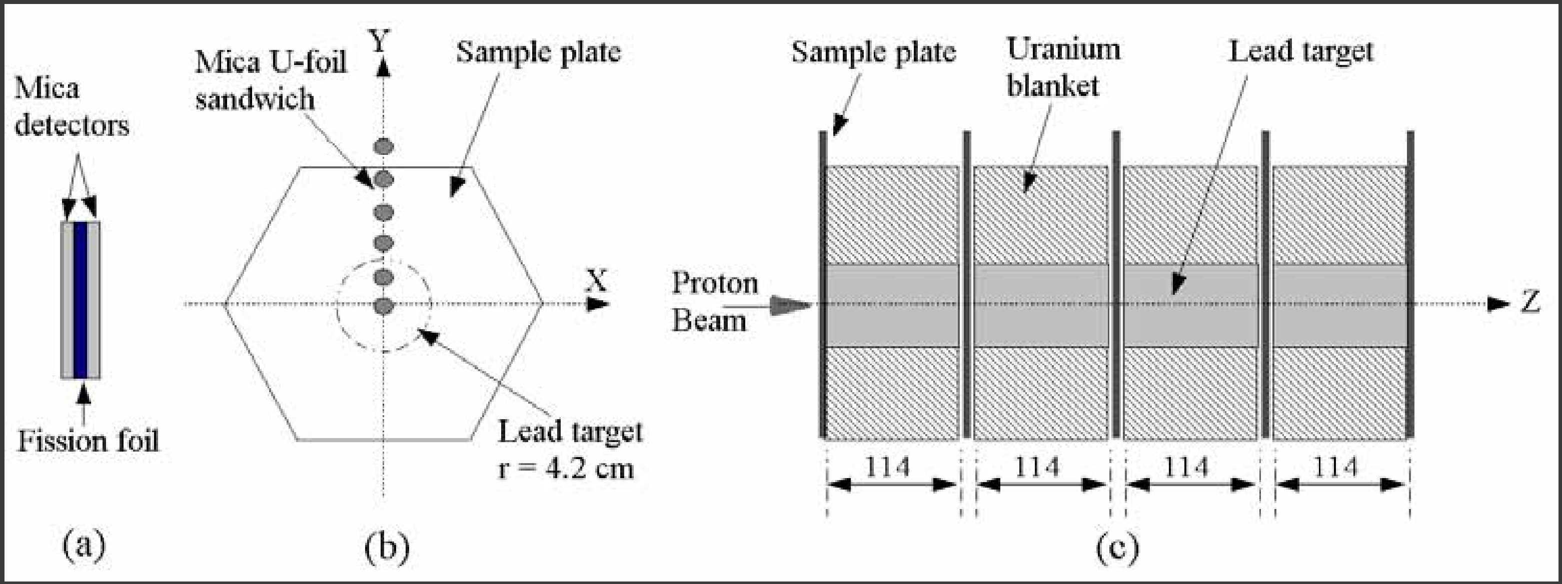
- Natural Uranium fission
- capture [$^{238}U(n,\gamma)$]
- Natural Lead fission

## Experimental techniques

- **Solid nuclear track detectors (SSNTD)**
  *(total fission reaction rates measurement)*
- **Standard activation method (γ-spectrometry)**
  *(neutron induced fission reaction rates measurement; capture reaction rates measurement)*

EXPERIMENT

# Sensors location




(a) The schematic drawing of the fission-foil-track-detector assembly used in the experiments.

(b) Schematic drawings of the sample plates and $^{nat}U$-mica detector sandwiches used in the experiment and

(c) placement of the sample plate within EPT assembly. Each target section is 114 mm long and there is a gap of 8 mm between each pair of target-blanket sections.

E X P E R I M E N T

# Experiment: $^{238}$U fission rate

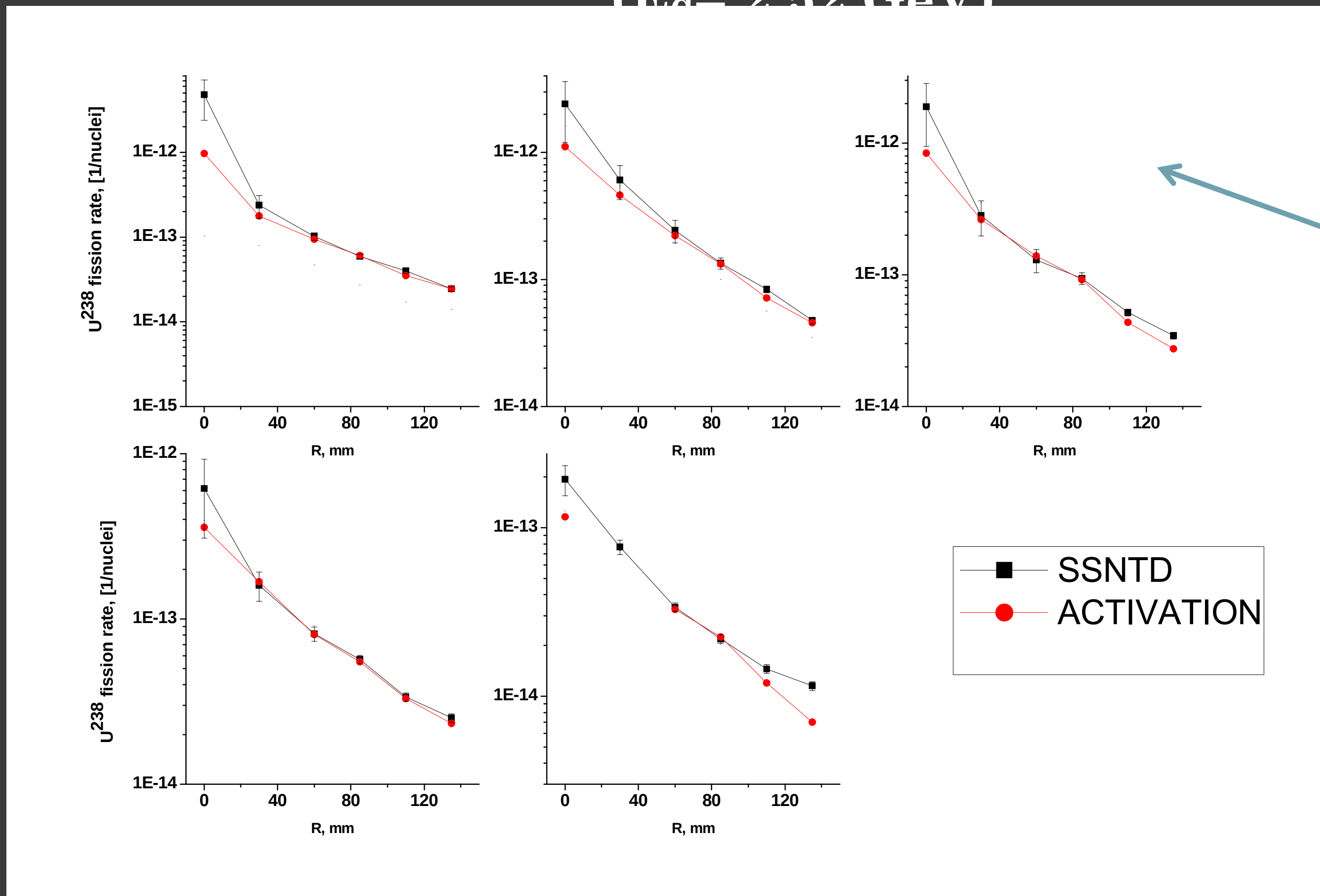
1. The fission rate - $R_f^{U238} = \int_{1.5}^{\infty} \sigma_f^{U238}(E)\varphi(E)dE$ describes fast part of neutron spectra (with the energy higher fission threshold of 238-U 1.5 MeV) in the experimental setup.

2. The fission rate was measured using two methods: Activation and SSNTD technique

3. All data are in good agreement (except region near the axis of the setup )

EXPERIMENT

# Experiment: two independent experimental methods for uranium fission rates determination (Ed= 2.52 GeV)



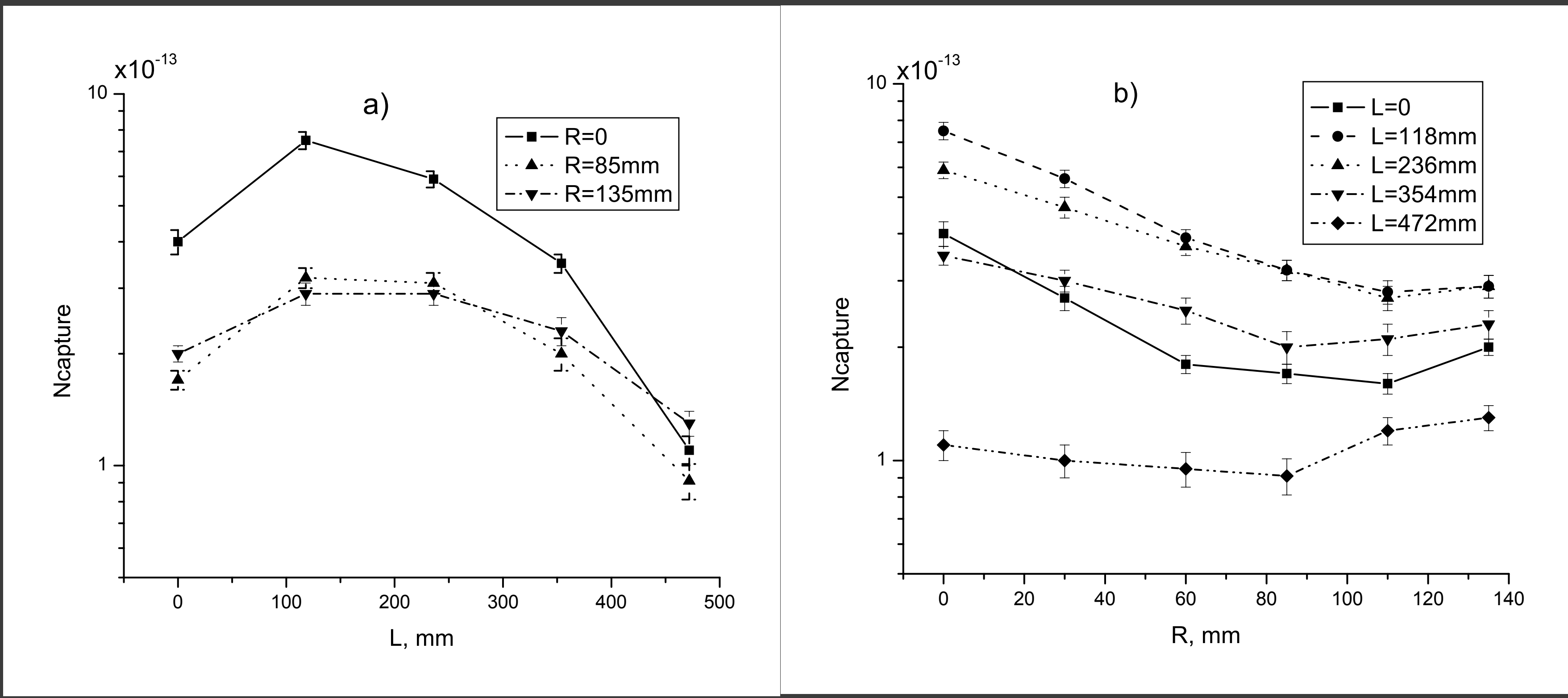

Note that the vertical scale ranges are not the same for all plots. Maximum is Situated on the third plate

Radial distributions of $^{238}$U fission rates inside the Pb-target and U-blanket for the five detector plates. R is the radial distance from the axis of the lead target. Lines are drawn to guide the eyes.

EXPERIMENT

# Spatial distributions of the number of radiative-capture reactions $^{238}U(n,\gamma)$ for the U/Pb assembly.

The data are given per $^{238}U$ nucleus, the deuteron fluence is $2.1\times10^{13}$, (**$E_d$=1.6 GeV**)




a)-axial (at R =0, 85,135 mm) and b)-radial (at plate 1, 2, 3, 4, 5 ).

E X P E R I M E N T

# Experiment: $^{239}$Pu accumulation

The procedure of combining the track counting and gamma-spectrometry techniques for the determination of spectral indices is a new development. It involves reception of information from the same sample by SSNTD-methods, i.e. counting the fission tracks of $^{238}$U, and by γ-ray spectrometry methods, i.e. counting a γ-line from the nuclide $^{239}$Np at 277.6 keV.

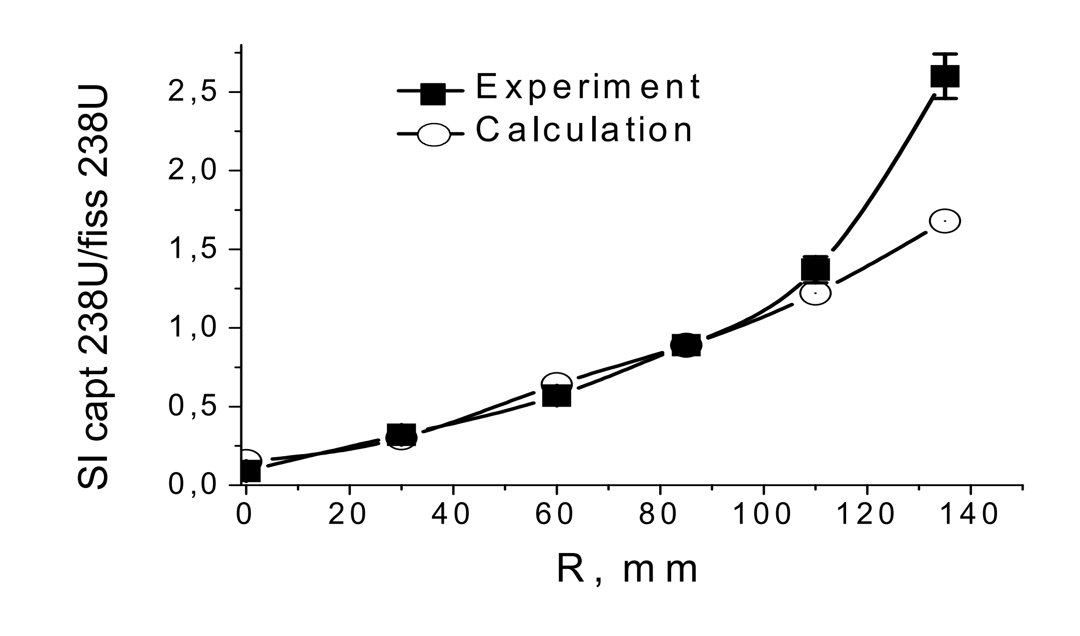
$$^{238}U(n,\gamma)^{239}U^{*} \xrightarrow{\beta^{-};T_{1/2}=23,4\min} {}^{239}Np^{*} \xrightarrow{\beta^{-};T_{1/2}=2,36d} {}^{239}Pu^{*} \xrightarrow{\alpha;T_{1/2}=2,41.10^{4}\,y}$$

| Deuteron Energy, GeV | Number of deuterons | 239-Pu accumulation in blanket, g | 239-Pu accumulation in blanket, g/deuteron·GeV |
|---|---|---|---|
| 1.60 | $2.1\times10^{13}$ | $(4.2\pm0.4)\cdot10^{-8}$ | $(1.2\pm0.1)\cdot10^{-21}$ |
| 2.52 | $5.9\times10^{12}$ | $(1.6\pm0.2)\cdot10^{-8}$ | $(1.1\pm0.1)\cdot10^{-21}$ |

E X P E R I M E N T

# Experiment: capture and fission

**The spectral index (ratio of reaction rates) characterizes a ratio between average cross-sections of neutron capture and neutron induced fission in the uranium blanket** $\bar{\sigma}_{capt}^{238U} / \bar{\sigma}_{f}^{238U}$




Neutron capture begin to prevail with increasing radial distance, probably because neutrons are moderated by inelastic collisions with nuclei of the blanket material.
A difference (1.5 times) is observed at the periphery of the assembly. This shows the underestimation in calculations of the influence of neutrons moderated in and reflected by the biological shielding.

**Radial distribution of a spectral index for the second plata (Z=118 mm)**

**spectral indexes are the best for comparison with Monte Carlo simulation: uncertainty in the number of primary particles are rejected**

LEAD BLOCK

# “Background” of the Project “Energy plus Transmutation” in JINR

- 1963-69, Investigation of neutron multiplicity in massive targets from metallic Uranium under proton irradiations (energy range 0.3-0.66 GeV) Vasillkov et al.

- 1965-68, Investigation of neutron multiplicity and neutron yields in massive Lead targets under proton irradiations (energy range 0.3-0.66 GeV) Vasillkov et al.

- 1979-84, Investigation of neutron multiplicity and neutron yields in massive Lead targets under proton irradiations (energy range 0.8-8.1 GeV) Vasillkov et al.

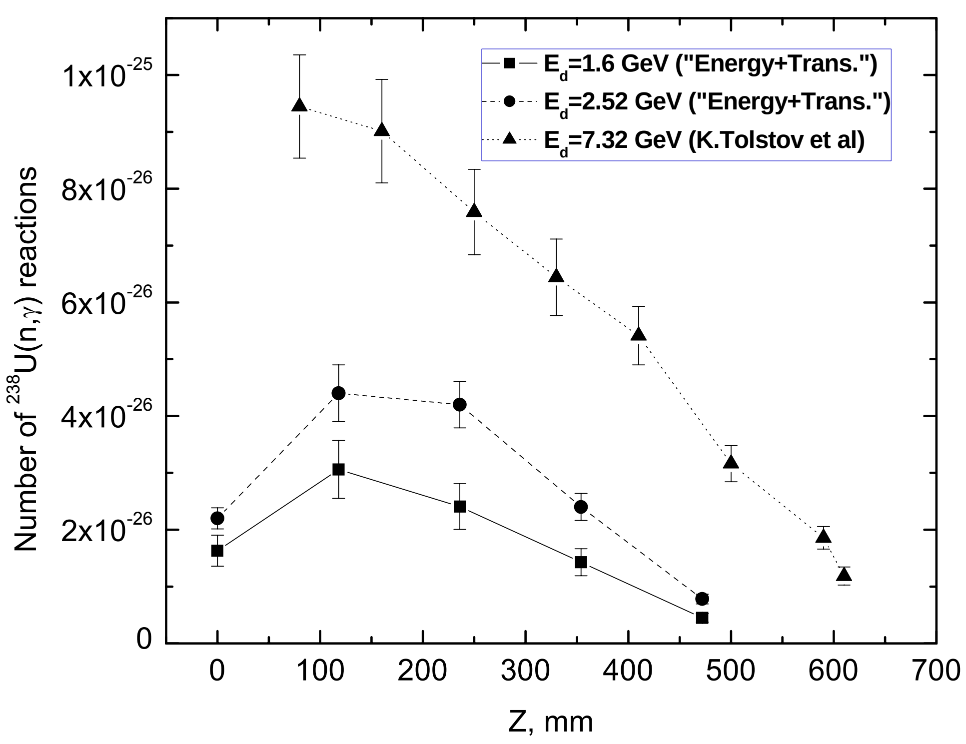
- 1987-92, Investigation of neutron generation and transport in massive Lead targets 50×50×80 cm$^3$ under charged particle (protons, alpha-particles, deuterons, $^{12}$C ions) irradiations (energy range 3.6-8.1 GeV) – project “Energy” - Tolstov et al. (see K.D.Tolstov, V.Ya.Migalenya, V.A.Voronko et al. Journal ”Atomic Energy”. (Publisher: Springer, New York) 1989, V. 67 , pp. 784-785.)

⇓

Project “Energy plus Transmutation” within the framework of research program ***“Investigations of physical aspect of electronuclear energy generation and atomic reactors radioactive waste transmutation using high energy beams of synchrophasotron/nuclotron JINR (Dubna)”***

LEAD BLOCK

# Comparison between “Lead block” and “Energy plus transmutation” experimental setups. Axial distribution of capture reaction rates.

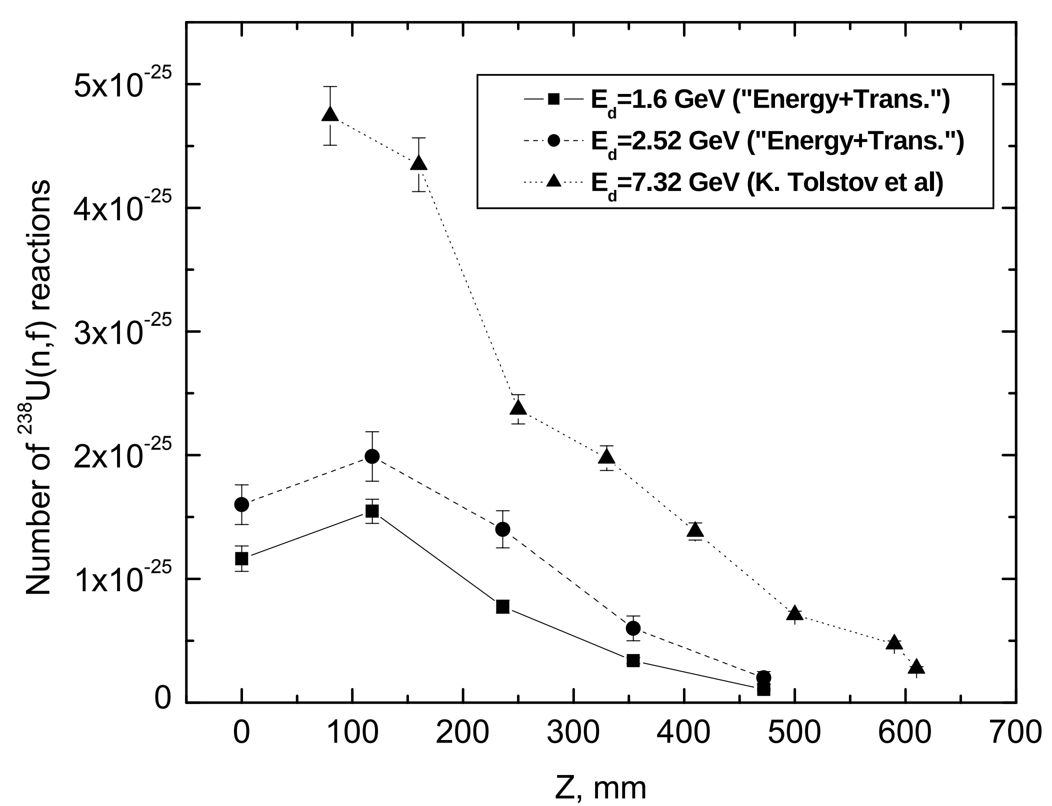



Axial distribution of radiation capture reaction rates $^{238}U(n,\gamma)$ for U/Pb assembly of the “Energy plus Transmutation” setup ($E_d$=1.6 и 2.52 GeV) and for lead block 50×50×80 cm ($E_d$=7.32 GeV). Normalized data (on nuclei of $^{238}U$ and deuteron) are given

Point for R=0 cm (axes of the setup)

L E A D B L O C K

# Comparison between "Lead block" and "Energy plus transmutation" experimental setups. Axial distribution of fission reaction rates.

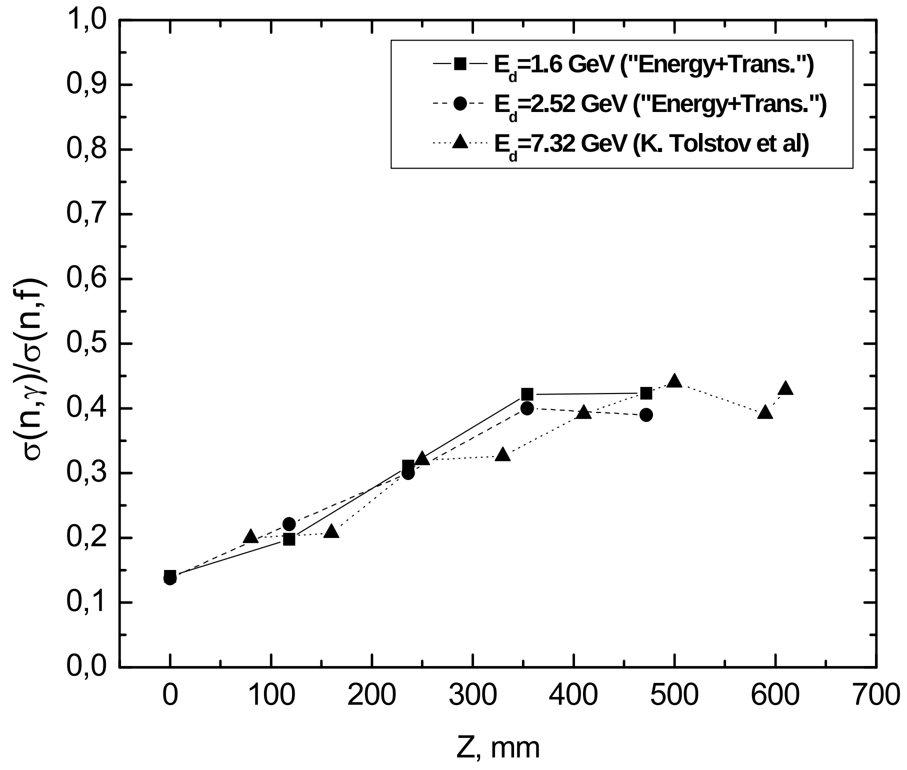



Axial distribution of $^{238}U$ fission reaction rates for U/Pb assembly of the "Energy plus Transmutation" setup ($E_d$=1.6 и 2.52 GeV) and for lead block 50×50×80 cm ($E_d$=7.32 GeV). Normalized data (on nuclei of $^{238}U$ and deuteron) are given

Point for R=0 cm (axes of the setup)

L E A D B L O C K

# Comparison between "Lead block" and "Energy plus transmutation" experimental setups. Axial distribution of spectral index $\overline{\sigma}_{capt}^{238U} / \overline{\sigma}_{f}^{238U}$




Axial distribution of spectral index U/Pb assembly of the "Energy plus Transmutation" setup ($E_d$=1.6 и 2.52 GeV) and for lead block 50×50×80 cm ($E_d$=7.32 GeV).

Point for R=0 cm (axes of the setup)

# “Background” of the Project “Energy plus Transmutation” in JINR

L E A D B L O C K

With increasing of axial distance a primary beam particles and the neutron cascade (a curve n, f) decreases faster than capture curve (n, y). It well shows by increase of spectral index values from value of 0.13 in an input of deuteron beam (Z=0) to the target, to value 0.4 for Z=300.

Such behavior of a spectral index means redistribution of the contribution of fast and resonant neutrons at their moderation. Really, assuming scattering length in a lead as a constant and equals to 3 cm, we can obtain, that energy of neutrons decreases with axial distance almost in three times with length of lead - 30 cm. Value of a spectral index becomes equal 0.4 on axial distance of 30 cm.

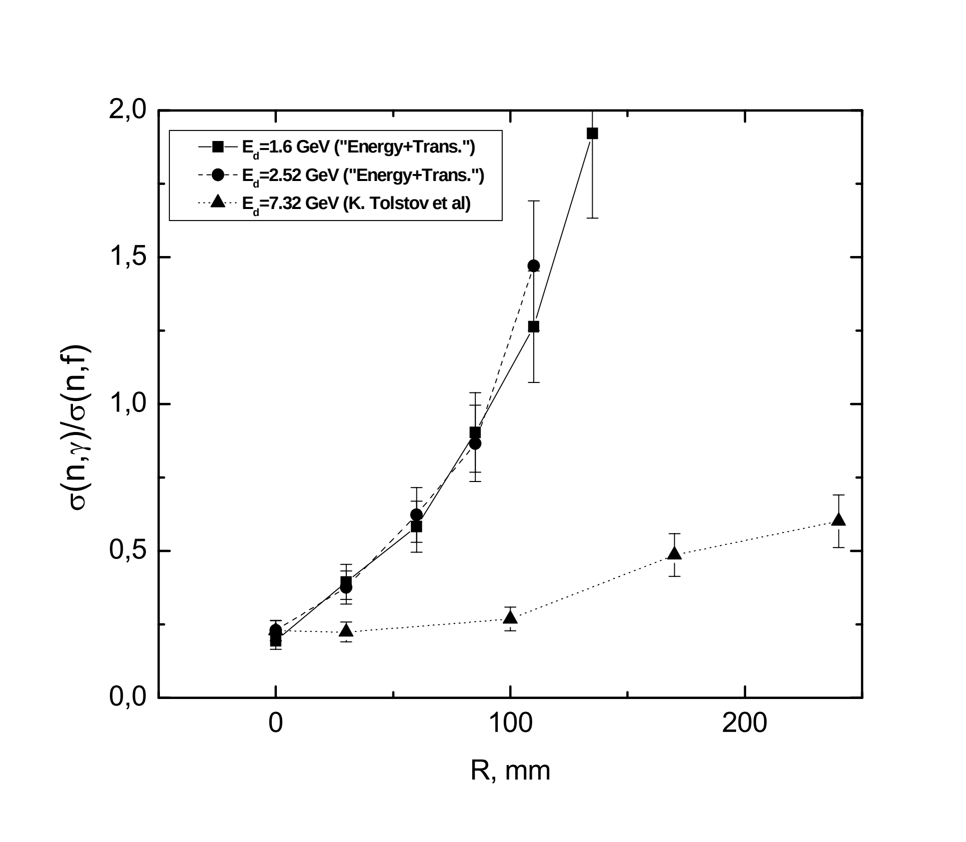
Further decrease of curves (n, f) and (n, y) occurs equally and value of spectral index from Z=300 to Z=600mm within an measurements errors remains to constants.

Thus energy of neutrons along the beam remains much more above than energy at which fission starts to affect Uranium 235.

Thus, distribution of a spectral index on a beam axis carries absolutely identical character as for assemblage uranium-lead so and for the big lead target.

L E A D B L O C K

# Comparison between "Lead block" and "Energy plus transmutation" experimental setups. Radial distribution of spectral index $\bar{\sigma}_{capt}^{238U} / \bar{\sigma}_{f}^{238U}$




Radial distribution of spectral index for U/Pb assembly of the "Energy plus Transmutation" setup $E_d$=1.6 и 2.52 GeV (Z=118 mm, the second experimental plate) and for lead block 50×50×80 cm ($E_d$=7.32 GeV, Z=150 mm).

Point Z=118 mm for "Energy plus transmutation" setup
Point Z=150 mm for "Lead block"

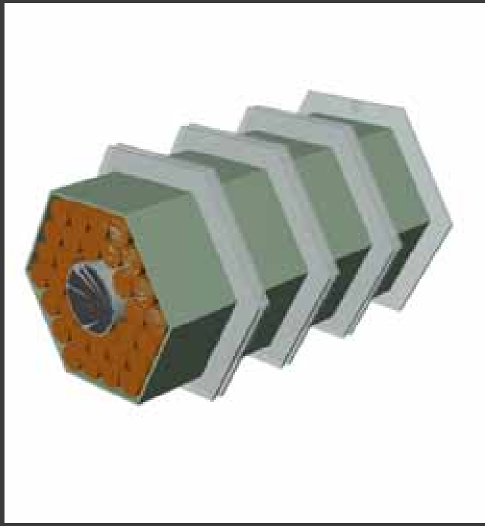
S
I
M
U
L
A
T
I
O
N
S

# Simulations: codes

Codes : **MCNPX** - version - 2.6C [calculation was made by S.R. Hashemi-Nezhad, *University of Sydney, Australia*]

**FLUKA** - version - 2006.3b [calculation was made by A.S. Potapenko, JIPNR, *Belarus*]

From the real view …to the geometrical module for Monte Carlo codes…

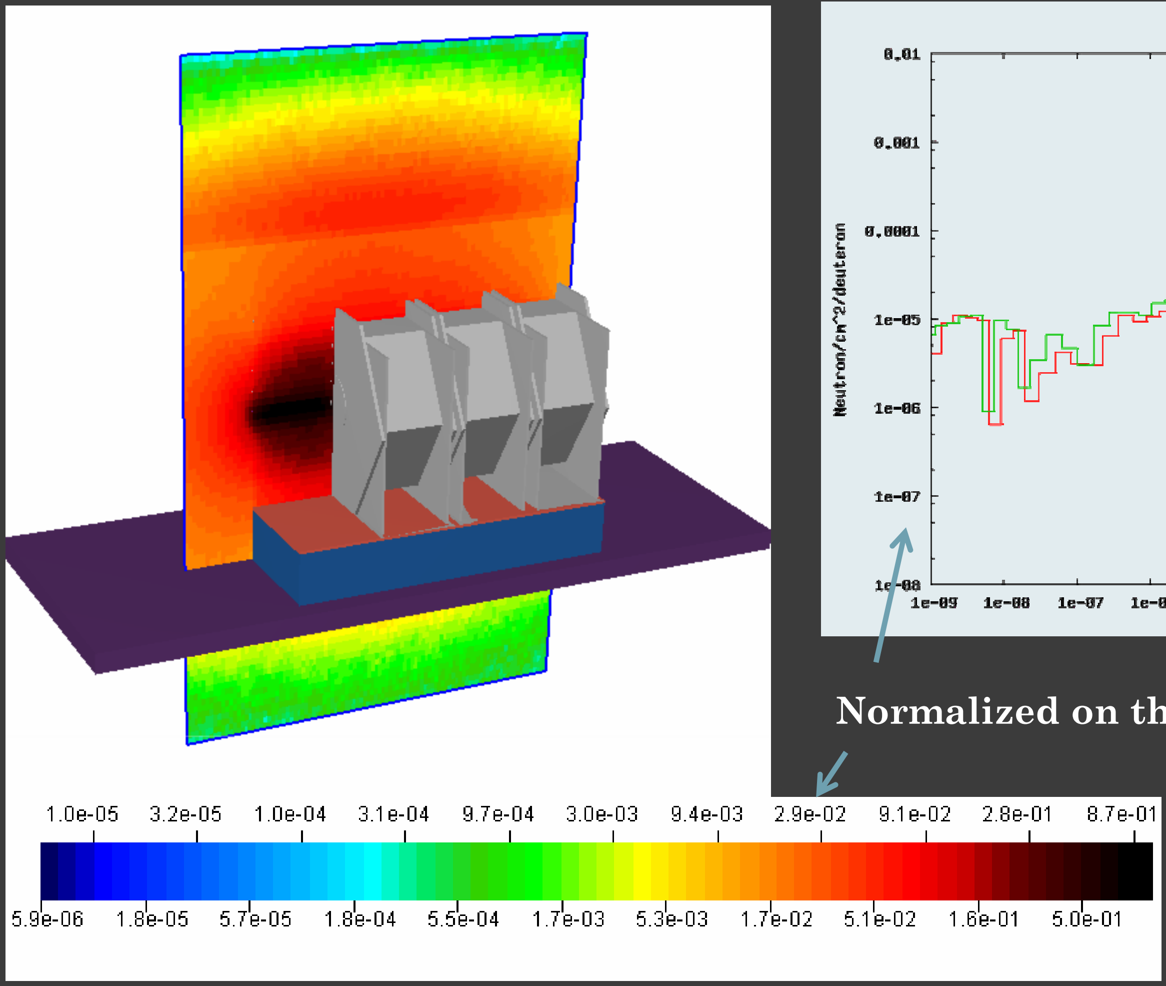


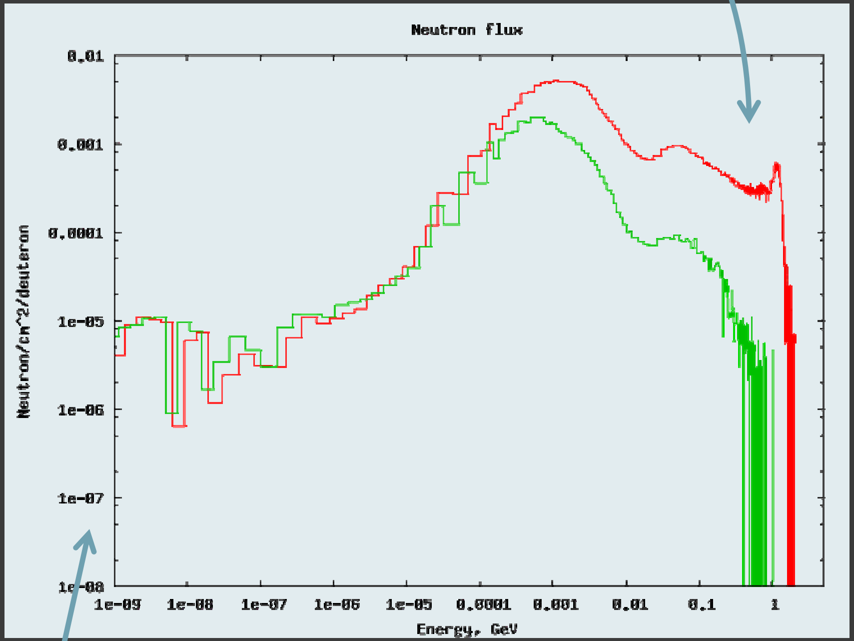
S I M U L A T I O N S

# Simulations: FLUKA results

Neutron spatial distribution: slice yz (the first section and shielding were removed for the best view)

Neutron spectrum for points:
Z=118 mm, R=0 mm (red line)
Z=118 mm, R=85 mm (green line)

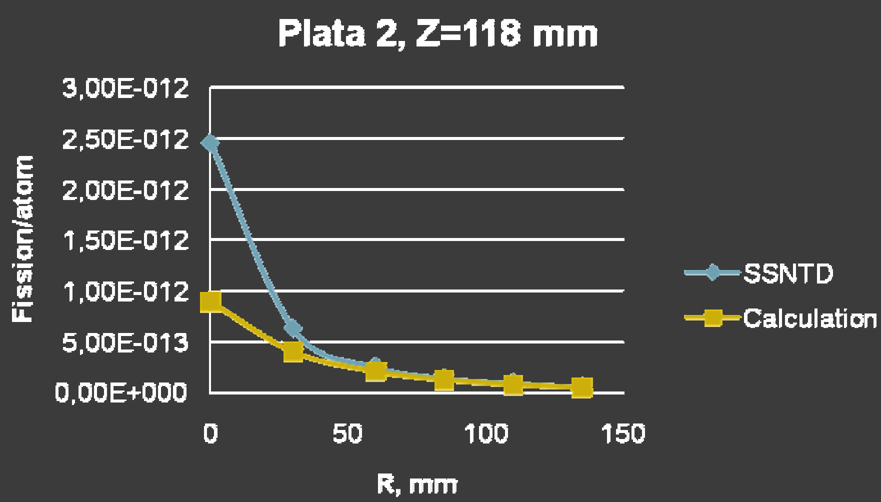


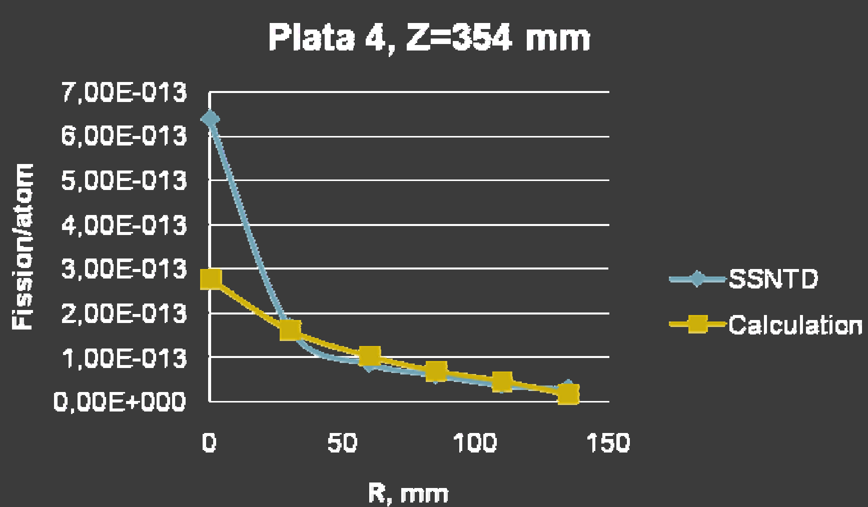




Normalized on the primary particles

SIMULATIONS

# Simulations: comparison FLUKA and SSNTD data




Natural Uranium fission rates: Experimental and calculated results

Radial distributions for Experimental plates No 2 and 4

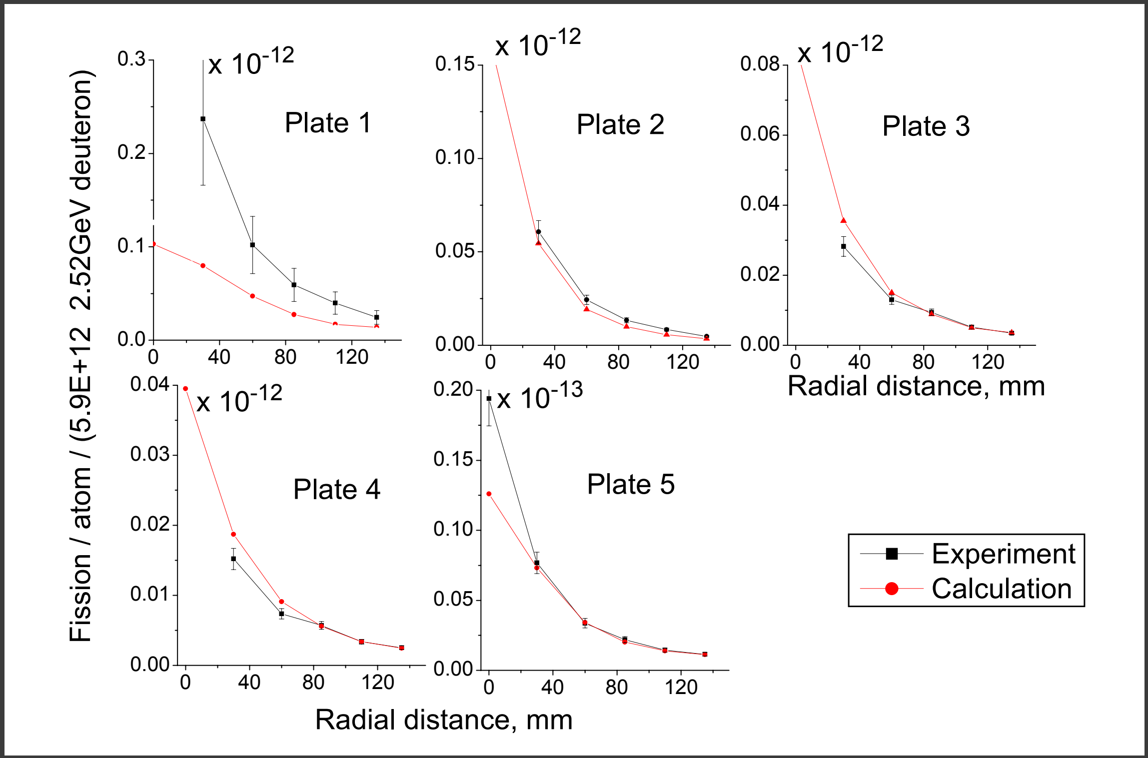
Experimental results are in good agreement with calculation in the “blanket region” (radial distances R>4.2 cm)
of the setup. It can be explained by overwhelming contribution of neutron induced fission reaction in the total uranium fission process

Note that the calculation Includes only neutron induced fission

S I M U L A T I O N S

# Simulations: comparison MCNPX and SSNTD data

## Natural Uranium fission rates: Experimental and calculated results




Experimental results are in good agreement (within the experimental error interval) with calculation in the “blanket region” (radial distances R>4.2 cm) of the setup.

Radial distributions of $^{238}U$ fission rates inside the Pb-target and U-blanket for the five detector plates. R is the radial distance from the axis of the lead target. Lines are drawn to guide the eyes.

# CONCLUSION

1. Research of neutron-physical characteristics of U/Pb-assembly with 4 section of uranium blanket, bombarded relativistic deutron with kinetic energies 1.6 and 2.52 GeV was carried out.
2. Comparison of experimental data with the calculation results obtained with using of computer codes MCNPX 2.5e and FLUKA were carried out. The good coincident of results of measurements and calculation of spectral indexes in blanket U/Pb-assembly testifies that calculation models, basically, correctly describe transport of particles in substance of blanket.
3. It have been measured the distribution of high-energy neutrons (with En> 30 MэB) in volume of a lead target and blanket in U/Pb-assembly.
4. Calibration factor for "lead+mica" sensor was obtained theoretically and experimentally.
5. It was developed combined track - γ -spectrometer measurement of a spectral index which give information from the same sample by SSNTD methods (fission tracks density of 238U) and by a γ-spectrometer method (on a γ-line nuclide 239Np with energy 277.6 keV) and which allows to measure spectral index with the error no more than 15 %. The developed technique will allow to determine accumulation 239Pu blanket of U/Pb-assembly.
6. New experimental values of fission Unat(d,f), Pbnat(d,f), 209-Bi(d,f), cross sections were obtained for deuteron energy 1.6 GeV.

# AKNOWLADGEMENTS

We would like to thank

Veksler and Baldin Laboratory of High Energies Physics (VBLHE), Joint Institute for Nuclear Research (JINR), Dubna, Russia and staff of the Nuclotron accelerator for providing us the research facilities used in these experiments.

JINR for the hospitality during their stay in Dubna.

National academy of science of Belarus for support of this work

# THANK YOU