

Andrzej Wojciechowski

andrzej.wojciechowski@ncbj.gov.pl

National Center for Nuclear Research, NCNR Otwock-Swierk, Polska

Research program: Energy + Transmutation of Radioactive Wastes
(E+T RAW) led by prof. S.Tiutiunnikov.

Kierownik UZ3 w NCBJ: prof. M.Dąbrowski.

'Measurement method of number fission
reactions of U-238 in proton or deuteron
beam by activation detectors'

October 2020

Outline

- 1. Introduction
 - Geometry and materials of the QUINTA assembly
 - main goals of the E+TRAW research program
- 2. Measurement method of number of fission reactions of U-238 inside relativistic protons or deuterons beam.
- 3. Experimental data and calculation results
 - comparison of data from SSNT, Activation detectors and results from MCNPX code.

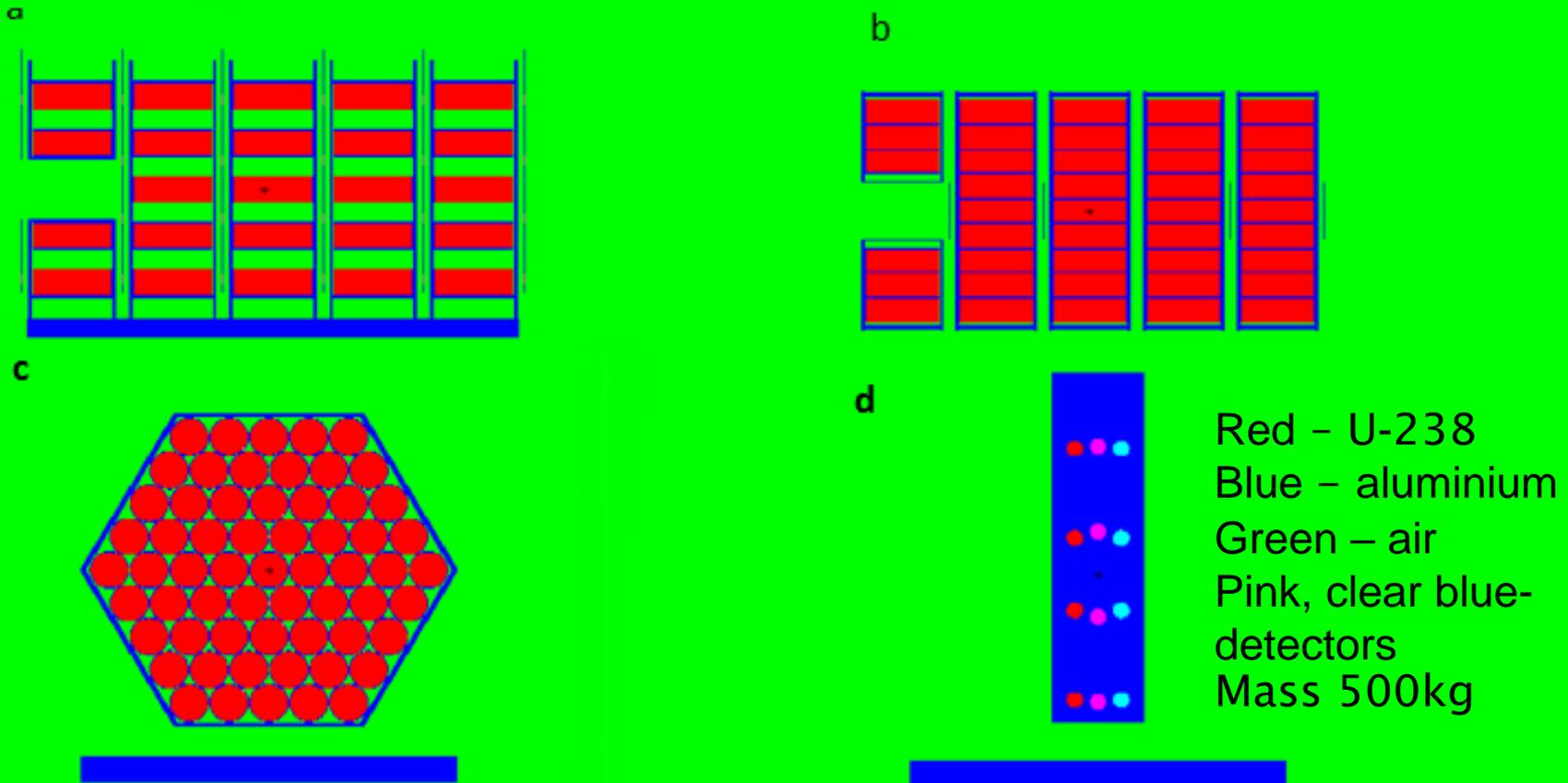
Outline

- [1] A. Wojciechowski, V. Voronko, et al., 2019, Simultaneous measurement of the neutron- and proton-induced fissions by activation detectors, *Measurement*, 146 (2019) 972–981

- [2] A. Wojciechowski, V. Voronko, et al., Measurement of fission reactions in deuteron beam by activation detectors, *Measurement*, In revision

Program E+T RAW

Geometry of the Kwinta detektor

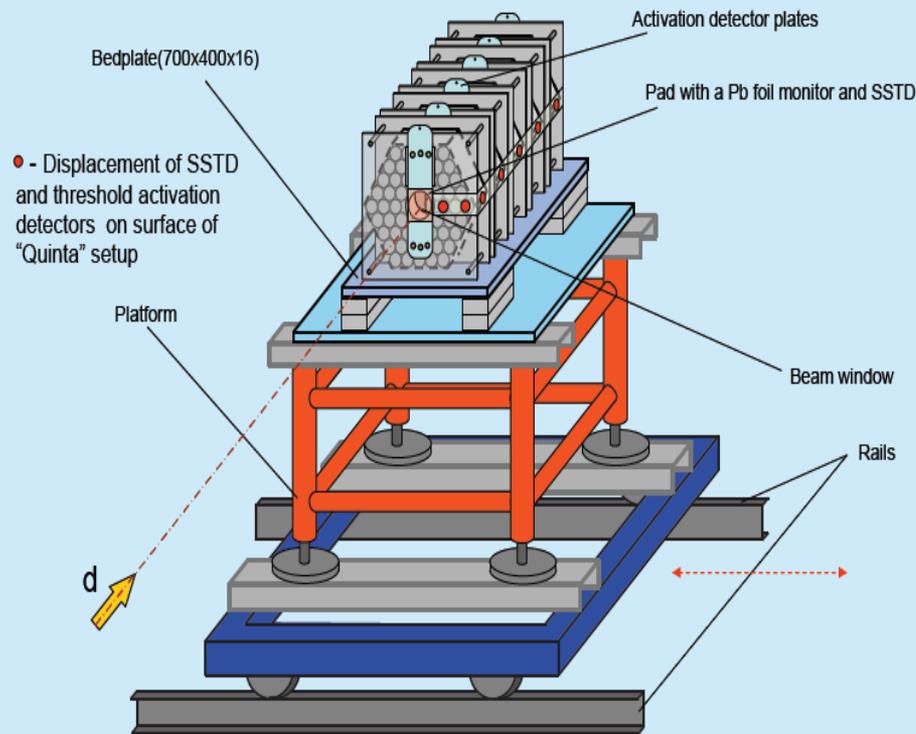


- a) The Z-Y cross section of assembly along the beam direction (at $X=0$)
b) The X-Y cross section of assembly at front wall of lead reflector.
c) The X-Y cross section of assembly at ^{nat}U cylinder blocks.
d) The X-Y cross section of assembly at probe plane with detectors

Kwinta – main aims

E+TRAW

Layout of upgraded target assembly “Quinta” at the irradiation position



ISINN-19, Dubna 25-28 May 2011

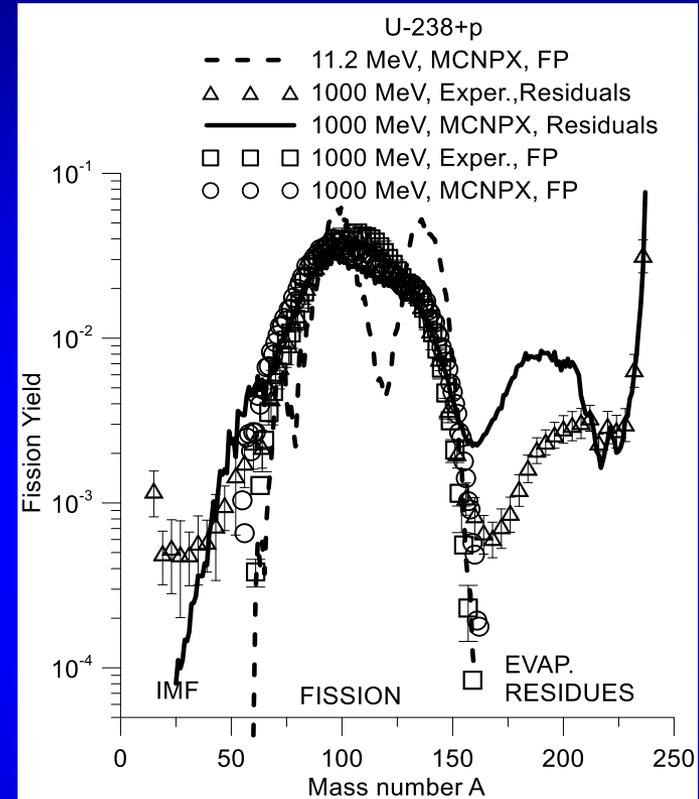
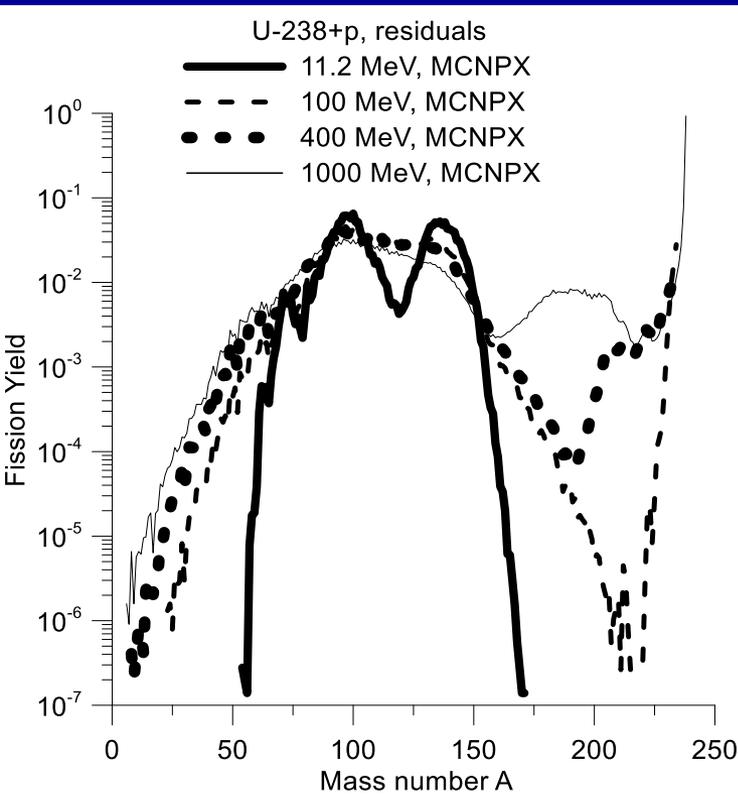
Research of

1. Deeply subcritical systems
2. Fission of U-238 i Th-232
3. Pu-239 i U-233 production
4. Transmutation of actynides i fission products
5. Measurement methods – mainly activation and track detectors.
6. Comparison experimental data and callculation results.

Number of fissions inside
proton beam

Second Estimation (SE) Method

Fission yields induced by protons



Calculation value of fission yield as a function of mass number of residuals for proton energy of 11.2, 100, 400 and 1000 MeV

Activation detector-Gamma spectroscopy

Experimental data [10-14] and calculation results of the fission yield. Residuals means evaporated IMF, the fission fragments and the heavy evaporation residues. FP means fission product for $28 < Z < 64$. Beam energy 1 AGeV.

Fissions in proton beam

Second estimation (SE) Method

$$n_{U(x,f)Z}^{\text{exp}} = \int_0^{\infty} (\phi^n(E) \sigma_{U(n,f)}(E) Y_{U(n,f)Z} + \phi^p(E) \sigma_{U(p,f)}(E) Y_{U(p,f)Z}) B dE$$

Number of fission product

$$\begin{bmatrix} n_{Zr97}^{\text{exp}} \\ n_{I131}^{\text{exp}} \\ n_{I133}^{\text{exp}} \\ n_{Ce143}^{\text{exp}} \end{bmatrix} = \begin{bmatrix} Y_{Zr97,U(n,f)} & Y_{Zr97,U(p,f)} \\ Y_{I131,U(n,f)} & Y_{I131,U(p,f)} \\ Y_{I133,U(n,f)} & Y_{I133,U(p,f)} \\ Y_{Ce143,U(n,f)} & Y_{Ce143,U(p,f)} \end{bmatrix} \begin{bmatrix} n_{U(n,f)} \\ n_{U(p,f)} \end{bmatrix}$$

Y-fission yield of fission product

Main problem:
determination of matrix Y

The minimal value of

$$\| \bar{n}^{\text{exp}} - \bar{Y} \bar{n} \|$$

satisfy the equation

$$\left. \begin{array}{l} n_{U(n,f)} \geq 0 \\ n_{U(p,f)} \geq 0 \end{array} \right\}$$

$$\frac{\partial \| \bar{n}^{\text{exp}} - \bar{Y} \bar{n} \|}{\partial \bar{n}} = 0$$

condition number is big

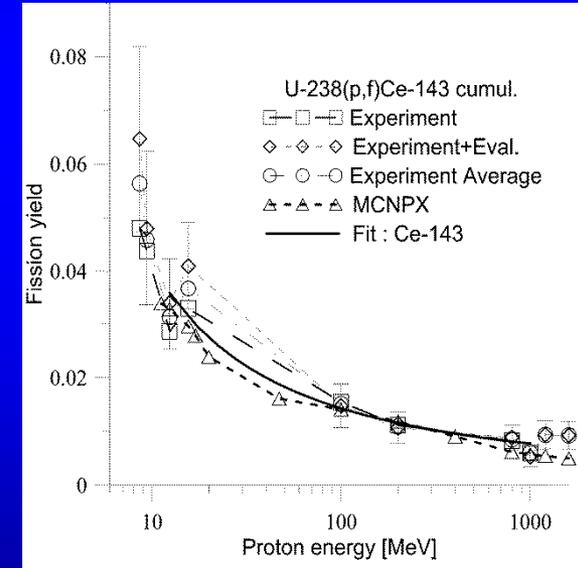
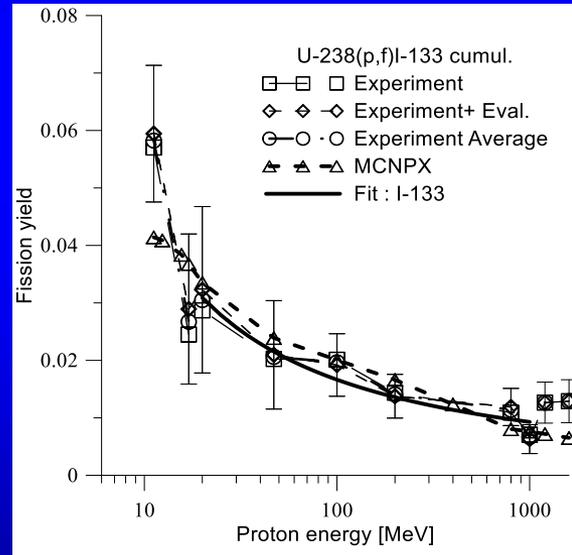
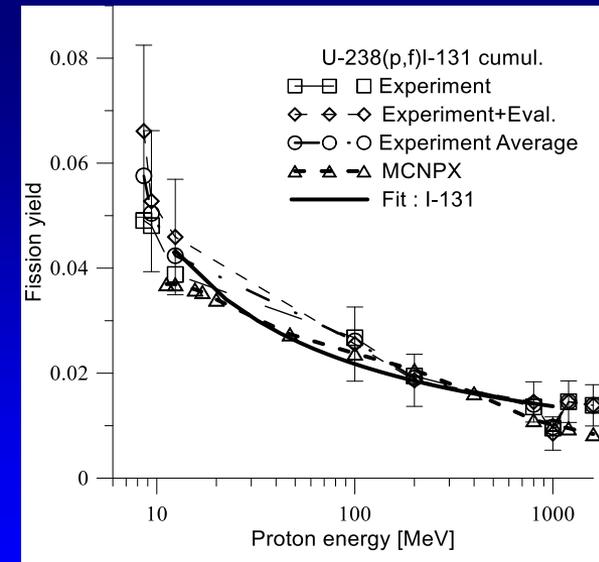
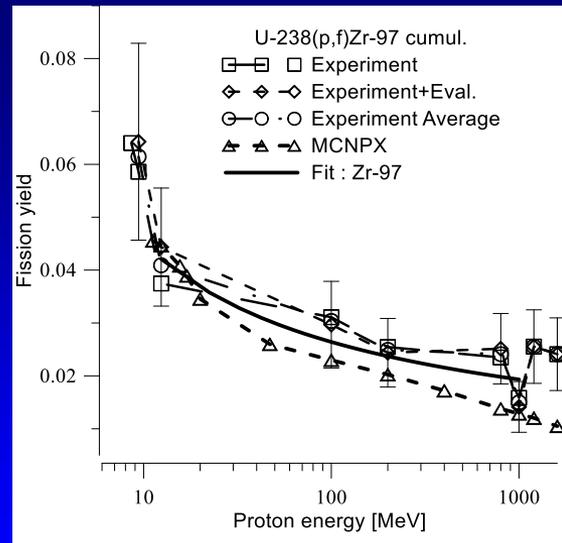
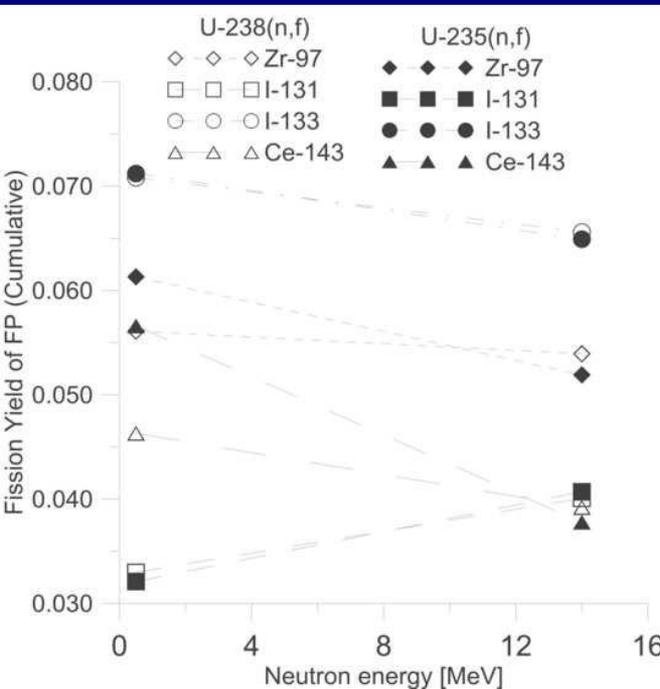
$$\bar{n} = (\bar{Y}^T \bar{Y})^{-1} \bar{Y}^T \bar{n}^{\text{exp}}$$

$$n_{U(n,f)}^{FE} = \frac{\sum_{z=1}^4 Y_z n_z^{\text{exp}}}{\sum_{z=1}^4 Y_z^2}$$

For low value of proton flux

$$n_{U(p,f)} = 0$$

Fission yields

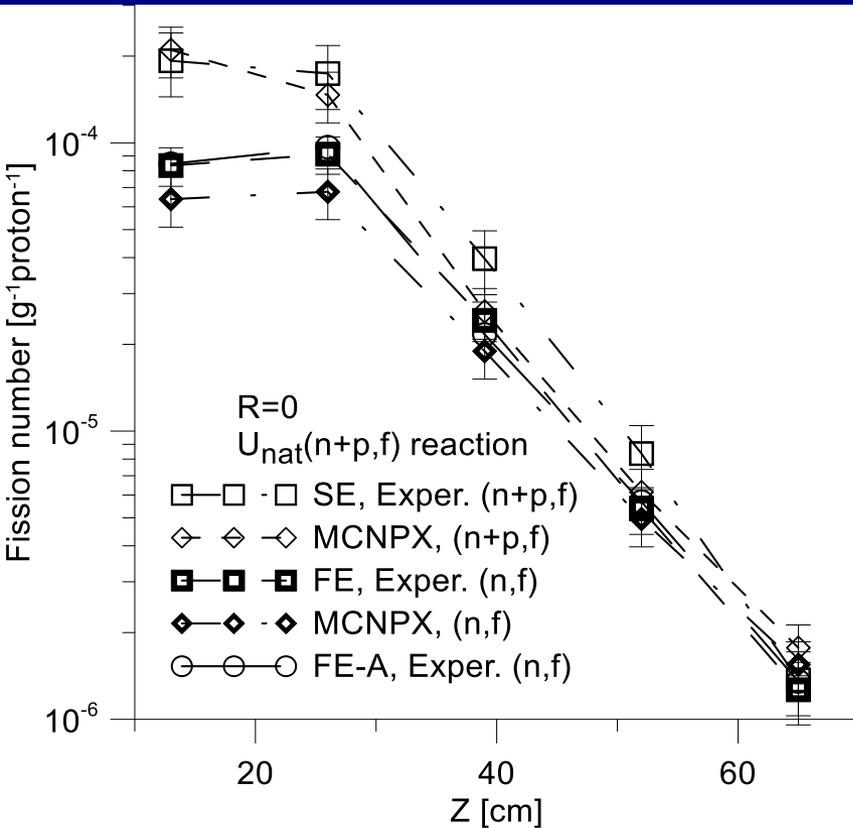


Fission yields of fission reaction induced by neutrons and protons

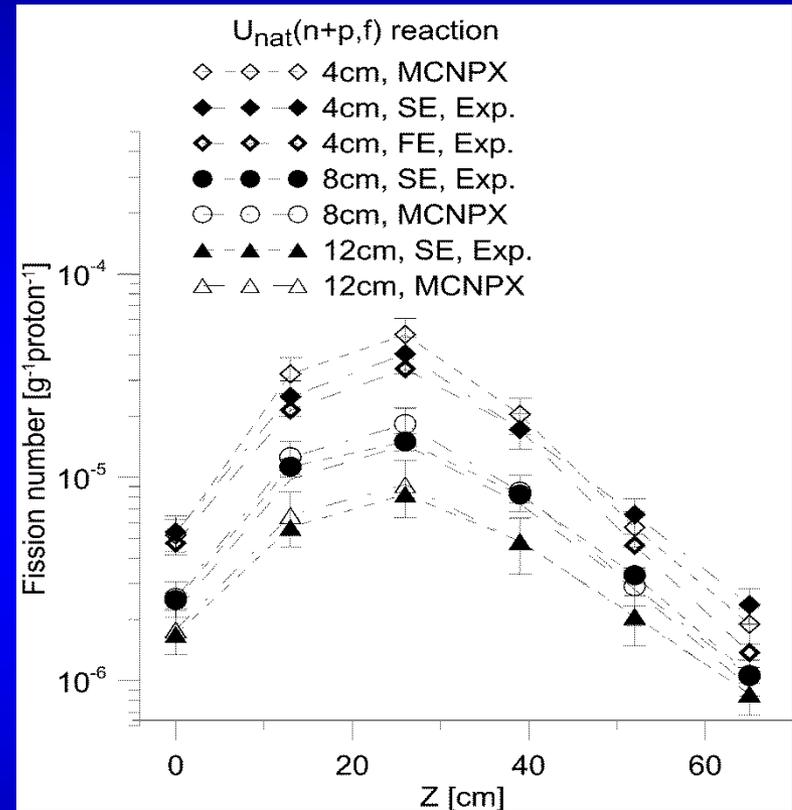
Fission yields function was obtain as fitting to experimental data

A.Wojciechowski, et al. , Measurement, 146 (2019) 972–981

Fissions - U-238(n+p,f), proton beam, 660MeV



-Second -Estimation (SE) method includes fissions induced by relativistic protons



The longitudinal distribution of fission number for detectors placed on the axis of assembly

Experiment : November 2014,
 The Ukrainian Group led by prof.Voronko

The same as on the left figure but for R=4, 8 and 12cm

Number of fissions inside
deuteron beam

Third Estimation (TE) Method

Why are we interested proton-induced fissions in deuteron beam?

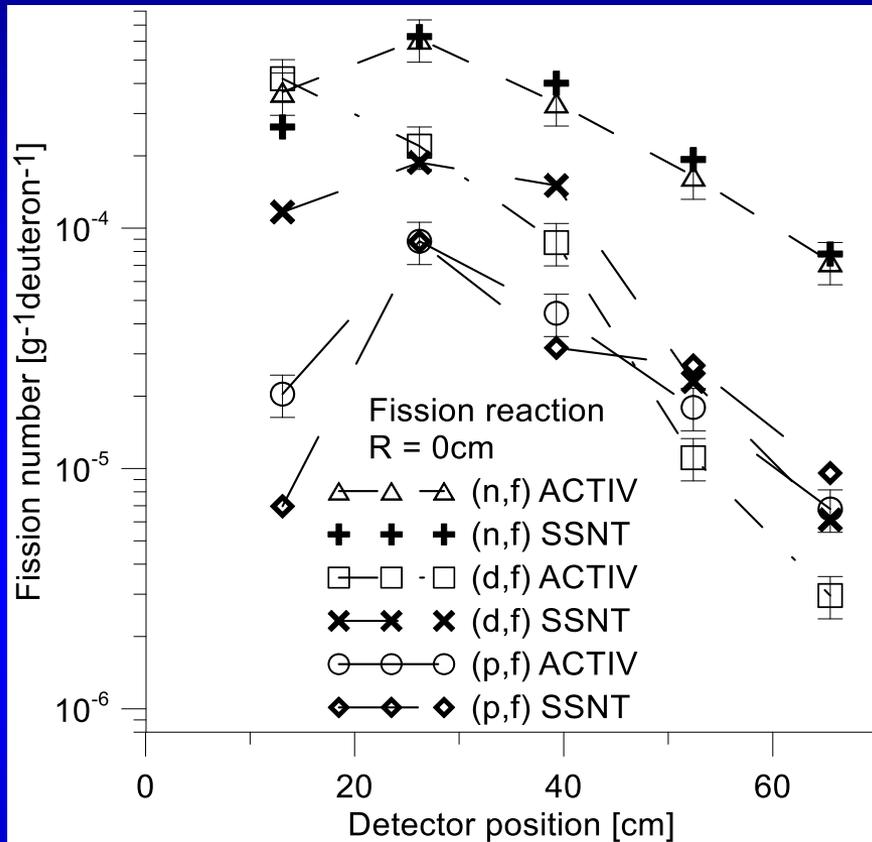


Fig.7 Neutron-, deuteron- and proton-induced fission number along the assembly axis for R=0. MCNPX simulation for experimental beam parameters based on SSNTD and Activation detector.

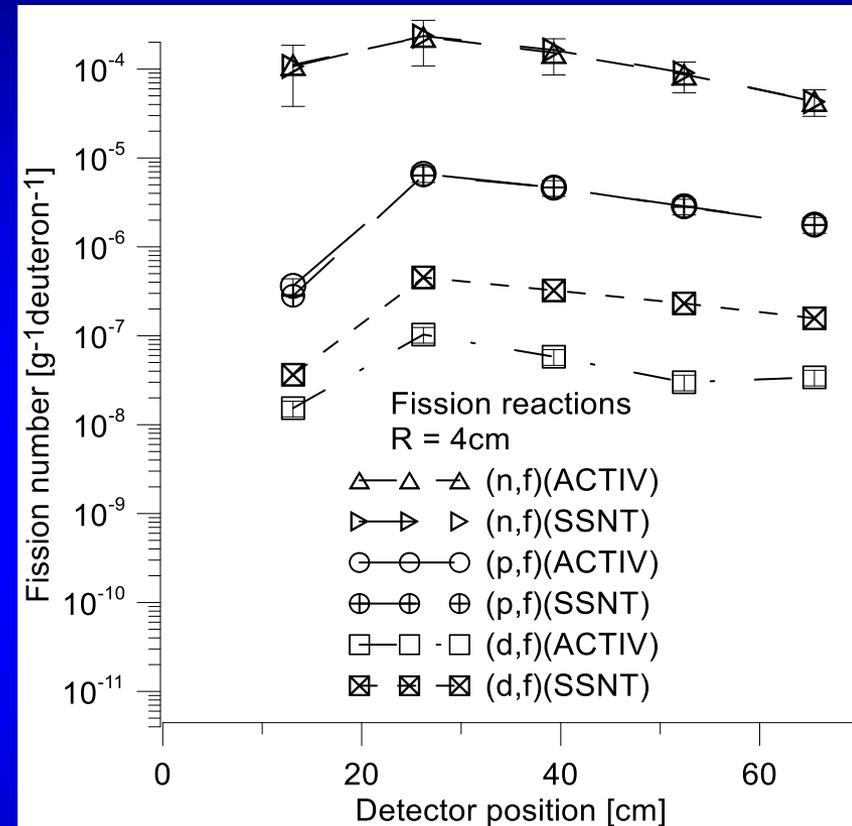


Fig 8. The same as in the Fig7. but for R=4cm

Fissions in deuteron beam

TE method

$$n_{U(x,f)Z}^{\text{exp}} = \int_0^{\infty} (\phi^n(E)\sigma_{U(n,f)Z}(E)Y_{U(n,f)Z} + \phi^d(E)\sigma_{U(d,f)Z}(E)Y_{U(d,f)Z} + \phi^p(E)\sigma_{U(p,f)Z}(E)Y_{U(p,f)Z})BdE$$

$n_{U(x,f)Z}^{\text{exp}}$ - the experimental number of fission products Z released in the reaction $U(x, f)Z$ per volume unit [cm^{-3}]

$\phi^n(E), \phi^d(E), \phi^p(E)$ - the energy spectrum of the neutron, deuteron or proton flux density [$\text{cm}^{-2}\text{MeV}^{-1}$] respectively. $\phi^d(E)$ is a sum of initial and secondary deuteron flux density.

$\sigma_{U(n,f)Z}, \sigma_{U(d,f)Z}, \sigma_{U(p,f)Z}$ - microscopic cross-section of $U(n, f)Z, U(d, f)Z$ and $U(p, f)Z$ fission reaction induced by neutron, deuteron or proton correspondently.

ρ - detector mass density [g/cm^3]

A - Avogadro constant

m - mass of 1 mole [g]

$$B = \rho A / m$$

Fissions in deuteron beam Third Estimation (TE) Method

$$\begin{bmatrix} n_{Zr97}^{\text{exp}} \\ n_{I131}^{\text{exp}} \\ n_{I133}^{\text{exp}} \\ n_{Ce143}^{\text{exp}} \end{bmatrix} = \begin{bmatrix} Y_{Zr97,U(n,f)} & Y_{Zr97,U(d,f)} & Y_{Zr97,U(p,f)} \\ Y_{I131,U(n,f)} & Y_{I131,U(d,f)} & Y_{I131,U(p,f)} \\ Y_{I133,U(n,f)} & Y_{I133,U(d,f)} & Y_{I133,U(p,f)} \\ Y_{Ce143,U(n,f)} & Y_{Ce143,U(d,f)} & Y_{Ce143,U(p,f)} \end{bmatrix} \begin{bmatrix} n_{U(n,f)} \\ n_{U(d,f)} \\ n_{U(p,f)} \end{bmatrix}$$

and

$$\left. \begin{array}{l} n_{U(n,f)} \geq 0 \\ n_{U(d,f)} \geq 0 \\ n_{U(p,f)} \geq 0 \end{array} \right\}$$

The minimal value of
the equation

$$\left\| \mathbf{n}^{\text{exp}} - \mathbf{Y} \mathbf{n} \right\| \text{ satisfy}$$

Fissions in deuteron beam

To solve this problem we can define a value of the fission vector

$$\bar{n}$$

for which the following norm achieves a minimal value

$$\|\bar{n}^{\text{exp}} - \bar{Y}\bar{n}\| = \text{minimum}$$

The vector \bar{n} satisfies the above equation one can presents in the standard form

$$\bar{n} = (\bar{Y}^T \bar{Y})^{-1} \bar{Y}^T \bar{n}^{\text{exp}}$$

One can solve the problem it is equation system of equations and conditions) simultaneously using the ALN1R program [24]. This program employ the Nonnegative Least Squares (NNLS) method [25]

[24] http://num-anal.srcc.msu.ru/lib_na/cat/cat59.htm -ALN1R

[25] Lawson C.L., Hanson R.J. "Solving Least Squares Problem", Prentice - Hall Inc., Englewood Cliffs, New Jersey, 1974

Fission yields induced by neutron

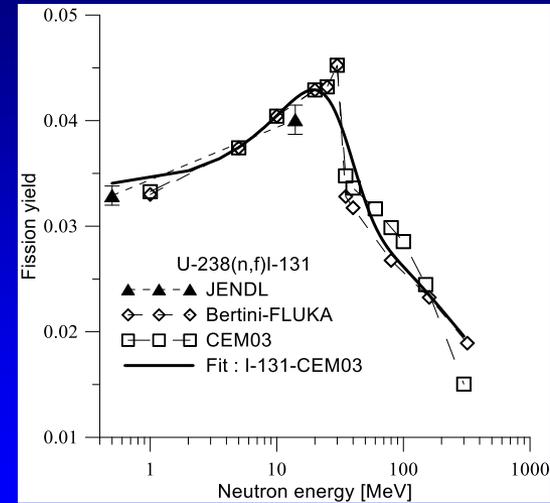
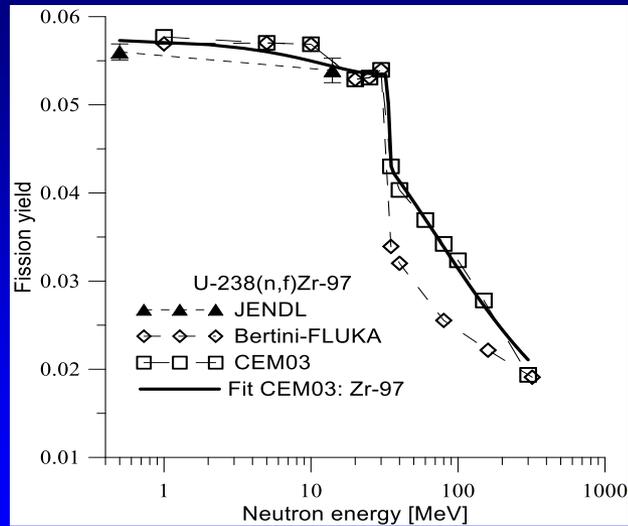


Fig.3. Cumulative neutron-induced FY for Zr-97. The experimental data are from JENDL library

Fig.4. The same as in the Fig,3 but for I-131

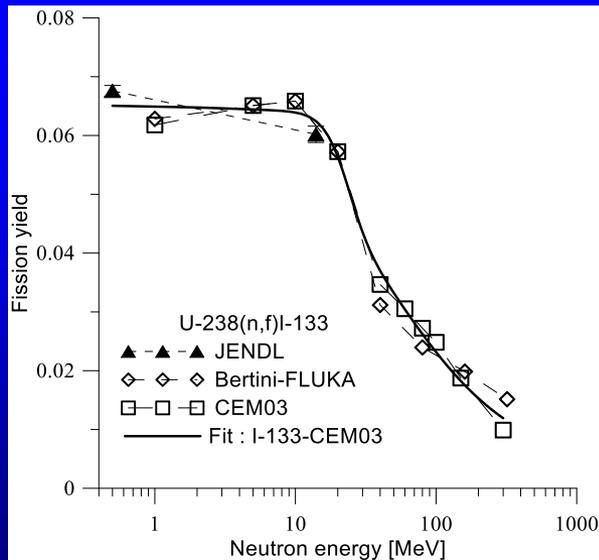


Fig.5. The same as in the Fig,3 but for I-133

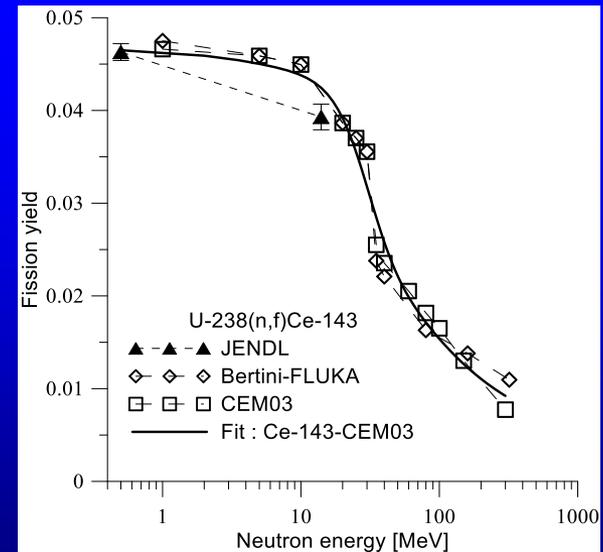


Fig.6. The same as in the Fig,3 but for Ce-143.

Fission yields induced by deuterons

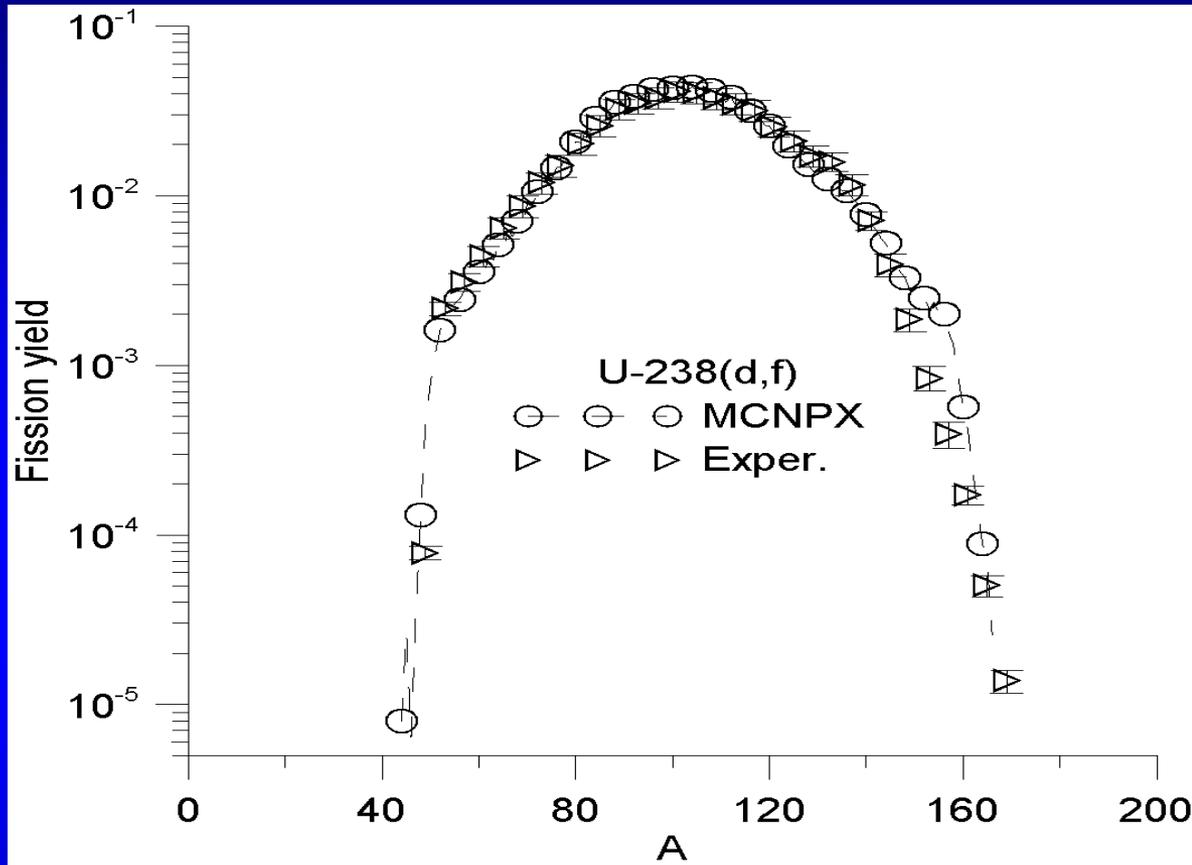


Fig.9. Fission yield for U-238(d,f) reaction as a function atomic mass A for Z=23-69 and A=49-169. Experimental data are from [16 and 17]. Calculations are obtained from Abl-FLUKA-LAQGSM model of MCNPX code. Beam energy 1 AGeV.

[16] J. Pereira, J. Benlliure, E. Casarejos, et al., 2007, Isotopic production cross sections and recoil velocities of spallation-fission fragments in the reaction $^{238}\text{U}(1\text{A GeV}) + d$, PHYS. REV. C 75, 014602.

[17] E. Casarejos, J. Benlliure, J. Pereira, et al., 2006, Isotopic production cross sections of spallation-evaporation residues from reactions of $^{238}\text{U}(1\text{A GeV})$ with deuterium, PHYS. REV. C 74, 044612 (2006)

Fission yields induced by deuterons

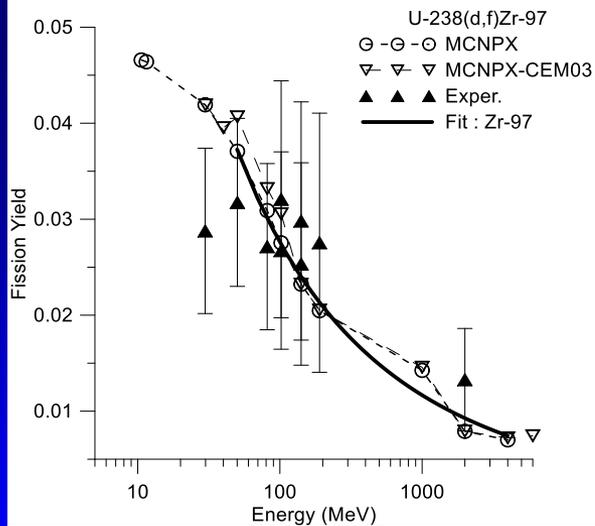


Fig.10. Cumulative FY of U-238(d,f) reaction for Zr-97. Experimental data are from [16, 27],], MCNPX means AbLa-FLUKA-LAQGSM model of MCNPX code.

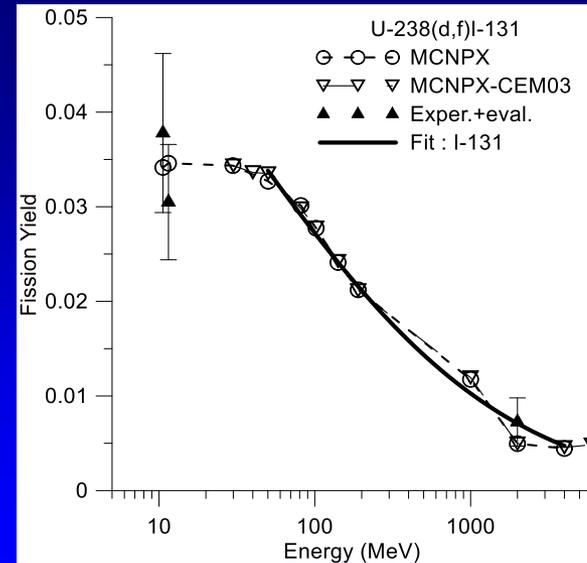


Fig.11. The same as in the Fig.10. but for I-131.

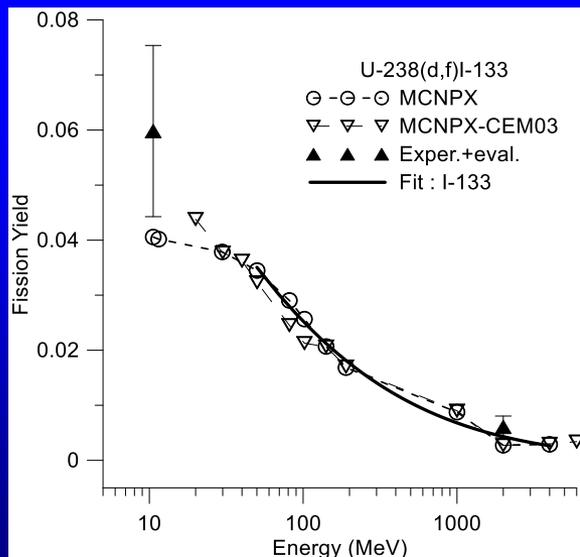


Fig.12. The same as in the Fig.10. but for I-133.

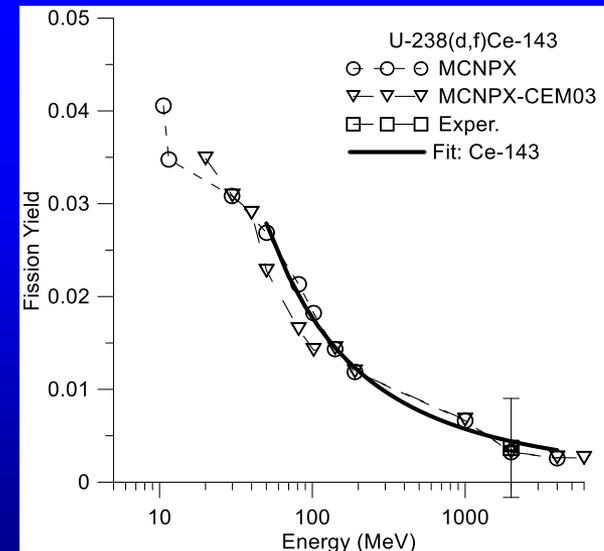


Fig.13. The same as in the Fig.10. but for Ce.143.

Experiment, Dubna,2011, deuteron beam 4GeV

Tab.1. Beam parameters for energy 4GeV

Measuring method	Beam center, [cm]		FWHM for Gaussian, [cm]		Total number of deuterons
	Xc	Yc	FWHM _x	FWHM _y	
SSNTD	2.0±0.2	-0.59±0.05	1.3±0.1	1.61±0.05	(1.4±0.17)×10 ¹³
Activate (ACTIV)	1.2	-0.7	2.2	2.3	(1.42±0.18)×10 ¹³

The Belarusian group led by prof. Zhuk - SSNT detector

The Ukrainian group led by prof. Voronko - Activation detector

Experimental data and calculation results Deuteron beam, 4GeV

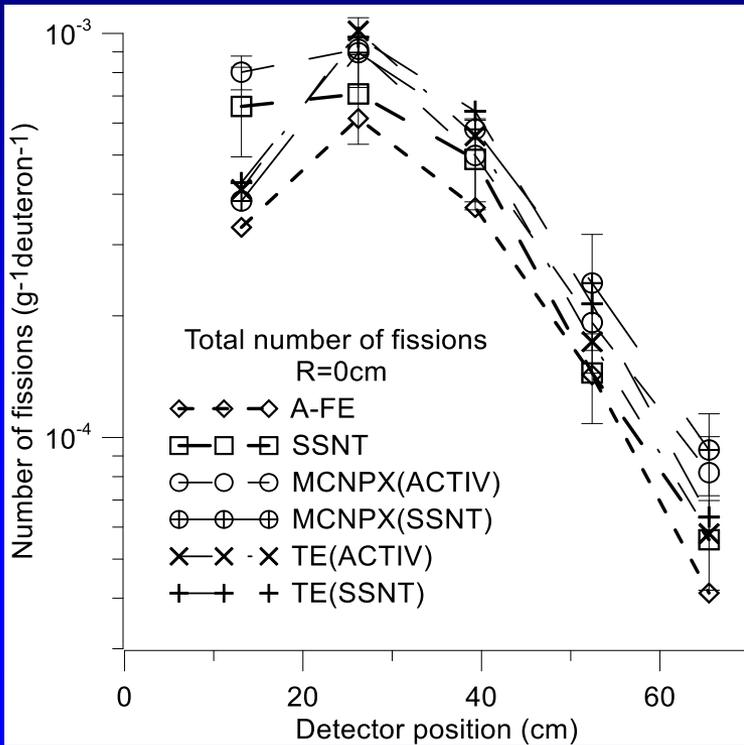


Fig.14. Total number of fissions placed on assembly axis obtained by SSNTD and activation detectors, predicted by MCNPX code and obtained by TE method

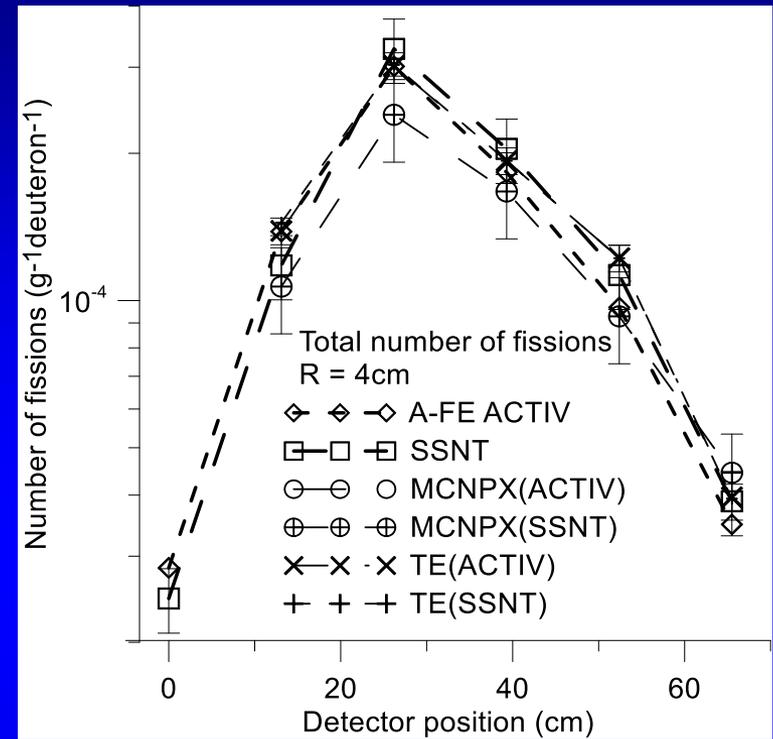


Fig.15 The same as in the Fig.14. but for R=4cm.

Experiment, Dubna, 2011

The Belarusian group led by prof. Zhuk - SSNT detector

The Ukrainian group led by prof. Voronko - Activation detector

Final verification

$$RD = \frac{\left| \overline{n^{\text{exp}}} - \overline{Yn} \right|}{\left| \overline{n^{\text{exp}}} \right|}$$

relative deviation

	A-FE	MCNPX (SSNT)	MCNPX (ACTIV)	TE (SSNT)	TE (ACTIV)
Average RD for axial detectors	0.091	0.38	0.43	0.11	0.083
Average RD for not axial detectors	0.046	0.19	0.21	0.031	0.032

Conclusion and remarks

- The measurement of total number of fission reactions inside volume of high energy deuterons or protons beam has key meaning for investigation ADS assembly, so is important from a practical viewpoint.
- The TE method is an effective method to estimate total number of fissions employing activation detectors and good statistic characteristic for these detectors.
 - Activation detectors have significantly greater statistic than SSNTDs.
 - This method determined total number of fissions induced by neutron, deuteron and proton, simultaneously.
- The neutron- and deuteron induced fission yields are determine employing experimental data and calculation results. However the experimental data are few. For these reasons neutron- and deuteron- induced fission yields require experimental confirmation in correspondent energy range.
- The average energy of different kind of particle are key magnitude to determine FY of relevant particles used by TE method. This magnitude we computed by MCNPX code. However one can determine in experimental method describe in Ref.[34].

[34] A.Wojciechowski, et al., A method of measuring the neutron energy spectrum by activation detector Measurement, 90 (2016) 118–126

The End

Thank you for your attention
Questions?

Reverse geometry

- [10] M. Bernas et al, FISSION-RESIDUES PRODUCED IN THE SPALLATION REACTION $^{238}\text{U} + p$ at 1 A GeV, Nucl.Phys. A725 (2003) 213-253, nucl-ex/0304003.
- [11] M. V. Ricciardi, P. Armbruster, J. Benlliure, et al, Light Nuclides Produced in the Proton-Induced Spallation of ^{238}U at 1 GeV, Physical Review C 73(1), (2005).
- [12] M. Bernas, P. Armbruster, J. Benlliure, et al., Very heavy fission fragments produced in the spallation reaction $^{238}\text{U} + p$ at 1A GeV, Nuclear Physics A 765 (2006) 197–210.
- [13] J. Taïeb, K.-H. Schmidt , L. Tassan-Got, et al., Evaporation residues produced in the spallation reaction $^{238}\text{U} + p$ at 1 AGeV, Nuclear Physics A 724 (2003) 413–430
- [16] J. Pereira,1,* J. Benlliure,1 E. Casarejos, et al., 2007, Isotopic production cross sections and recoil velocities of spallation-fission fragments in the reaction $^{238}\text{U}(1\text{AGeV}) + d$, PHYS. REV. C 75, 014602.
- [17] E. Casarejos,1,* J. Benlliure,1 J. Pereira, et al., 2006, Isotopic production cross sections of spallation-evaporation residues from reactions of $^{238}\text{U}(1\text{A GeV})$ with deuterium, PHYS. REV. C 74, 044612 (2006)

Ważniejsze ostatnie publikacje

- A.Wojciechowski, Production of U-232 in thorium cycle, Progress in Nuclear Energy , 106, (2018), 204-214
- A.Wojciechowski, V.Voronko, V.Sotnikov.A.Zhadan, Simultaneous measurement of the neutron and proton induced fissions by activation detectors, Measurement, (2019), 146 (2019) 972–981
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- A. Pacan, B. Słowinski, M. Szuta, A. Wojciechowski, A thorium loaded external neutron source driven setup as a multipurpose tool for nuclear power, Annals of Nuclear Energy 62 (2013) 109–116, (2013)
- M.Bielewicz, E.Strugalska-Goła, M.Szuta, A.Wojciechowski, M.Kadykov, and S. Tyutyunnikov, Measurements of High Energy Neutron Spectrum (>10 MeV) by Using Yttrium Foils in a U/Pb Assembly, Nuclear Data Sheets 119 (2014) 296-298