Investigation of low- and high-energy region of the spallation neutron spectra using ^{235}U and ^{209}Bi activation detectors

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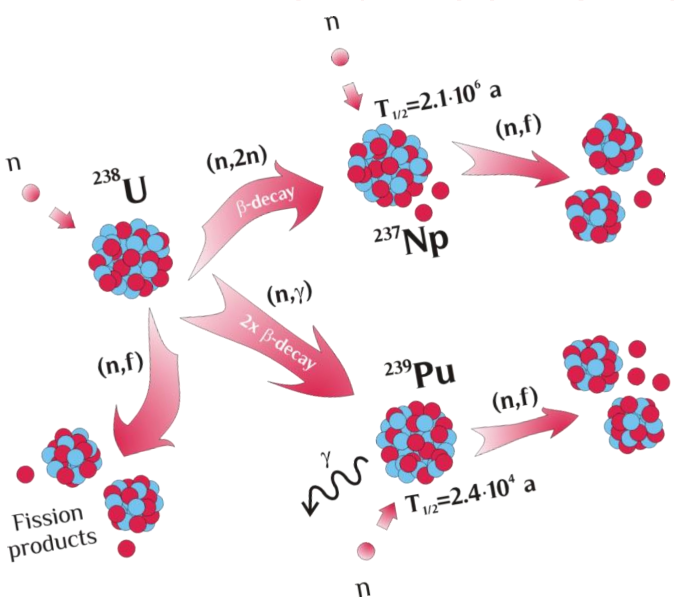
& other colleagues of the «E&T-RAW» collaboration

Outline

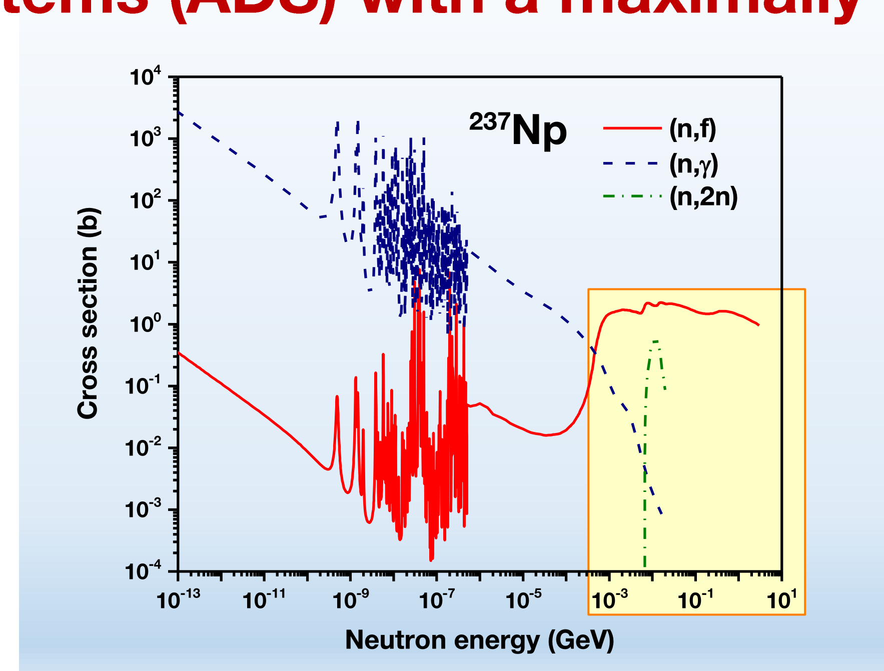
- Introduction
  - Motivation and goals
- Interaction of proton and deuteron beams with a massive uranium spallation target
  - Measurement of characteristics of neutron spectra using natural and enriched uranium activation detectors
  - Experimental determination of neutron flux with ^{209}Bi threshold activation detectors
- Validation of Monte Carlo radiation transport program MCNPX 2.7 and deterministic codes TALYS 1.6 and EMPIRE-3.2 Malta

Motivation

- **ADS Experimental research**
  - **Fundamental research on electronuclear production of neutrons for nuclear energy generation and spent nuclear fuel transmutation in the deep subcritical Accelerator Driven Systems (ADS) with a maximally hard neutron spectrum**



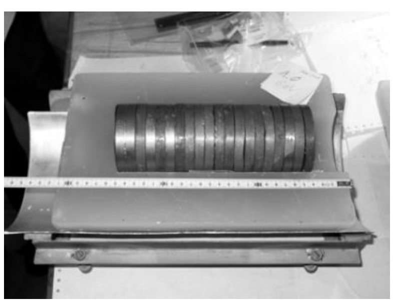
~~(n,γ)~~
(n,f)
(n,2n)



- **Validation of physics models in Monte Carlo codes**

Spallation targets @ Dubna

GAMMA-2
U/Pb + paraffin



M.Zamani+, J. of Physics: Conf.Ser. **41** (2006) 475

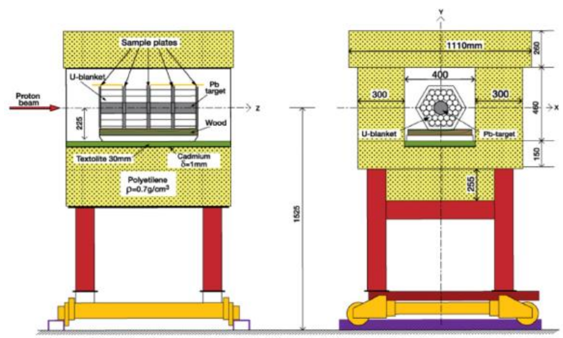
1995

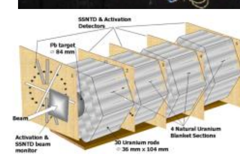
GAMMA-3
Pb + graphite

N.L.Asquith+, Rad. Meas. **67** (2014) 15
J.Adam+, EPJ A **47** (2011) 85

2000

Energy + Transmutation
U+ Pb + polyethylene

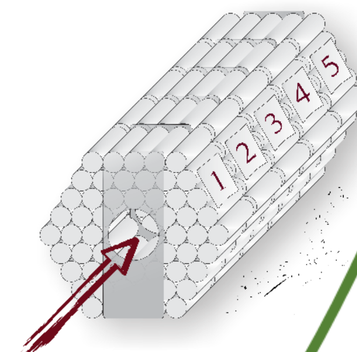




S.R.Hashemi-Nezhad+, NIM A **591** (2008) 517
J.J.Borger+, Rad. Meas. **46** (2011) 1765
J.Adam+, EPJ A **43** (2010) 159

2005

QUINTA
^{nat}U (+Pb)



L.Zavorka+, Annals of Nucl. Ene. **80** (2015) 178

2010

BURAN
depleted U

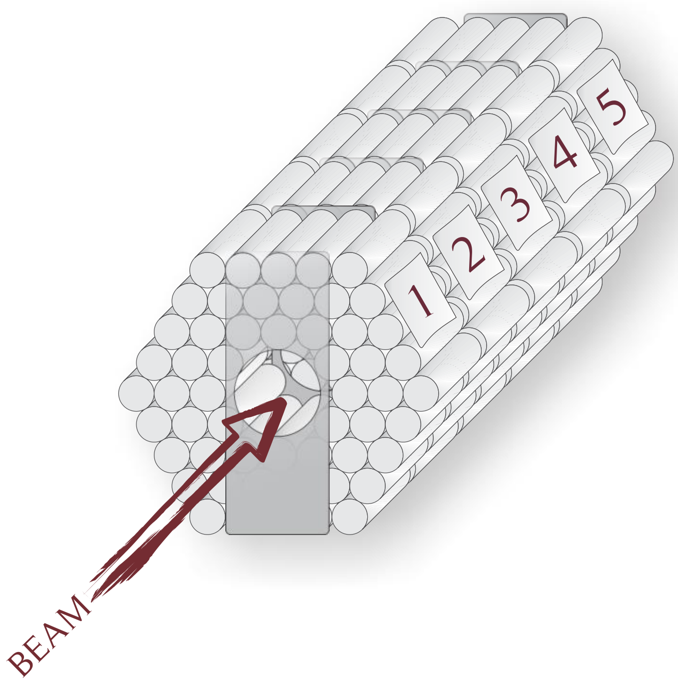
2015

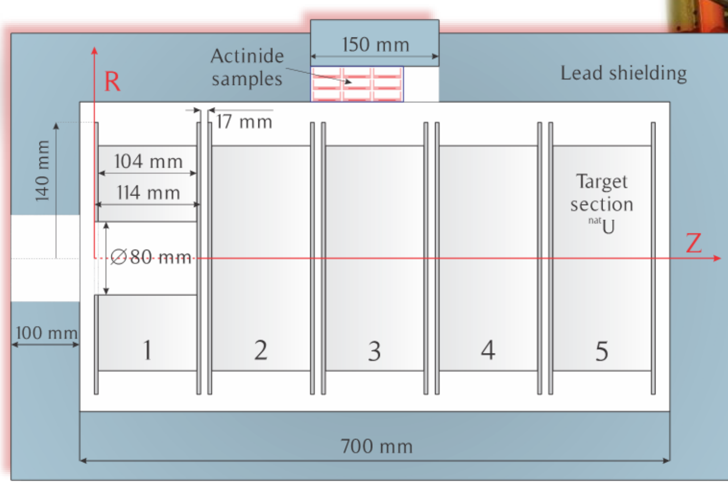
Current research on ADS @ Dubna

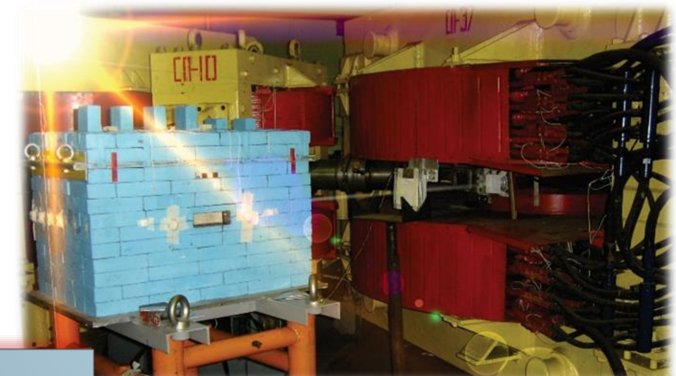
- Investigation of neutron production in a concept of Relativistic Nuclear Technology for nuclear energy production and transmutation of spent nuclear fuel (SNF) in a maximally hard neutron spectrum
  - Deep subcritical core composed of thorium or natural / depleted uranium with SNF elements
  - Contrary to a classical ADS scheme, the energy of primary beam will be increased to ~ 10 GeV
  - High-temperature helium coolant
- Since 2010, the research has been conducted at the massive natural uranium spallation target QUINTA that represents a central region of a true quasi-infinite spallation target for ADS purposes

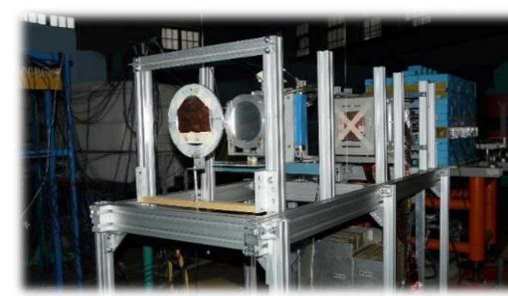
QUINTA target assembly

- **512 kg of metallic natural uranium in aluminum envelope and 10 cm thick lead shielding**
- **Irradiated with 660 MeV proton beam (0.4 μA) and relativistic deuteron beams 2 and 4 AGeV**
- **Calculated neutron leakage ~ 80%**
- **Beam Power Gain ≅ 2**







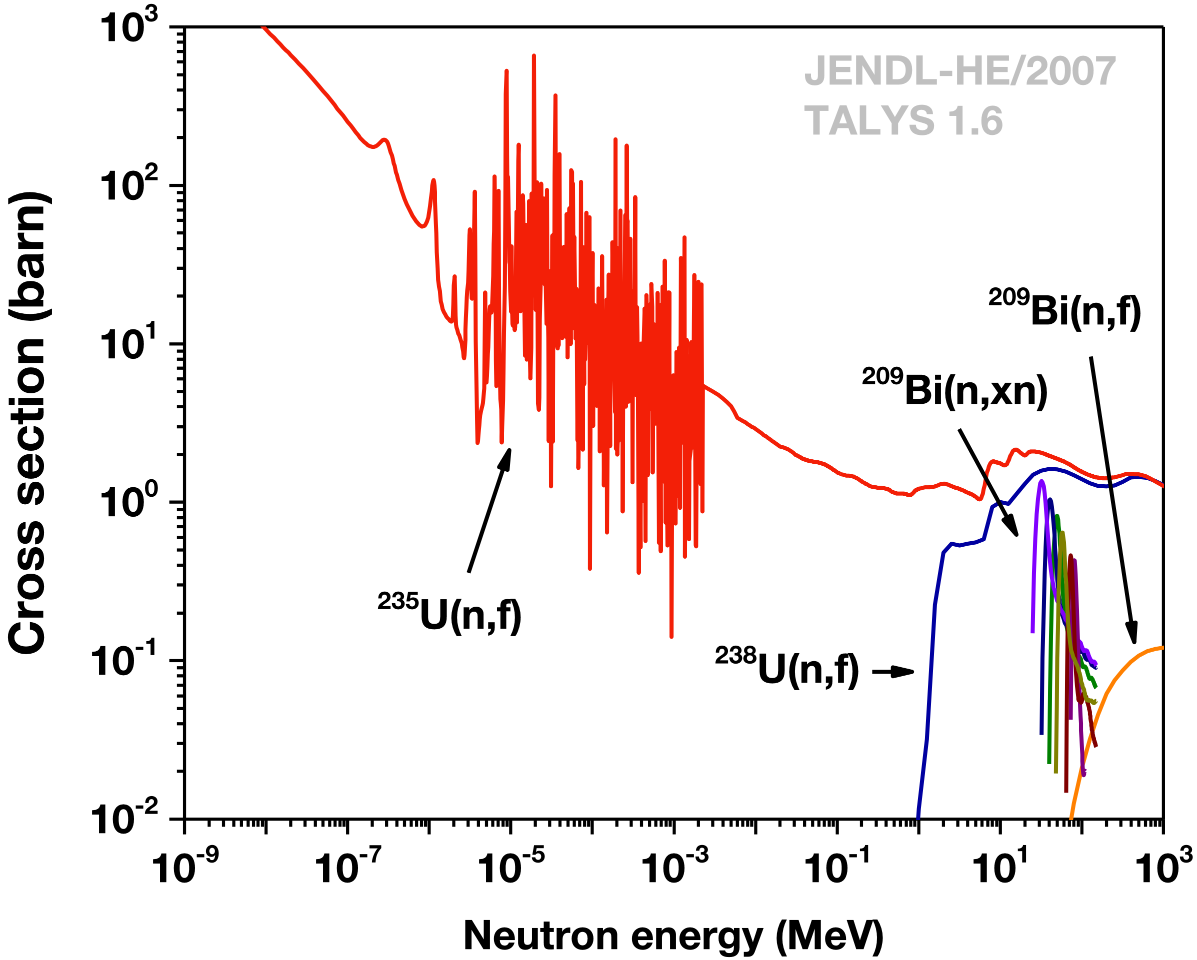


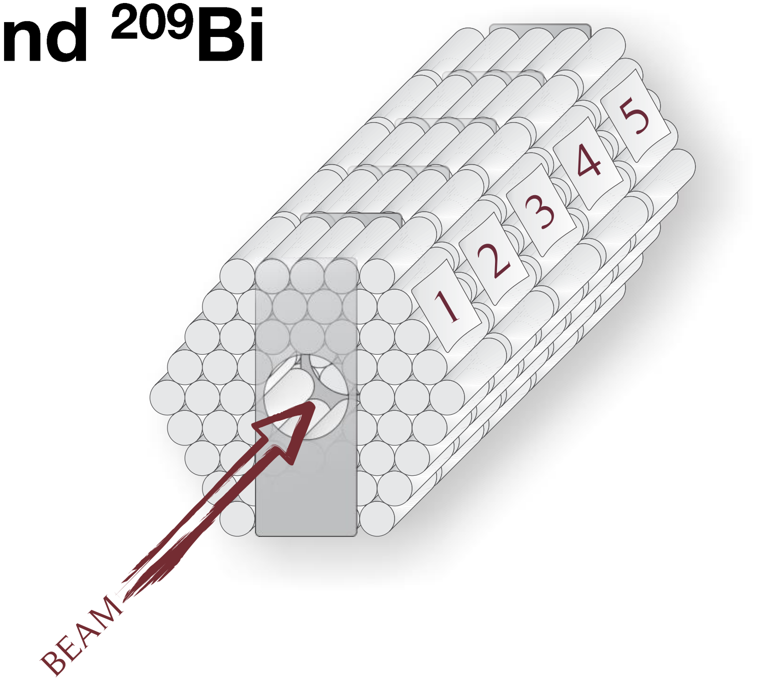
QUINTA target

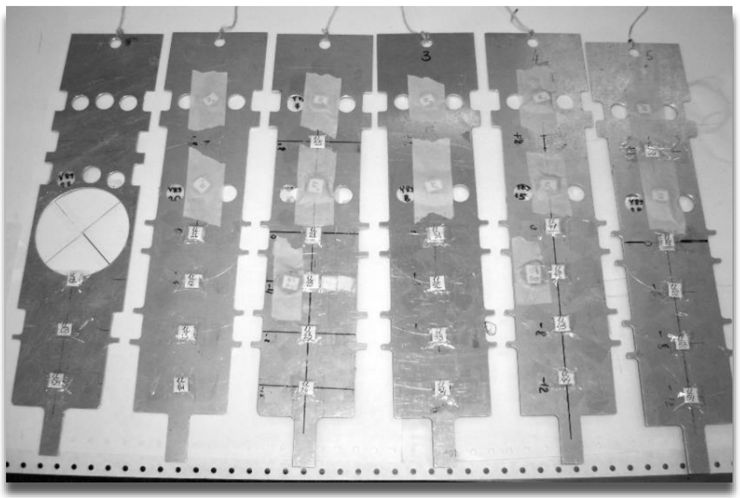
- **Experimental physics programme:**
  - Determination of a total number of fissions as well as production of ^{239}Pu
  - Role of ^{235}U fission in ^{nat}U spallation target
  - High-energy fission in ^{232}Th and ^{209}Bi detectors
  - Spectral indices of (n,f), (n,γ), and (n,2n) reactions in ^{238}U
  - Spectral characteristics of neutron spectrum based on results of threshold activation detectors (Al, Au, Bi, Co)
  - Total leakage of neutrons from the surface of the target
  - Transmutation of actinides (^{237}Np, $^{238,\,239}Pu$, ^{241}Am) and long-lived fission products (^{129}I)
  - Comparison with the results of simulations of MC codes MCNPX 2.7 (INCL4/ABLA) and MARS15 (LAQGSM03.03)

Activation detectors

- **Cross section in ^{235}U, ^{238}U, and ^{209}Bi**





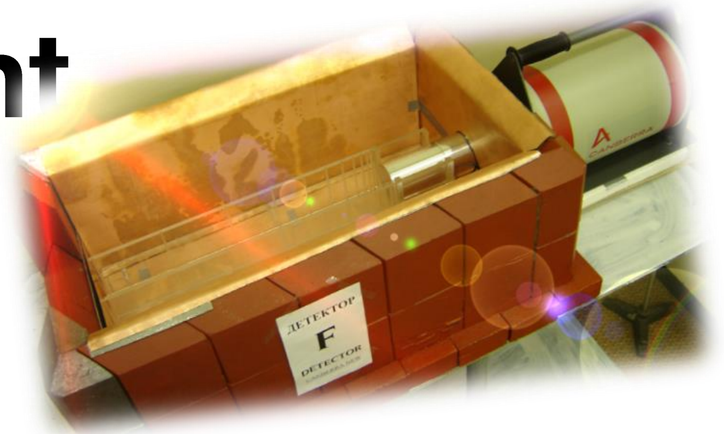


- **Samples of 1 ÷ 5 g in mass across the whole volume of the QUINTA target**

Experimental methods

- **Activation measurement technique**
- **Gamma-ray spectroscopy with the use of both planar and coaxial, P- and N-type HPGe detectors Canberra and ORTEC of up to 35% relative efficiency**
- **Calibrated with standard point gamma-ray sources from 5 keV to 3 MeV**
  - **Gamma-ray sources: ^{22}Na, ^{54}Mn, ^{57}Co, ^{60}Co, ^{65}Zn, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{133}Ba, ^{137}Cs, ^{139}Ce, ^{152}Eu, ^{228}Th, and ^{241}Am**
- **Efficiency compared with MC simulation, which is used for performing corrections for volume emitters**

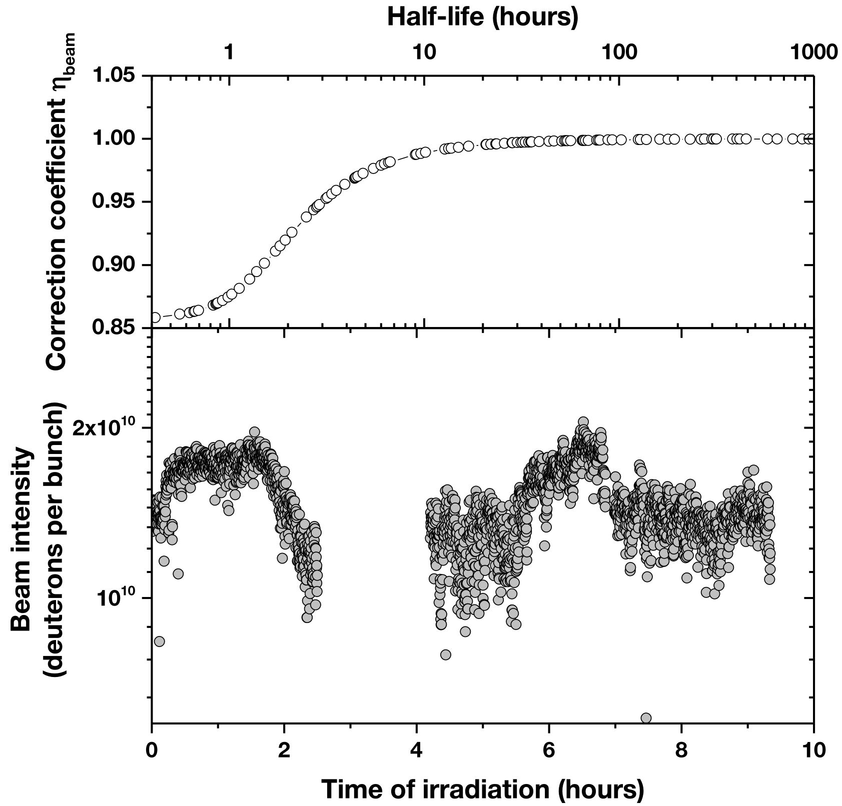
Activation measurement

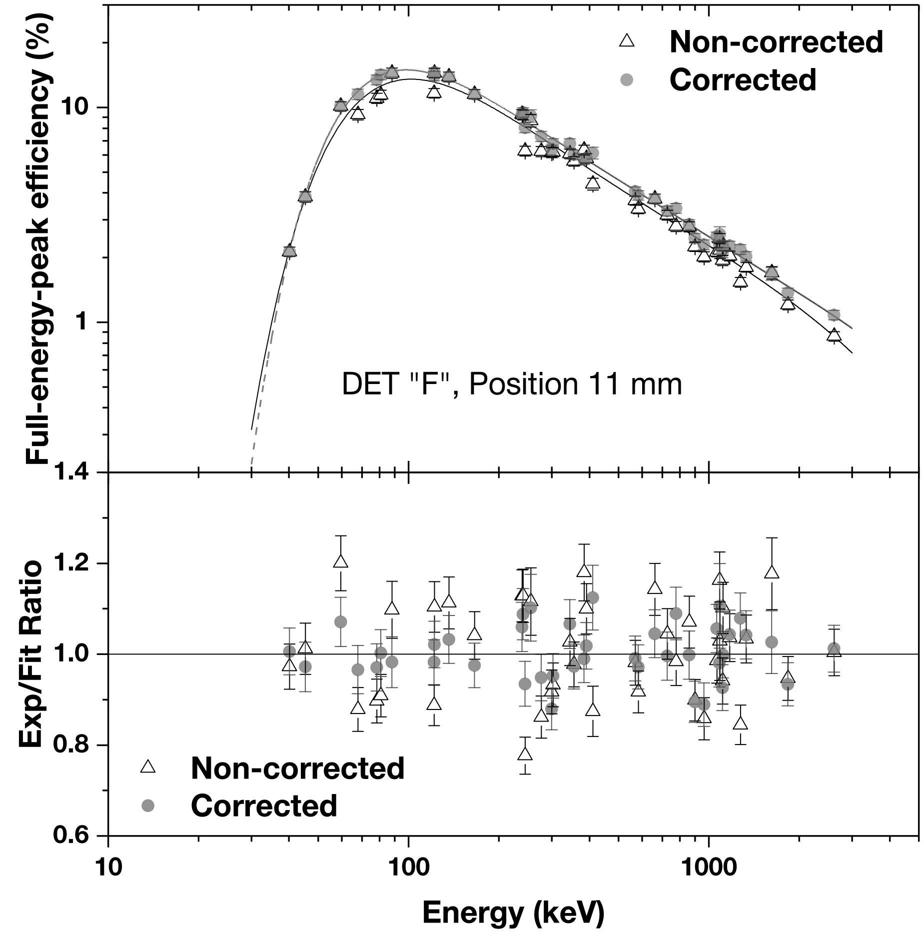


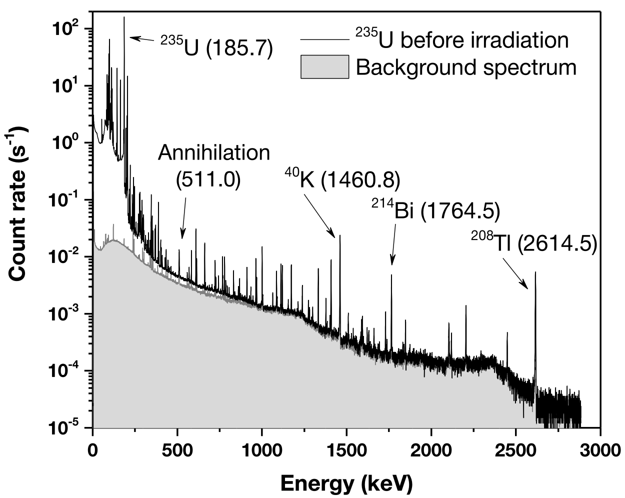
- Identification of products:
  - Half-life (min. 6 measurements)
  - Energy and intensity of gamma line
  - Reaction rates R calculated from measured activity
  - The following correction factors considered:

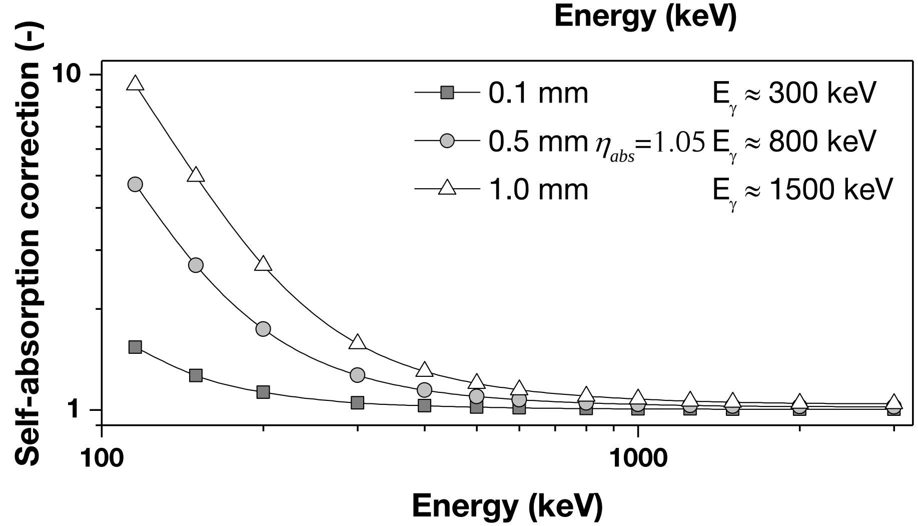
    background radiation, decay during irradiation, cooling and measurement, dead time, nonlinearity, detector efficiency, beam instability, nonpoint-like source, self-absorption, true coincidence summing

Correction factors

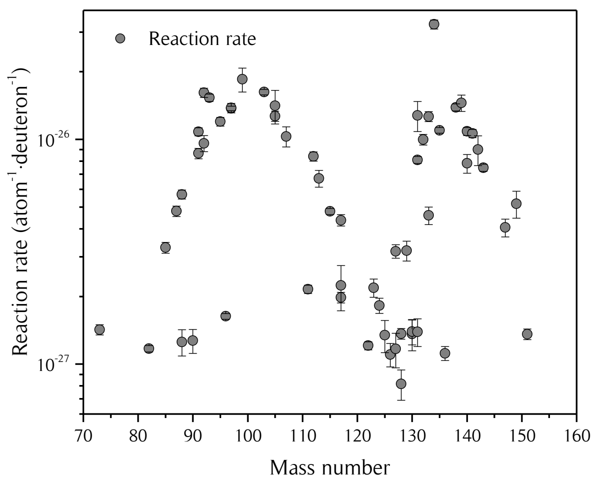


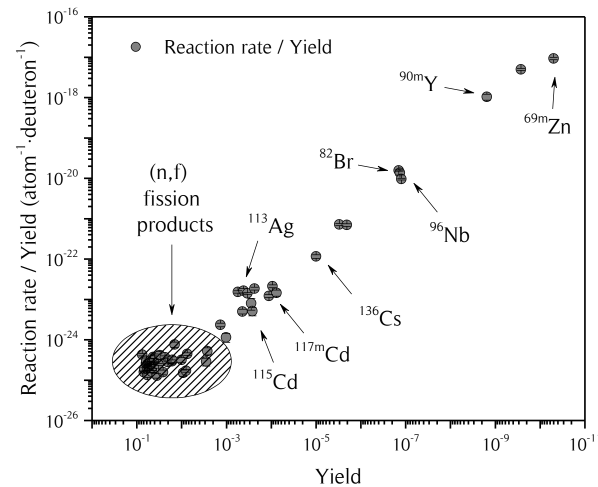






Fission rate determination

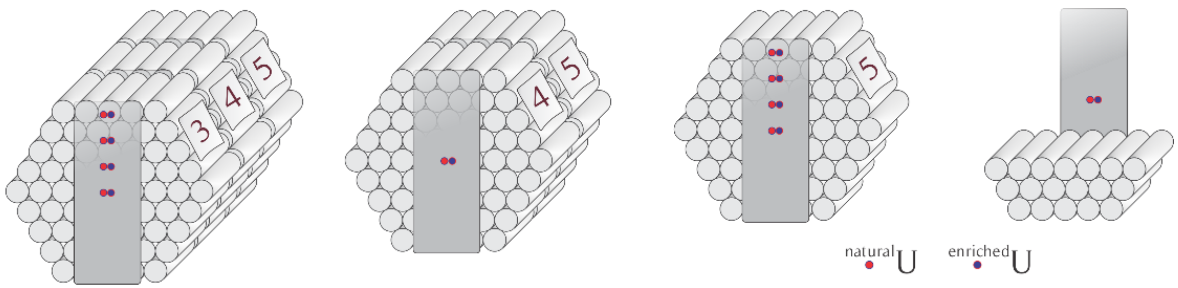




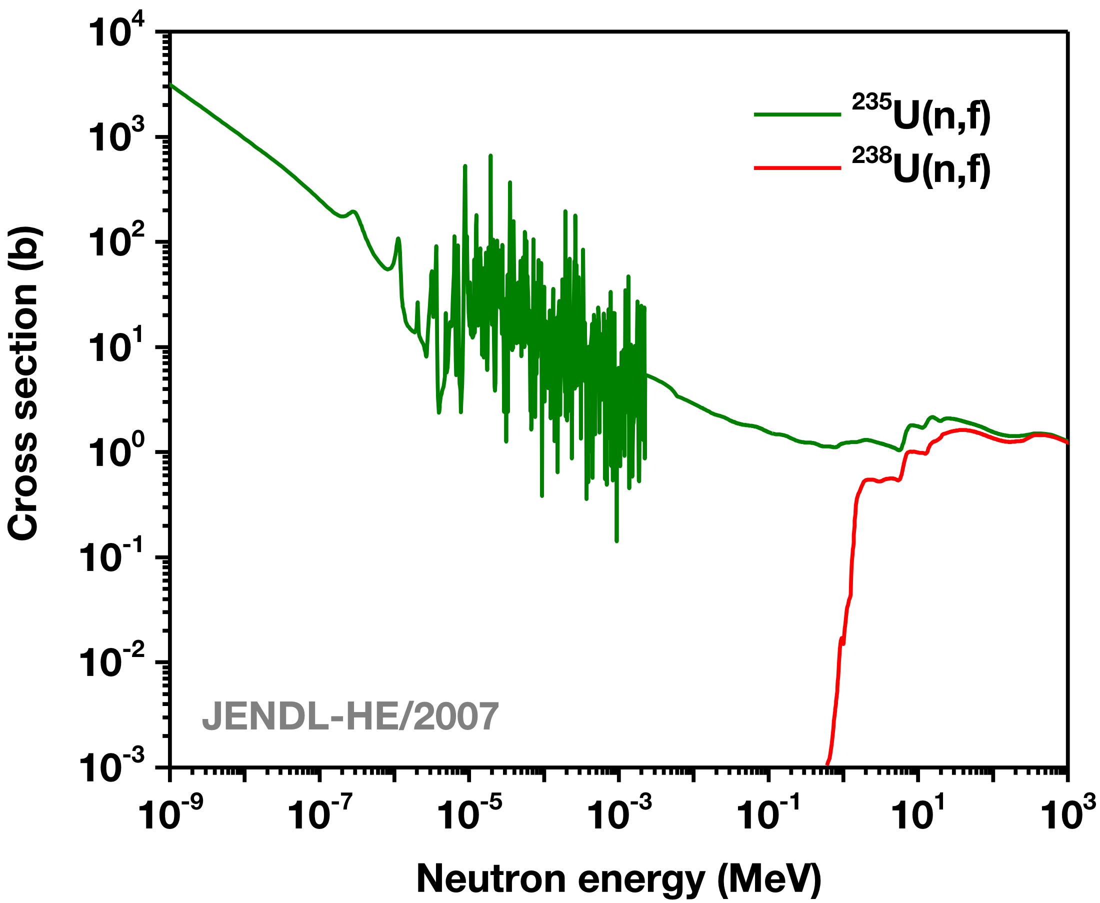
- **In measurements of uranium samples more than 70 fission products detected**
- **Proper selection of 18 fission products with suitable half-life, gamma-ray energy and intensity and cumulative fission yield:**

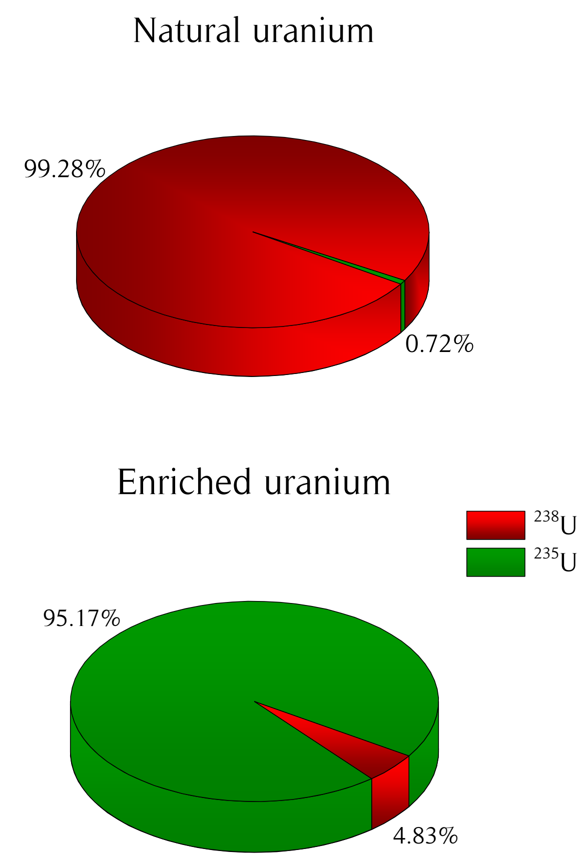
^{87}Kr, ^{88}Kr, ^{91}Sr, ^{92}Sr, ^{93}Y, ^{95}Zr, ^{97}Zr, ^{99}Mo, ^{103}Ru, ^{105}Ru, ^{129}Sb, ^{131}I, ^{133}I, ^{135}I, ^{140}Ba, ^{143}Ce, ^{147}Nd, and ^{149}Nd

Role of ^{235}U fission in ^{nat}U target

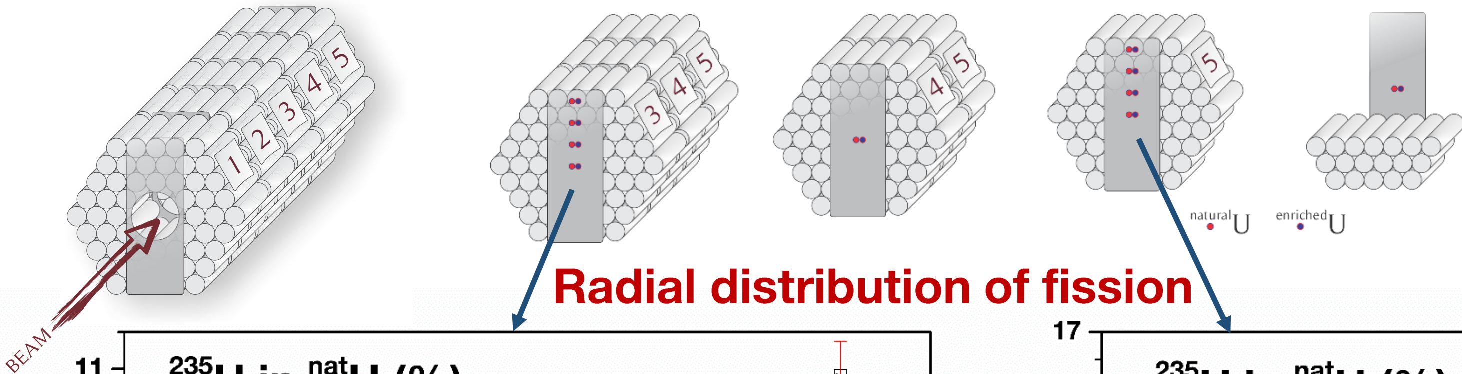


^{235}U vs ^{238}U

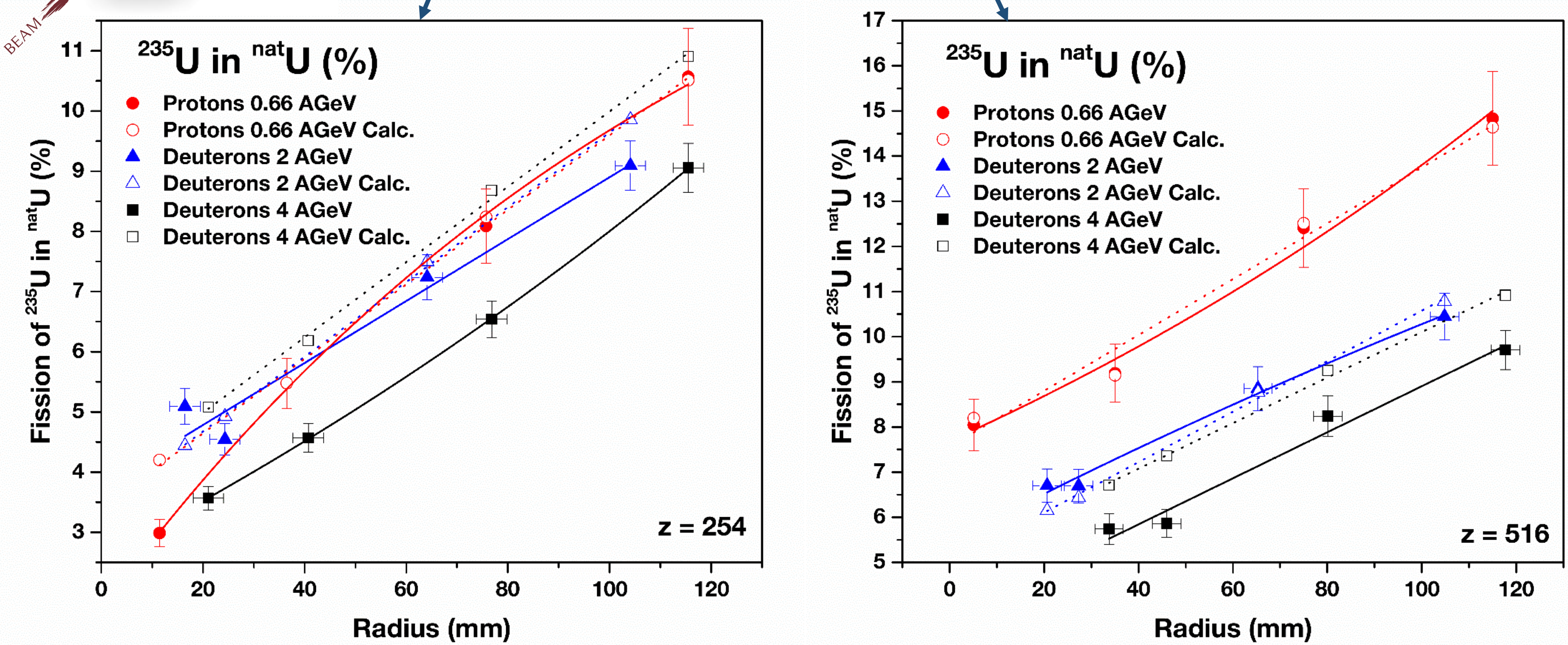




Role of ^{235}U fission in ^{nat}U target



- Contribution of ^{235}U to the total number of fission in QUINTA is 4.9(4) % for 660-MeV protons



- **Neutron spectrum becomes harder with increase of beam energy**

Bismuth threshold detectors

Eur. Phys. J. A **23**, 61–68 (2005)
DOI 10.1140/epja/i2004-10031-y

THE EUROPEAN PHYSICAL JOURNAL A

Spallation neutron spectrum on a massive lead/paraffin target irradiated with 1 GeV protons

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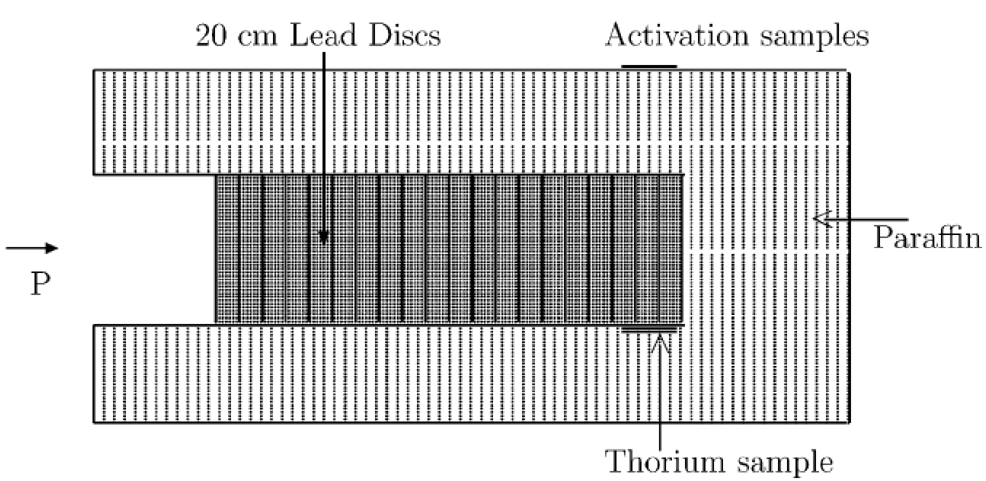
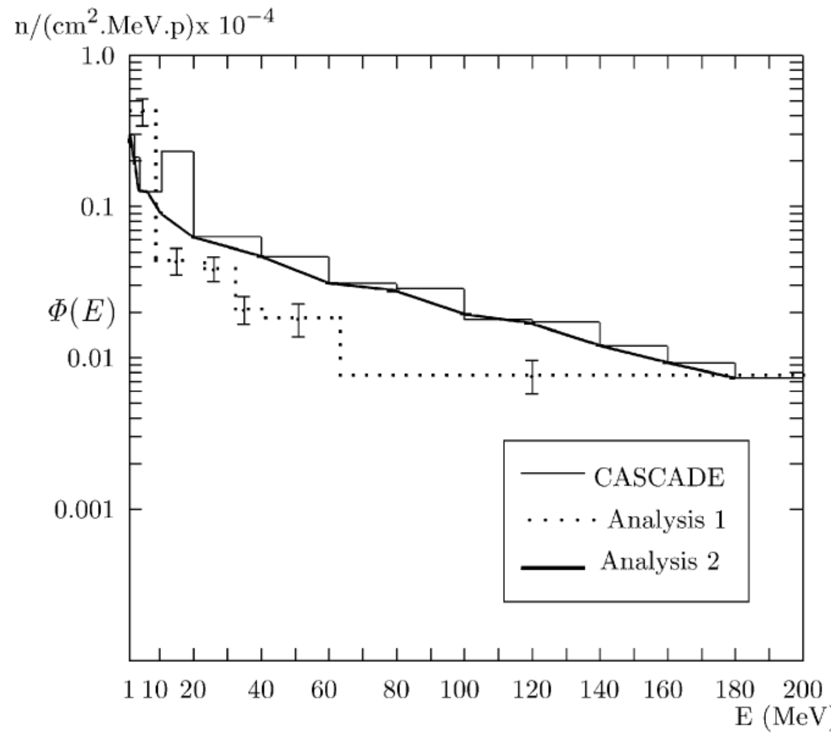




Bismuth threshold detectors

- **Pure experimental determination of spallation neutron spectrum initiated by collisions of 660 MeV protons using ^{209}Bi detectors**

Reaction	E_{theff} (MeV)	Half-life
^{209}Bi(n,3n)^{207}Bi	16.2	31.6 y
^{209}Bi(n,4n)^{206}Bi	24.8	6.2 d
^{209}Bi(n,5n)^{205}Bi	32.7	15.3 d
^{209}Bi(n,6n)^{204}Bi	40.9	11.2 h
^{209}Bi(n,7n)^{203}Bi	48.7	11.8 h
^{209}Bi(n,9n)^{201}Bi	64.2	1.7 h

$$R_{exp} = \int_{E_{th}}^{\infty} \sigma(E) \cdot \phi(E) \cdot dE$$

$$R_9 = \phi(9) \int_{E_{th(n,9n)}}^{E_{max}} \sigma_9(E) \cdot dE$$

$$\Rightarrow \phi(9) = \frac{R_9}{\int_{E_{th(n,9n)}}^{E_{max}} \sigma_9(E) \cdot dE}$$

$$R_8 = \phi(8) \int_{E_{th(n,8n)}}^{E_{th(n,9n)}} \sigma_8(E) \cdot dE + \phi(9) \int_{E_{th(n,9n)}}^{E_{max}} \sigma_8(E) \cdot dE$$

$$\Rightarrow \phi(8) = \dots$$

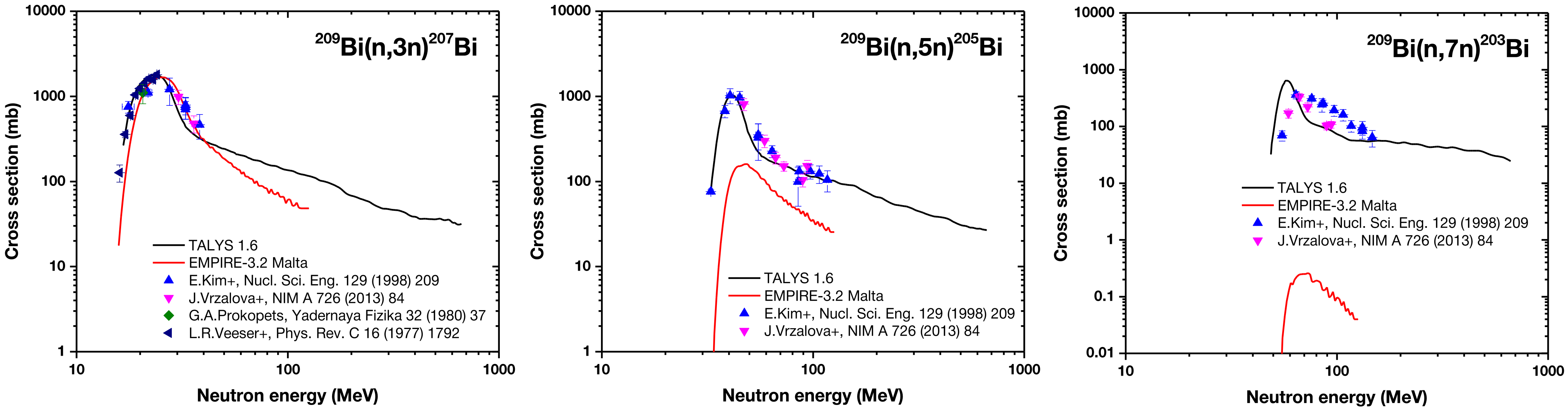
No initial guess flux in this approach!

- **Validation of stochastic and deterministic codes MCNPX 2.7 and TALYS 1.6 in a <u>very important energy region above 20 MeV</u>**

INCL4/ABLA

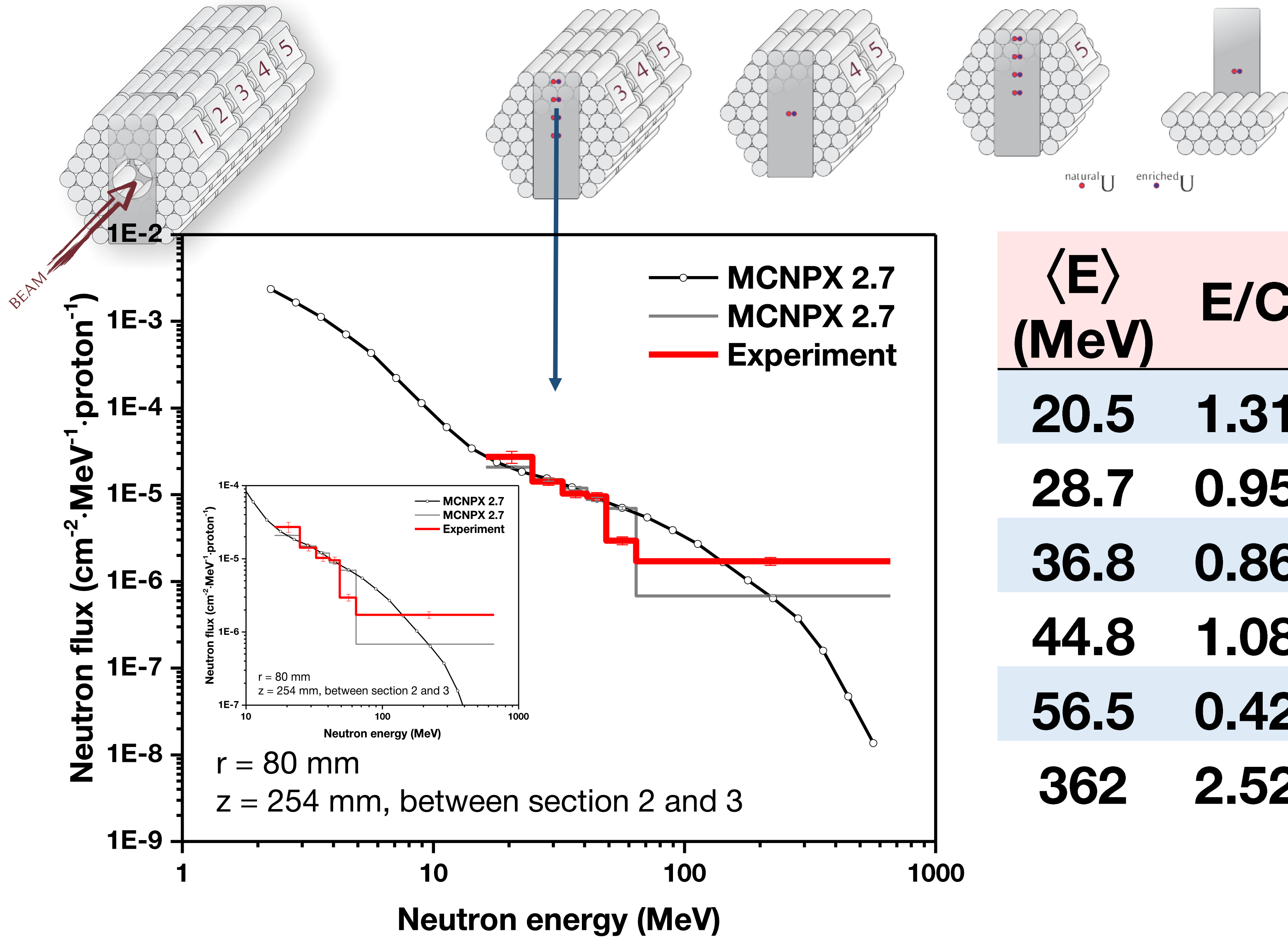
Cross section determination

- Cross section of (n,xn) reactions calculated with **TALYS 1.6** and **EMPIRE-3.2 Malta** codes and compared to the data available in EXFOR



- **EMPIRE is not suitable for our purposes**

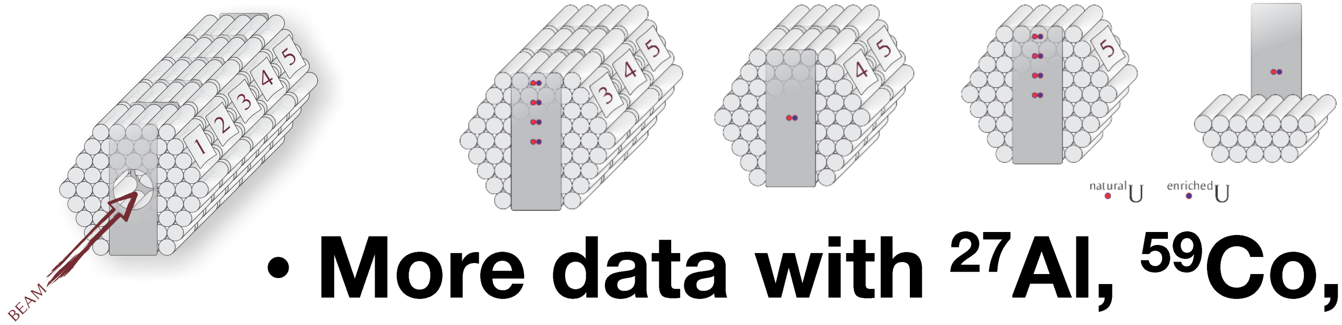
Spallation neutron spectra



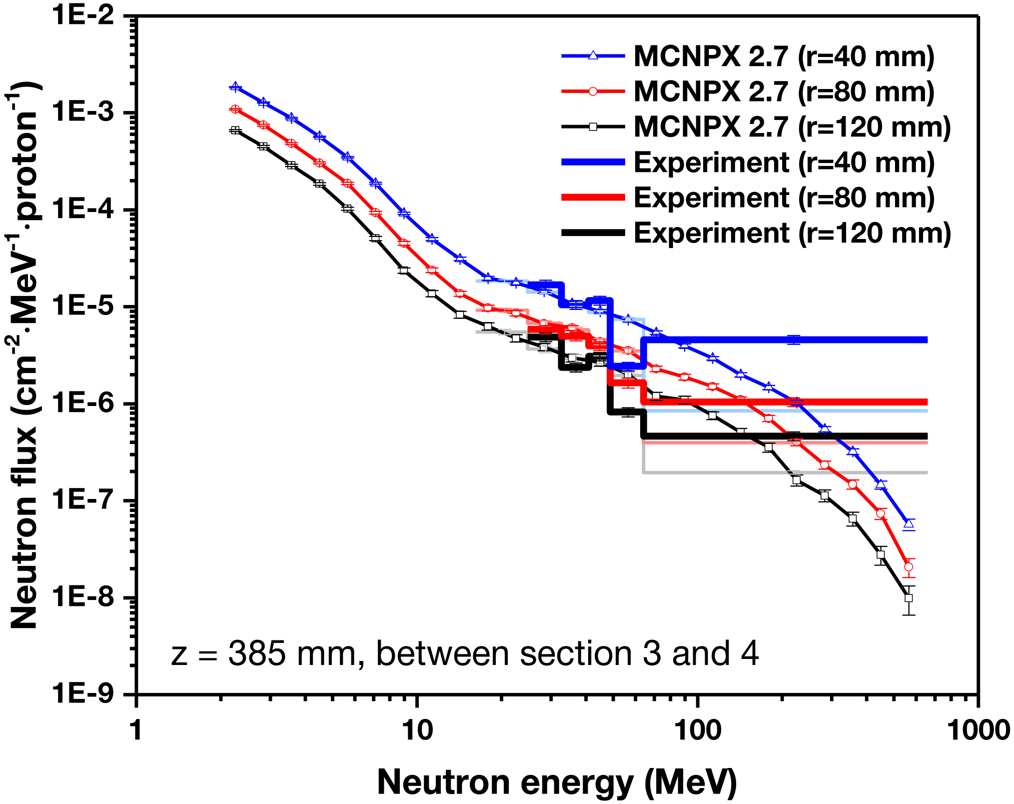
⟨E⟩ (MeV)	E/C	σ
20.5	1.31	0.21
28.7	0.95	0.10
36.8	0.86	0.09
44.8	1.08	0.12
56.5	0.42	0.05
362	2.52	0.27

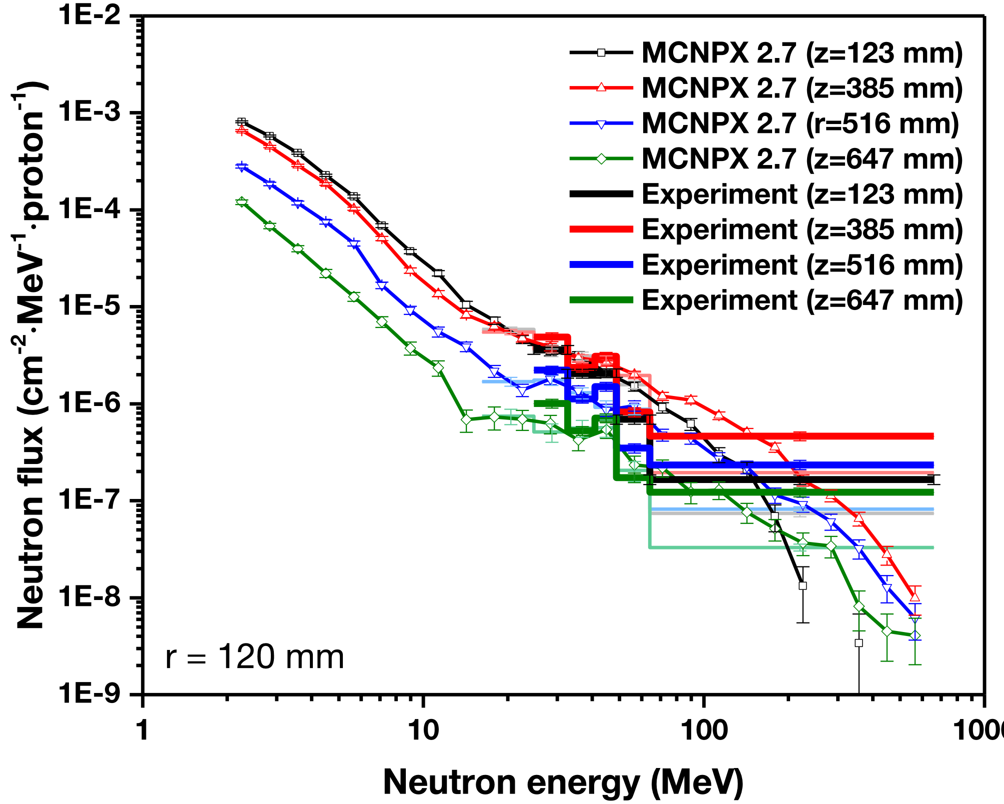
- **Experiment and MCNPX 2.7 with TALYS 1.6 data**

Spallation neutron spectra



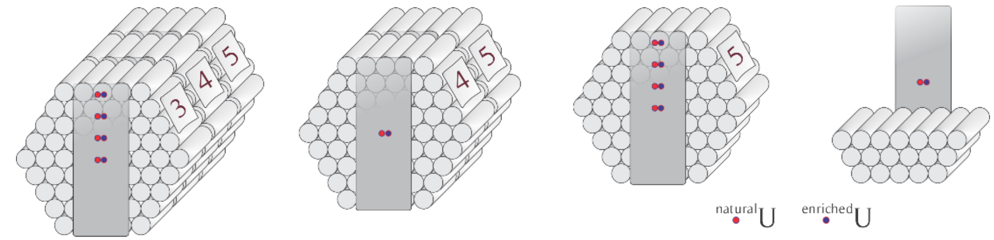
- More data with ^{27}Al, ^{59}Co, ^{89}Y, ^{197}Au





- Fission of ^{209}Bi (R~10^{-30}) detected in one sample

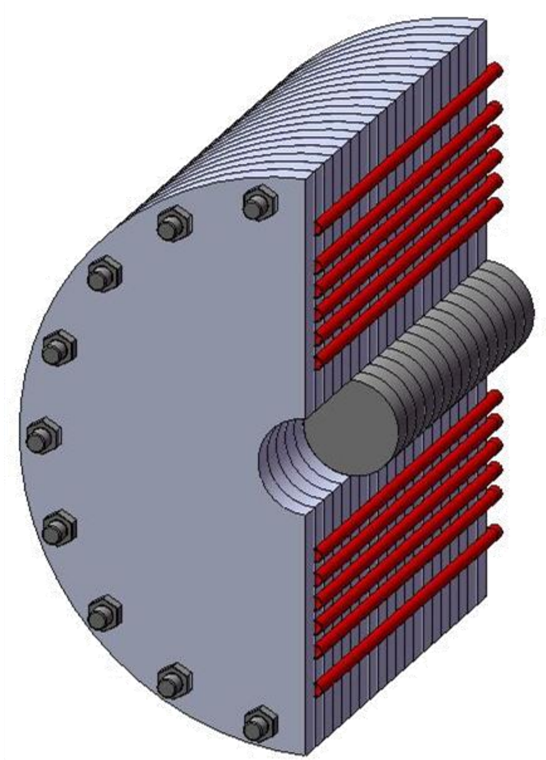
Conclusion

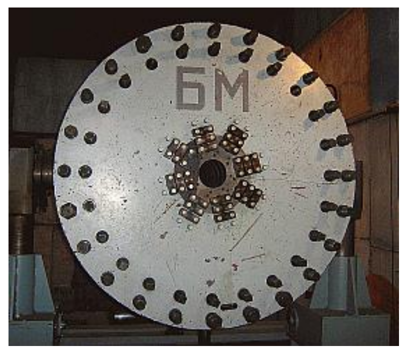


- **TALYS 1.6 provides reliable cross sections of (n,xn) reactions in ^{209}Bi and MCNPX 2.7 (INCL4/ABLA) generates reasonably good neutron spectra (above 20 MeV) in collisions of 660 MeV protons with the massive uranium spallation target**
- **Monte Carlo programs do not describe the increase in mean energy of neutron spectrum inside the spallation target as a function of deuteron energy, which was experimentally confirmed at the massive U target**
- **Measurement of fission of ^{209}Bi requires increase in beam integral by two orders of magnitude**

Spallation target BURAN

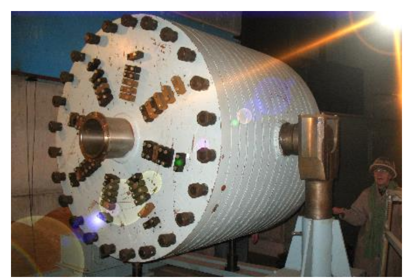
- **20 tons of depleted uranium (central part repl.)**
- **Simple geometry (diam. 1.2 m, length 1 m)**

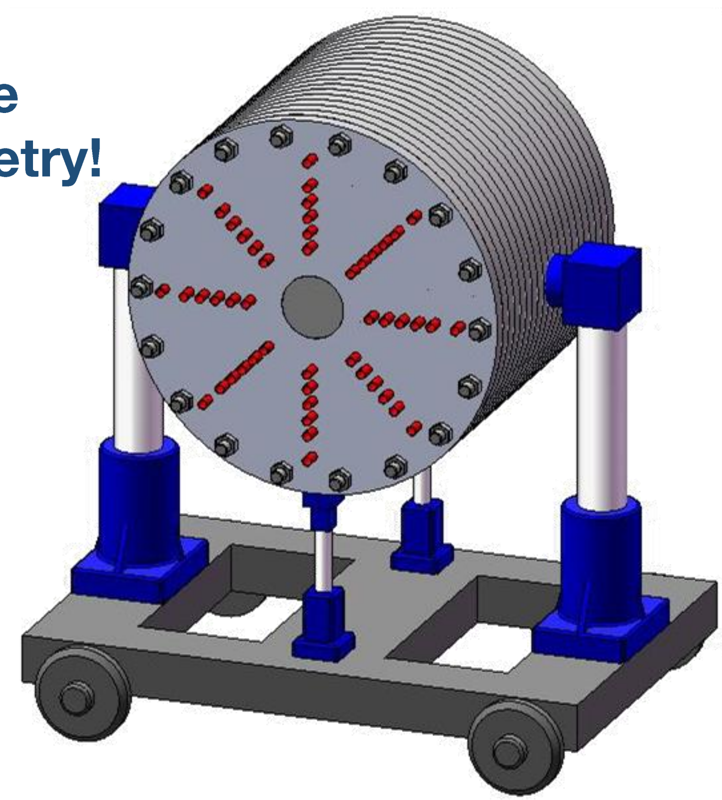




Simple geometry!

Minimal neutron leakage!





- **Validation of various codes and physics models**
- **to be launched in Autumn 2015 (660 MeV protons)**

Thank you for your attention.

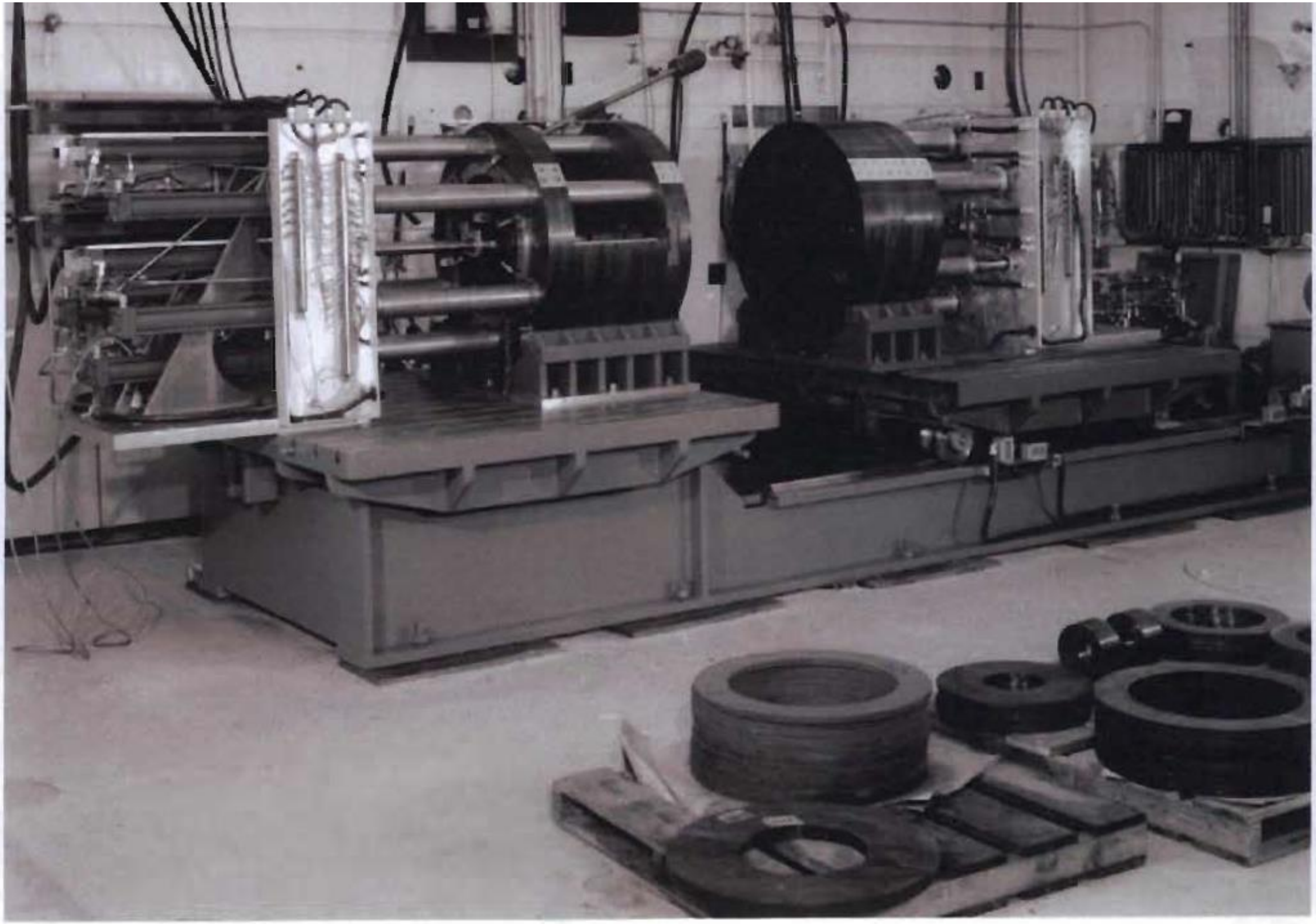


Figure 1. The Big Ten Assembly during Construction (1968).

Production cross sections

- **Spallation reactions play an important role in neutron production for Accelerator Driven Systems (ADS), are responsible for intensive production of radioactive beams using Isotope Separation On-Line (ISOL) technique, serve as a potential source of α-emitting radioisotopes for medical radiotherapy (^{225}Ac, ^{223}Ra)**

- **Measurement of cross sections of reaction residues implementing methods of inverse kinematics at GSI Darmstadt (max. 1 AGeV ^{197}Au, ^{208}Pb, ^{238}U + p/d target)**

or

- **using methods of direct kinematics at JINR Dubna (this experiment) to obtain new experimental data and validate nuclear physics models at the beams of relativistic energies - deuterons up to 3.5 AGeV**

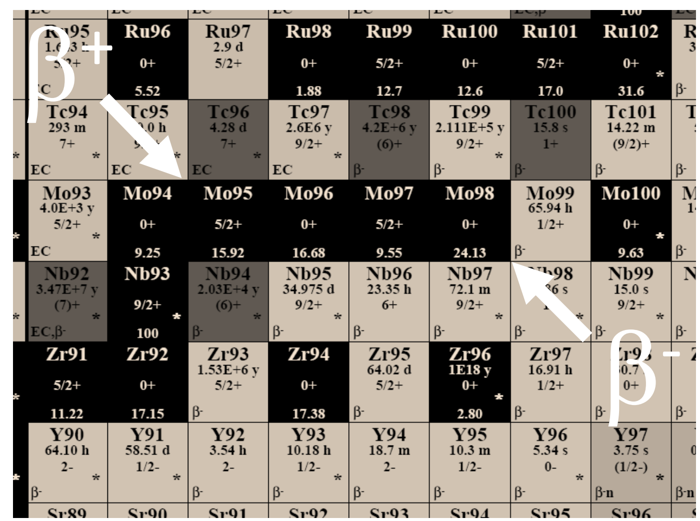
Cross section determination

- **Thin spallation targets made of natural, enriched, and depleted uranium irradiated with 2.2 AGeV and 3.5 AGeV deuteron beams at JINR Nuclotron in 2014 and 2015**
- **Time of irradiation: 23 and 40 hours**
- **Beam integral: 4×10^{9} – 6×10^{11} deuterons**
- **At least 13 measurements at JINR JaSNAPP gamma-ray spectrometry complex with HPGe detectors**
- **Important spectroscopy correction factors considered in careful data analysis**
- **Cross sections calculated from measured activity of the uranium samples**

Cross section determination

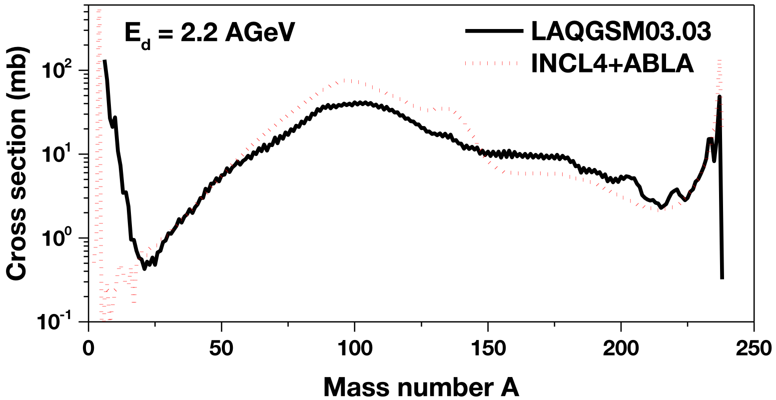
- **Cross sections obtained for a large number of neutron-rich nuclei, including some metastable states of produced residues, as well as some neutron-deficit nuclei and products of quasi-elastic reactions**
- **New experimental data compared to the results of simulations employing physics models available in MCNP6 v 1.0: INCL4/ABLA and LAQGSM03.03**
- **Independent residual nuclei cross sections and mass distributions calculated using GENXS option at TROPT card**

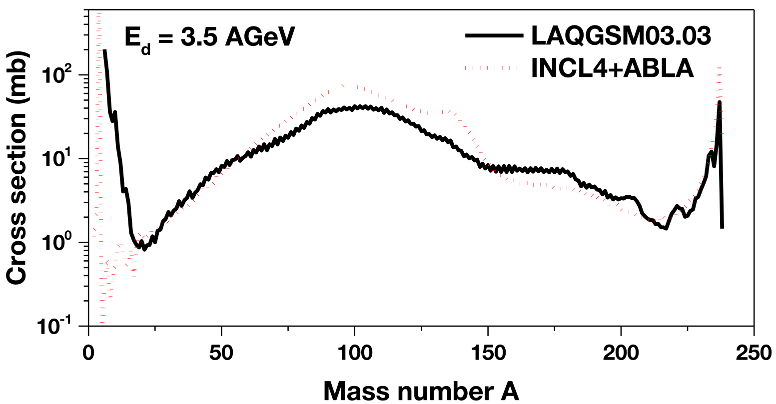
Isotope	^{69m}Zn	^{85m}Kr	^{106m}Ag	^{117m}Cd	^{120m}Sb
$T_{1/2}$	13.76 h	4.48 h	8.28 d	3.36 h	5.76 d



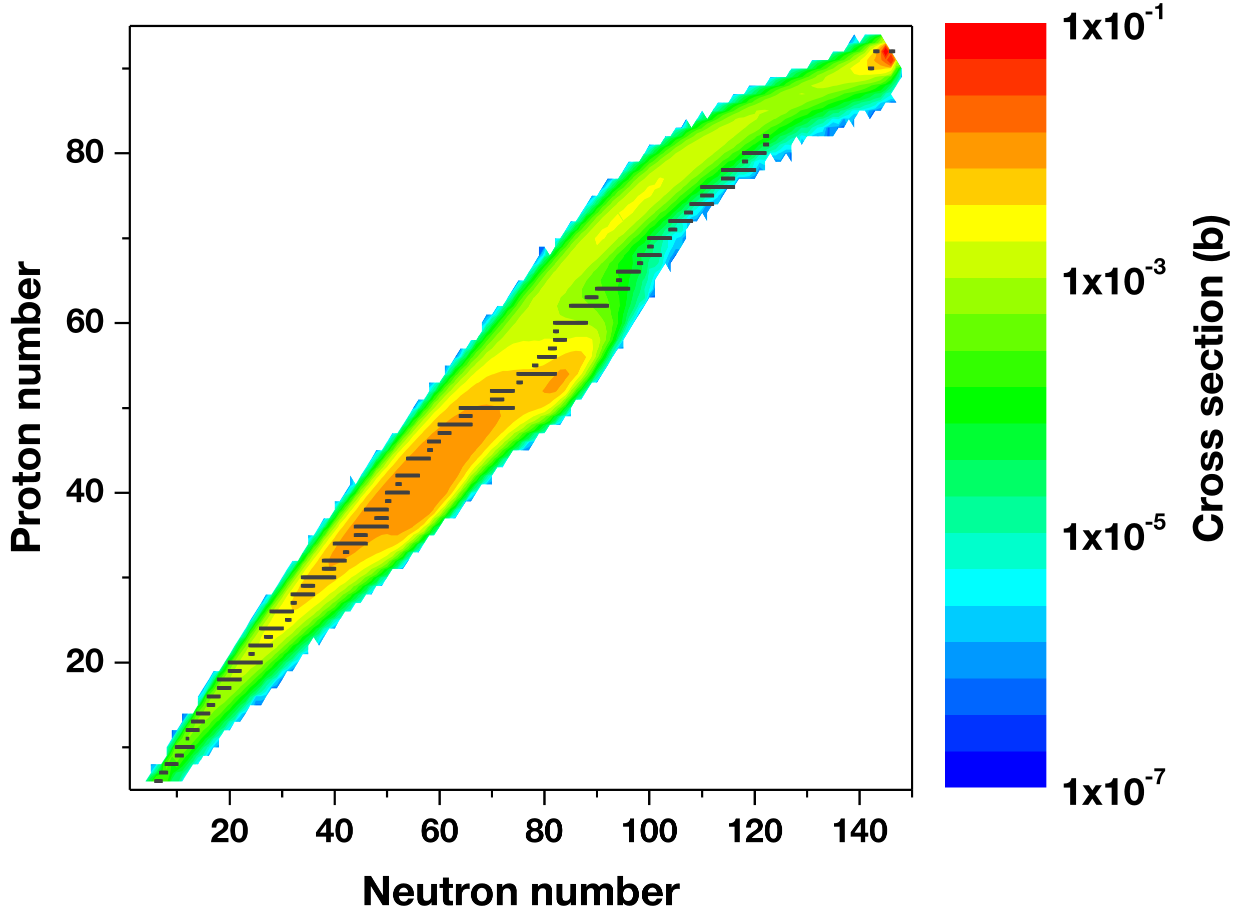
Cross section determination

- Cross sections in ^{nat}U using INCL4/ABLA and LAQGSM03.03 in MCNP6 v 1.0





Residual nuclei cross section in ^{nat}U irradiated with 2.2 AGeV deuterons. INCL4/ABLA used.



Cross section determination

- **Comparison between experiment and simulation employing INCL4/ABLA and LAQGSM03.03 at the deuteron beam energy 3.5 AGeV**