Activation Measurements in Collisions of Proton and Relativistic Deuteron Beams with Uranium Spallation Targets

L. Zavorka, J. Adam, A. A. Baldin, V. V. Chilap, W. I. Furman, J. Khushvaktov, Yu. Kish, V. Kumar, V. S. Pronskikh, A. A. Solnyshkin, M. Suchopar, V. M. Tsoupko-Sitnikov, S. I. Tyutyunnikov, R. Vespalec, J. Vrzalova, M. Zeman

Czech Technical University, Prague, Czech Republic
Joint Institute for Nuclear Research, **Dubna**, Russia
Center of Physics and Technical Projects "Atomenergomash",Russia
G.G.S. Indraprastha University, New Delhi, India
Fermi National Accelerator Laboratory, Batavia, IL, USA
Nuclear Physics Institute of the ASCR, Czech Republic
Brno University of Technology, Brno, Czech Republic

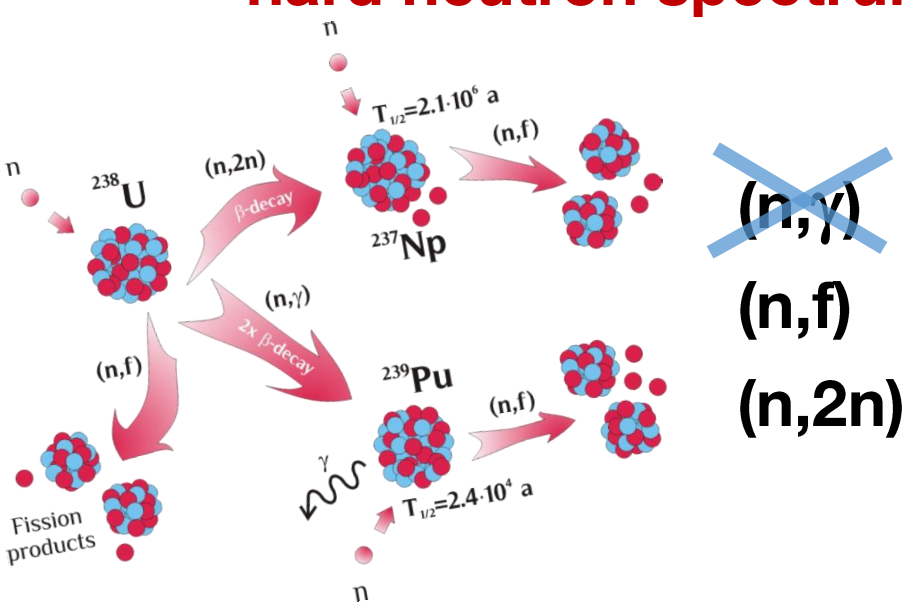
& other colleagues of the «E&T-RAW» collaboration

Outline

- **Introduction**
  - **Motivation and goals**
- **Interaction of proton and deuteron beams with a massive uranium spallation target**
  - **Experimental determination of neutron spectrum characteristics using activation detectors**
  - **Comparison with predictions of MCNPX and TALYS**
- **Collisions of relativistic deuteron beams with thin uranium spallation targets**
  - **Measurement of the production cross sections**
  - **Calculations of XS employing available physics models**

Motivation

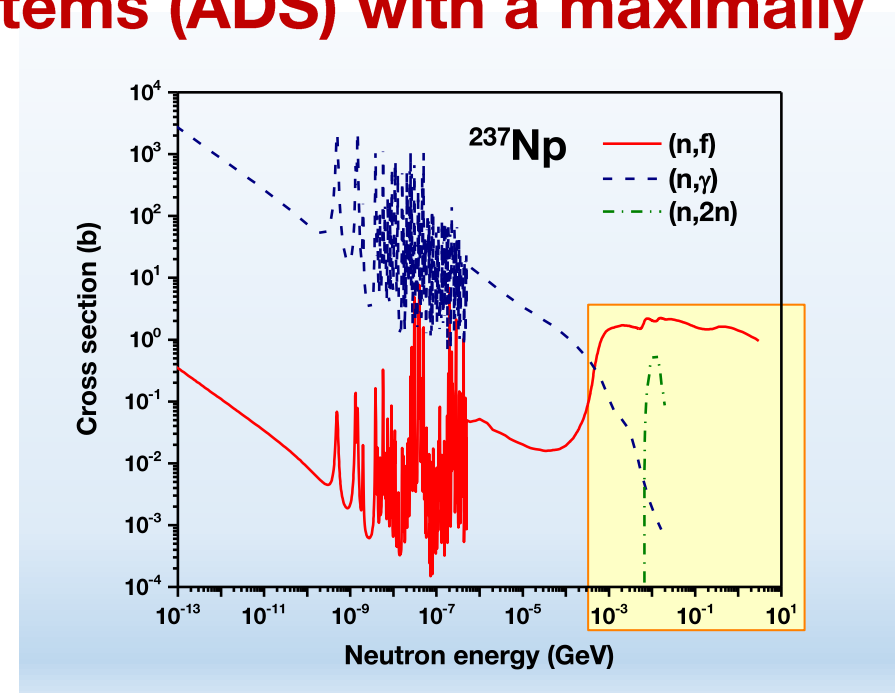
- **ADS Experimental research**
  - **Fundamental research on electronuclear production of neutrons for nuclear energy generation and spent nuclear fuel transmutation in the deep subcritical Accelerator Driven Systems (ADS) with a maximally hard neutron spectrum**



~~(n,γ)~~

(n,f)

(n,2n)

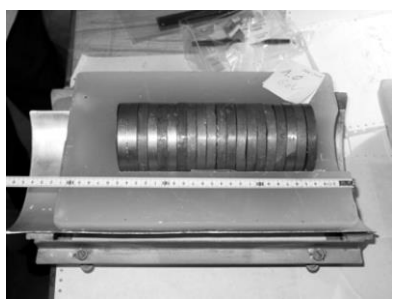


- **Validation of radiation transport codes**

Spallation targets @ Dubna

BURAN
depleted U

GAMMA-2
U/Pb + paraffin

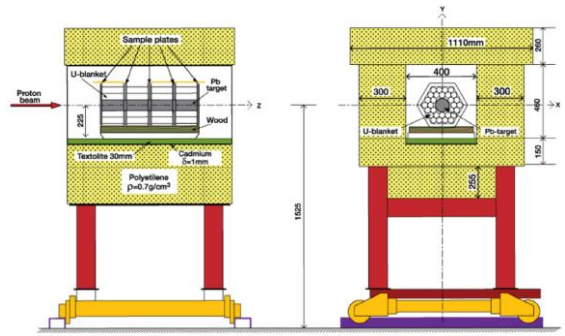


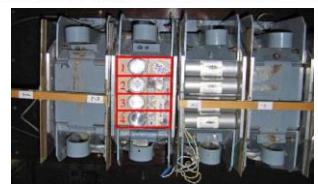
M.Zamani+, J. of Physics: Conf.Ser. **41** (2006) 475

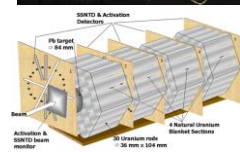
GAMMA-3
Pb + graphite

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

Energy + Transmutation
U+ Pb + polyethylene

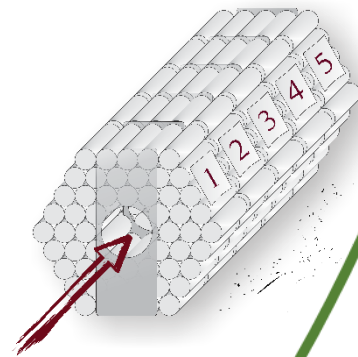






S.R.Hashemi-Nezhad+, NIM A **591** (2008) 517
J.J.Borger+, Rad. Meas. **46** (2011) 1765
J.Adam+, EPJ A **43** (2010) 159

QUINTA
^{nat}U (+Pb)

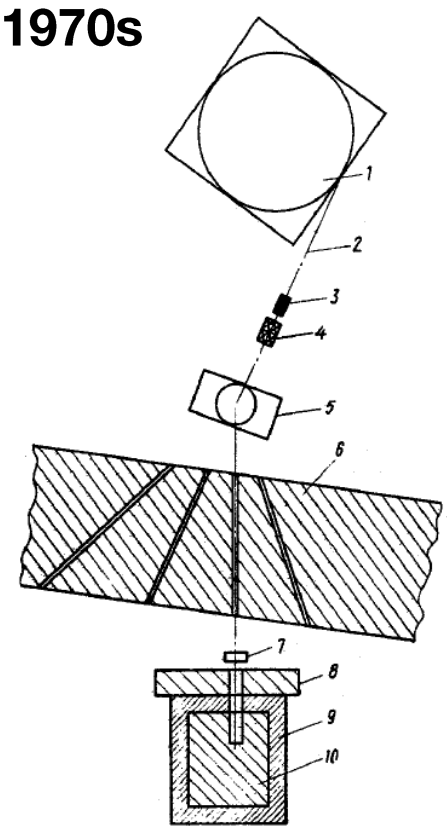


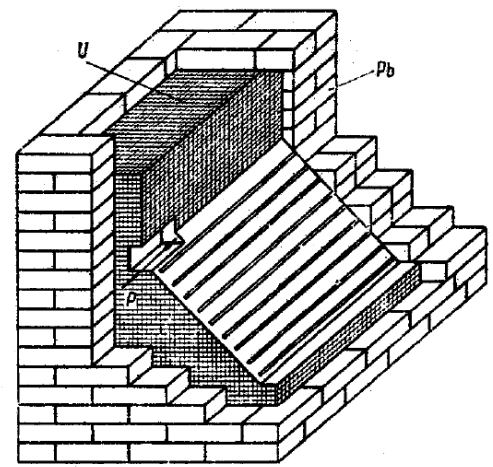
L.Zavorka+, Annals of Nucl. Ene. **80** (2015) 178

1995 2000 2005 2010 2015

Spallation targets @ Dubna

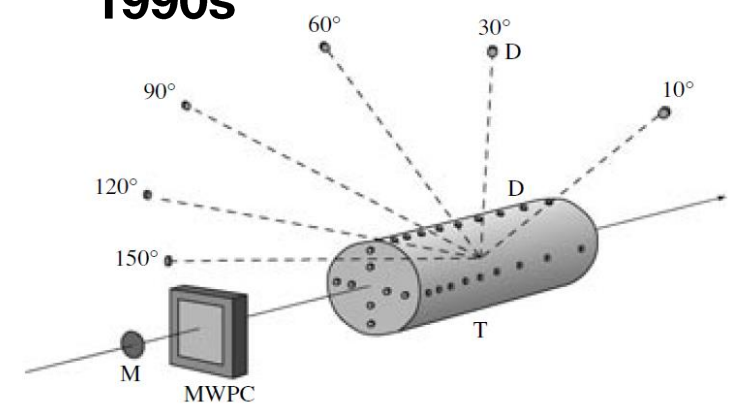
1970s





R.G.Vasilkov+, Atomnaya Energiya **44** (1978) 329

1990s



V.I.Yurevich+, Phys. Part. Nucl. Lett. **3** (2006) 169

ø 20 x 60 cm lead target, relativistic proton and deuteron beams

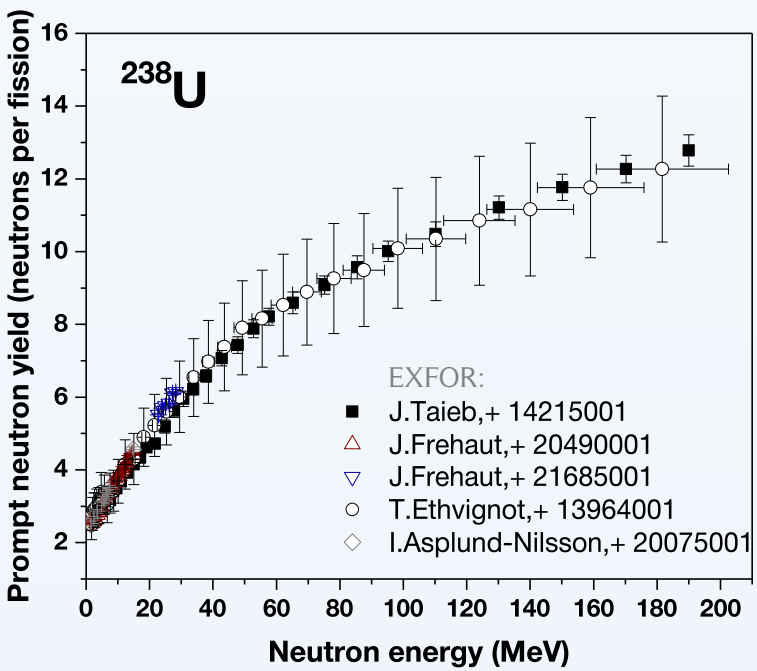
E_d (GeV)	$\langle E_n \rangle$ (MeV)	W/E_d (%)
1.03	6.5	32.6
1.98	7.9	43.9
3.76	10.4	45.7

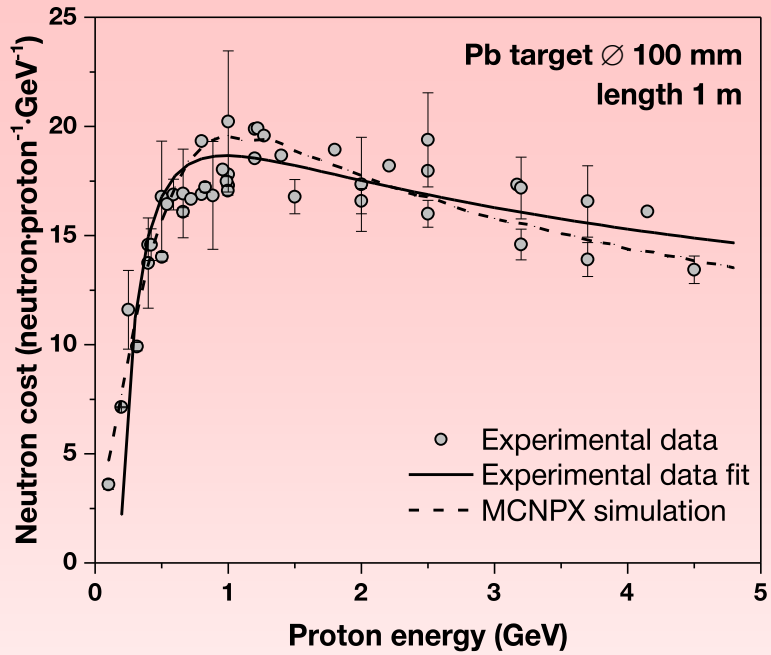
- **Natural / depleted uranium**
- **Intense proton beams with energy up to 660 MeV**
- **Beam Power Gain max. 7.4**

- **Increase in mean neutron energy $\langle E_n \rangle$ as well as energy W spent on neutron production as a function of beam energy**

Neutron production at high energies

- **Considerable increase in average multiplicity of prompt-fission neutrons in actinides as a function of energy of neutrons**
- **Prompt-fission neutron average kinetic energy (Watt spectrum) grows as well**





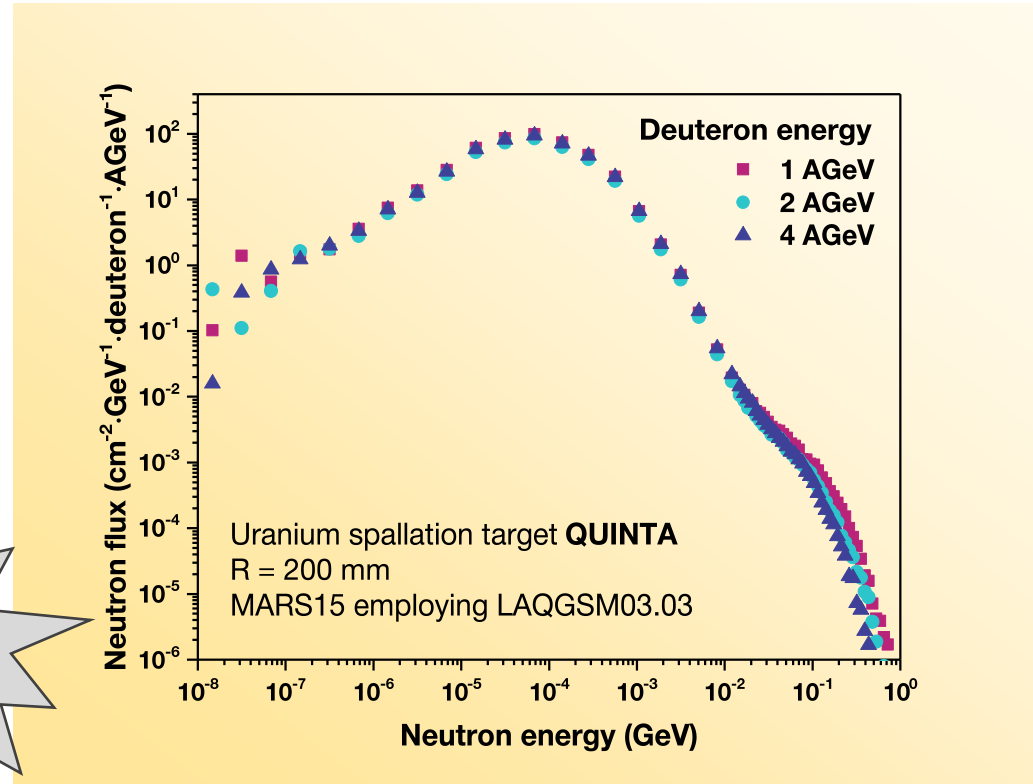
Data adopted from A. Krasa+, NIM A **615** (2010) 70

- **Neutron cost decreases as a function of beam energy above 1 GeV in lead target of limited dimensions**

Neutron production at high energies

- **Increase in**
  - Mean neutron energy f(E_{beam})
  - Neutron multiplicity due to (n,xn) reactions
  - Prompt neutron multiplicity f(E_n)
  - Average kinetic energy f(E_n)

- **Decrease in**
  - Neutron cost above 1 GeV



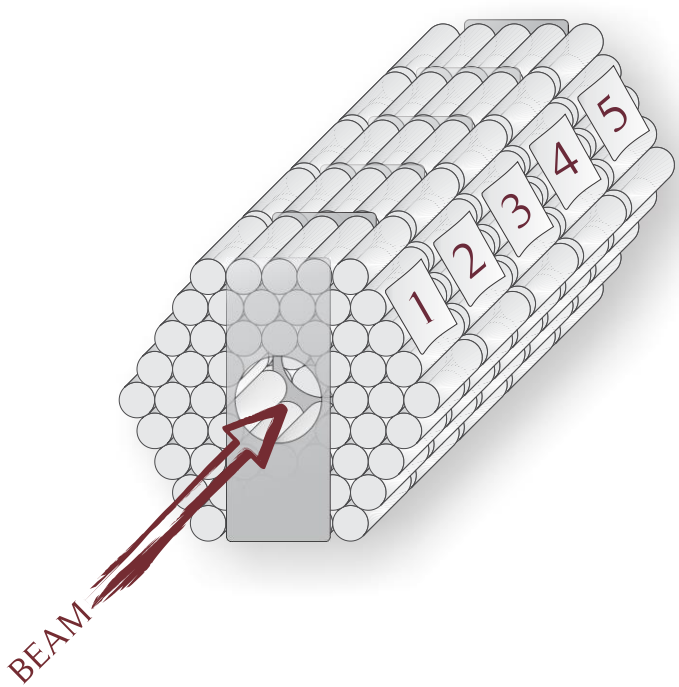
- Some experimental phenomena are not yet implemented in the physics models of radiation transport codes.
- These effects need to be scrutinized at the quasi-infinite spallation targets with minimal neutron leakage

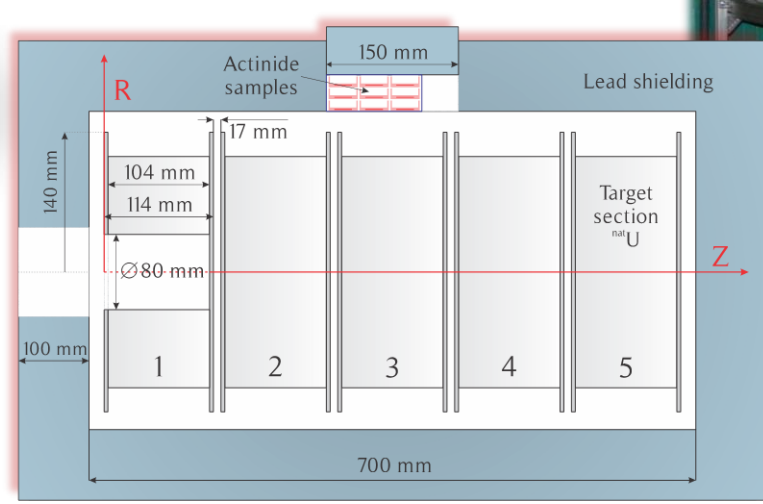
Current research on ADS @ Dubna

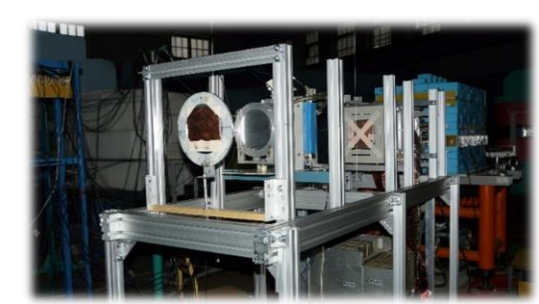
- **Investigation of neutron production in a concept of Relativistic Nuclear Technology for nuclear energy production and transmutation of spent nuclear fuel (SNF) in a maximally hard neutron spectrum**
  - **Deep subcritical core composed of thorium or natural / depleted uranium with SNF elements**
  - **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 up to 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)**

Experimental methods

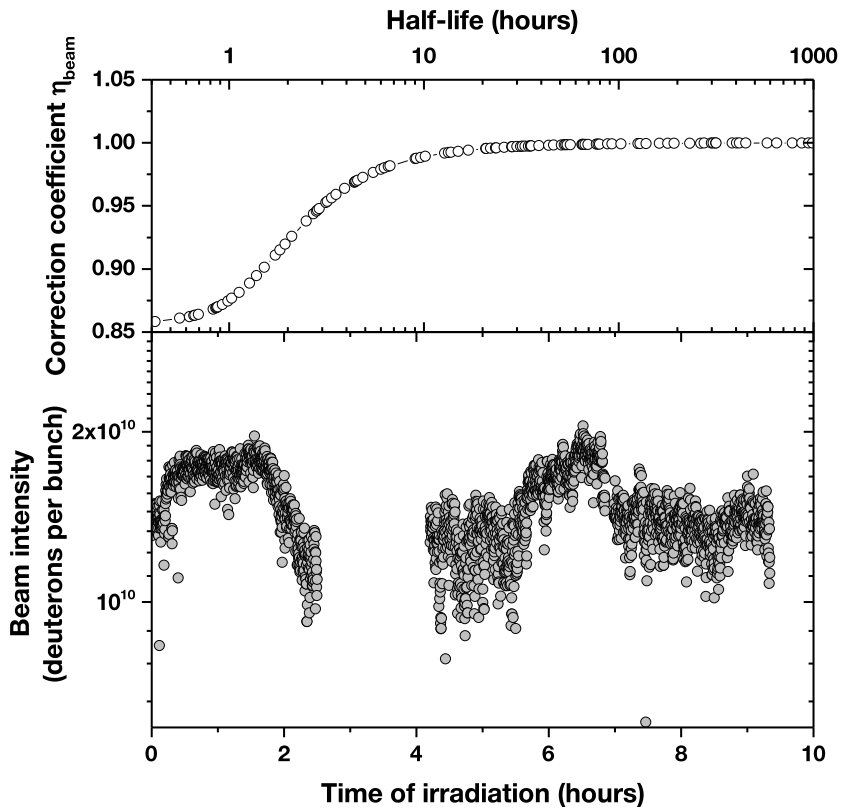
- **Activation measurement technique**
- **Gamma-ray spectroscopy with the use of both planar and coaxial, P- and N-type HPGe detectors Canberra and ORTEC of up to 35% relative efficiency**
- **Calibrated with standard point gamma-ray sources from 5 keV to 3 MeV**
- **Efficiency compared with MC simulation, which is used for performing corrections for volume emitters**

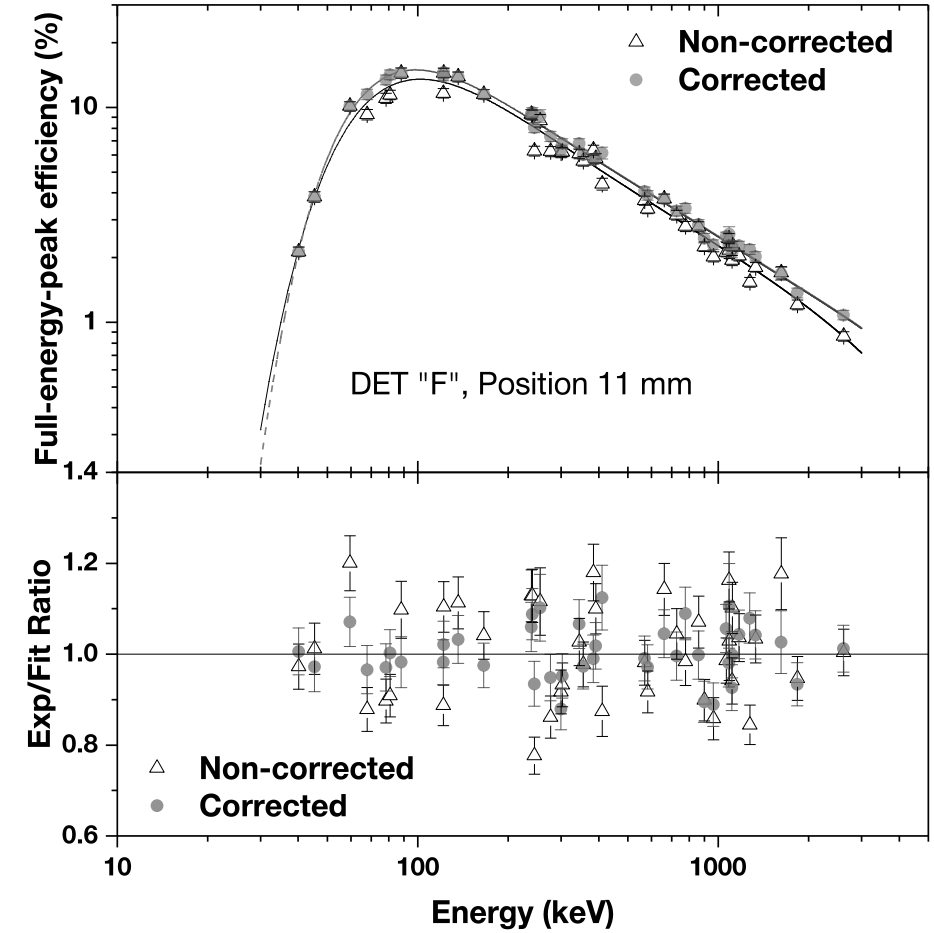
Activation measurement

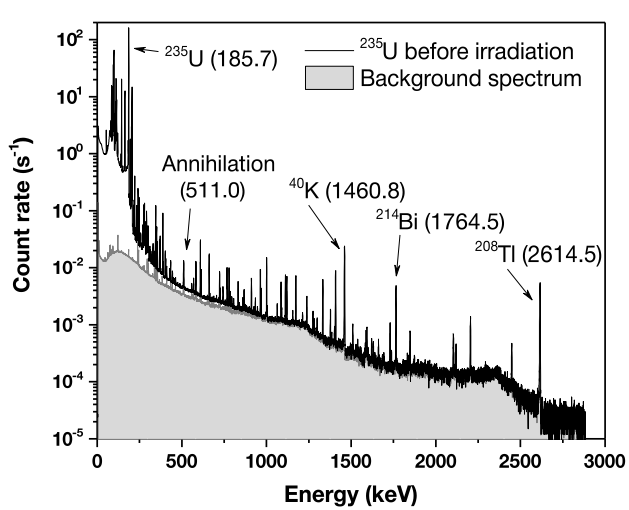
- Identification of products:
  - Half-life (min. 6 measurements)
  - Energy and intensity of gamma line
  - Reaction rates R calculated from measured activity
  - The following correction factors considered:

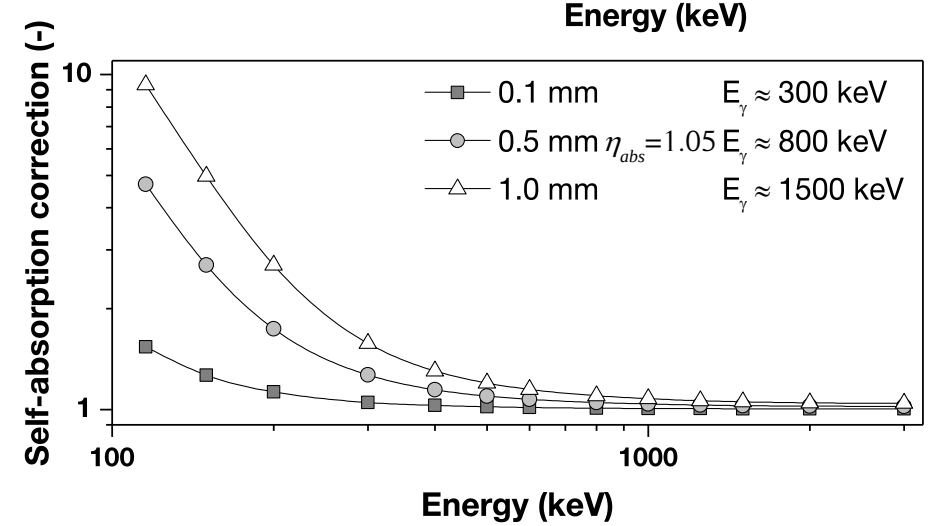
    background radiation, decay during irradiation, cooling and measurement, dead time, nonlinearity, detector efficiency, beam instability, nonpoint-like source, self-absorption, true coincidence summing

Correction factors

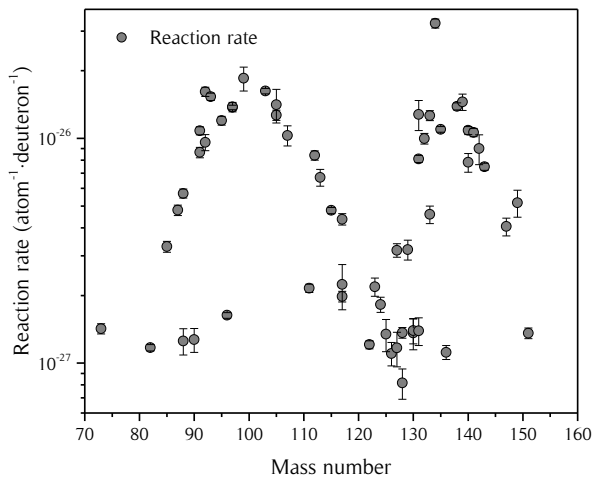


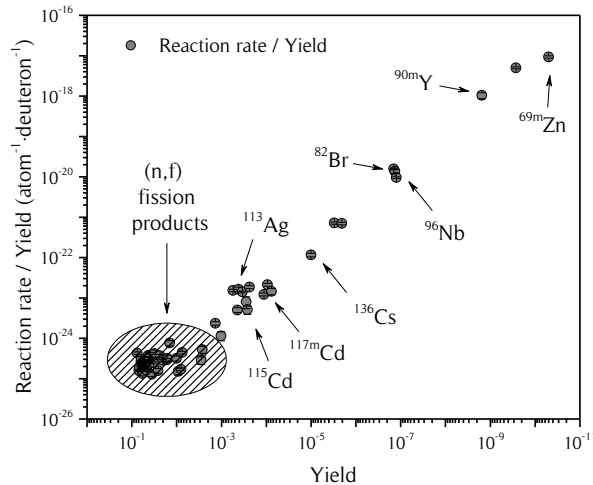






Fission rate determination

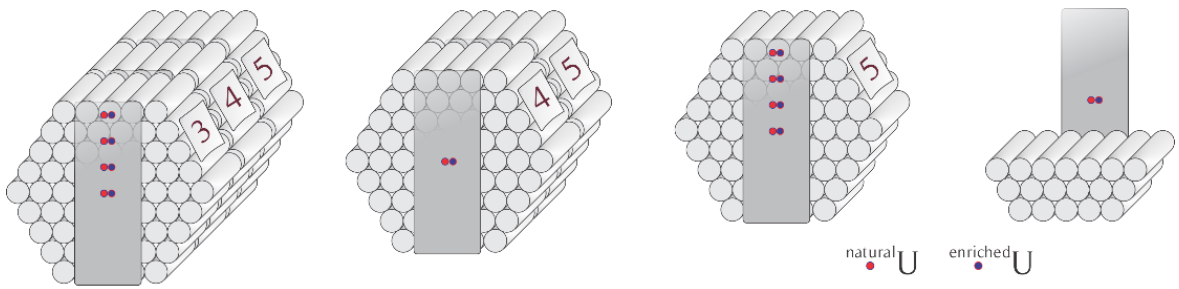


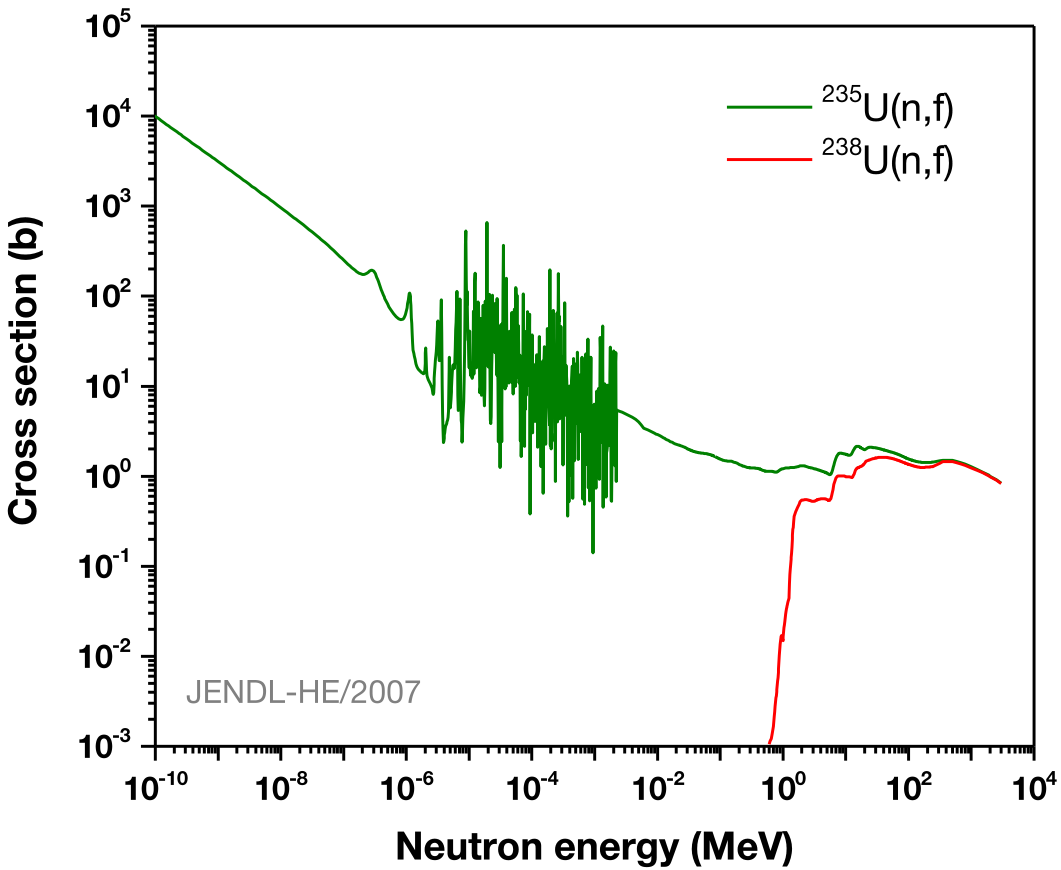


- **In measurements of uranium samples more than 70 fission products detected**
- **Proper selection of 18 fission products with suitable half-life, gamma-ray energy and intensity and cumulative fission yield:**

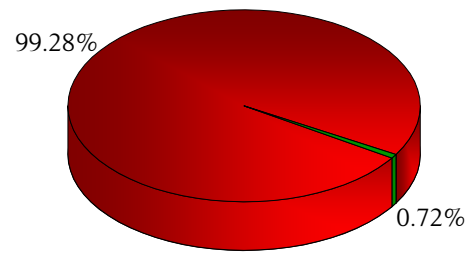
^{87}Kr, ^{88}Kr, ^{91}Sr, ^{92}Sr, ^{93}Y, ^{95}Zr, ^{97}Zr, ^{99}Mo, ^{103}Ru, ^{105}Ru, ^{129}Sb, ^{131}I, ^{133}I, ^{135}I, ^{140}Ba, ^{143}Ce, ^{147}Nd, and ^{149}Nd

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

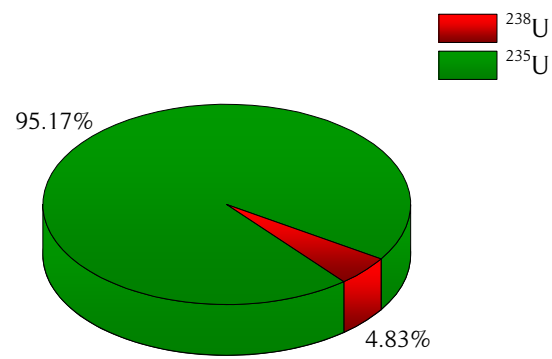




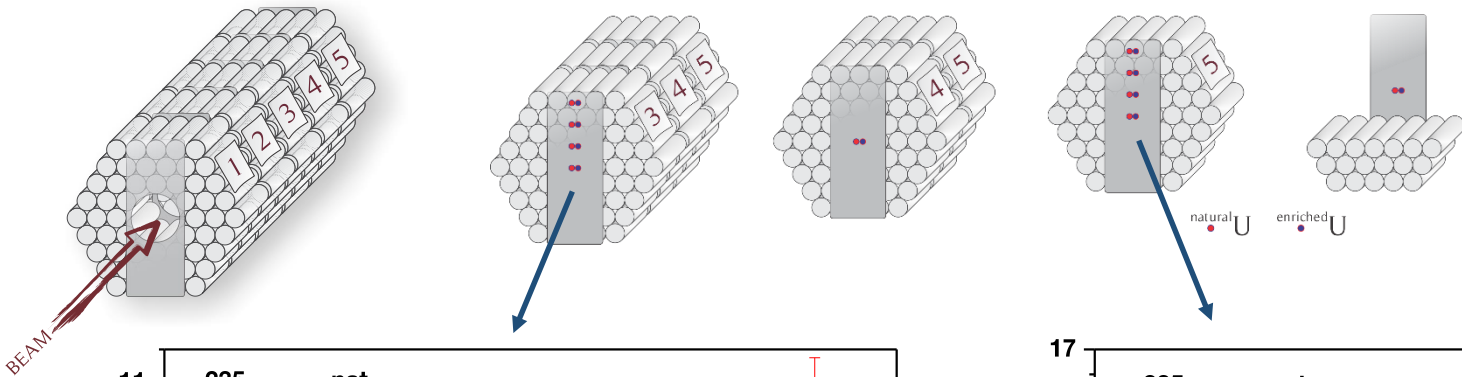
Natural uranium



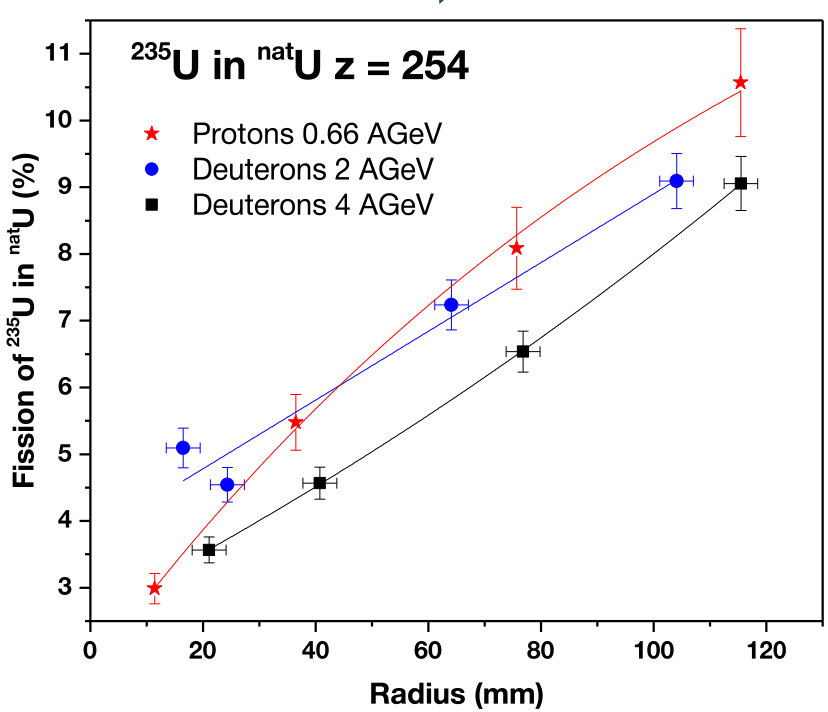
Enriched uranium

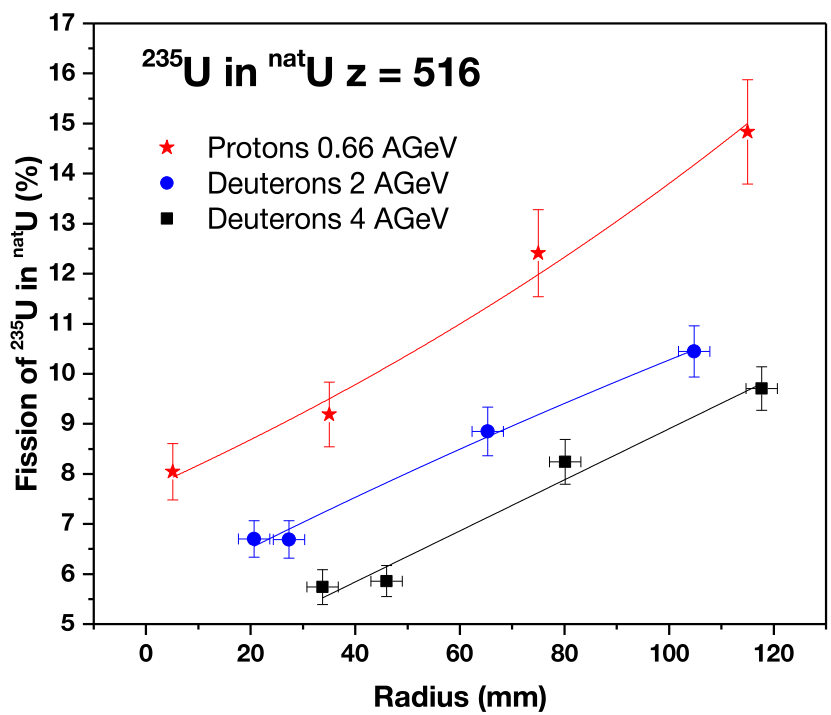


Role of ^{235}U fission in ^{nat}U target



- 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

- **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

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

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

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

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

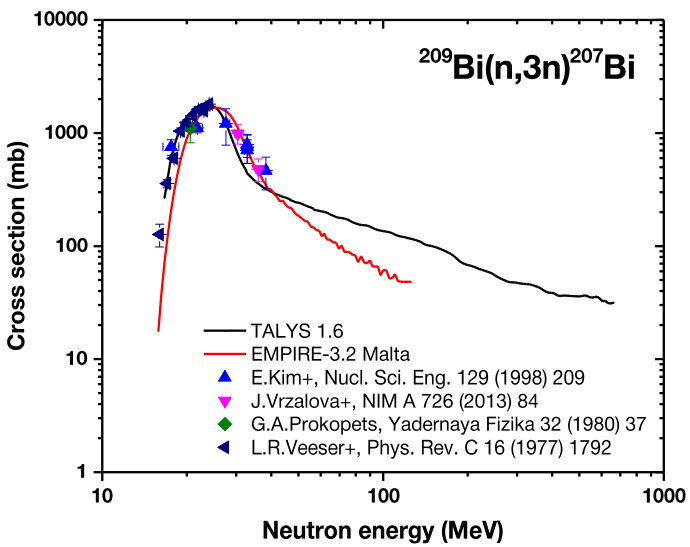
$$\Rightarrow \phi(8) = \ldots$$

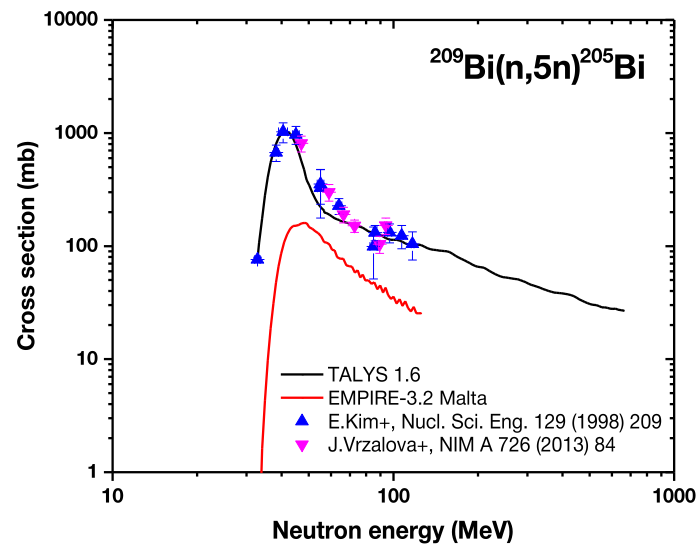
No initial guess flux in this approach!

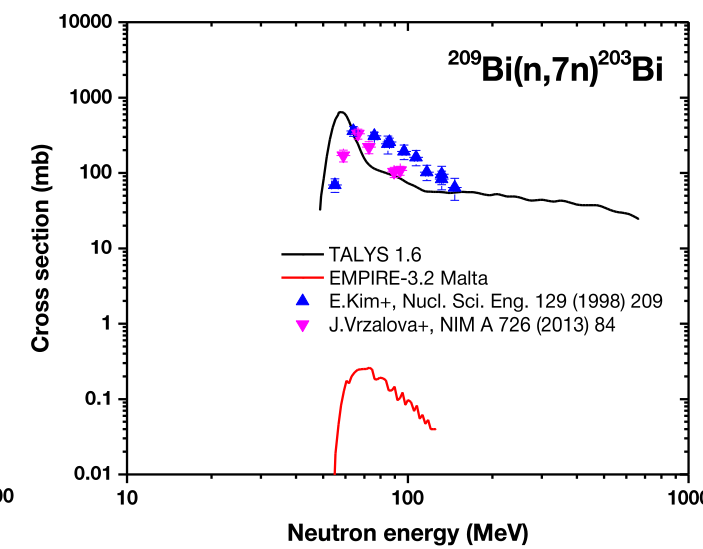
- **Validation of stochastic and deterministic codes MCNPX 2.7 and TALYS 1.6**

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**

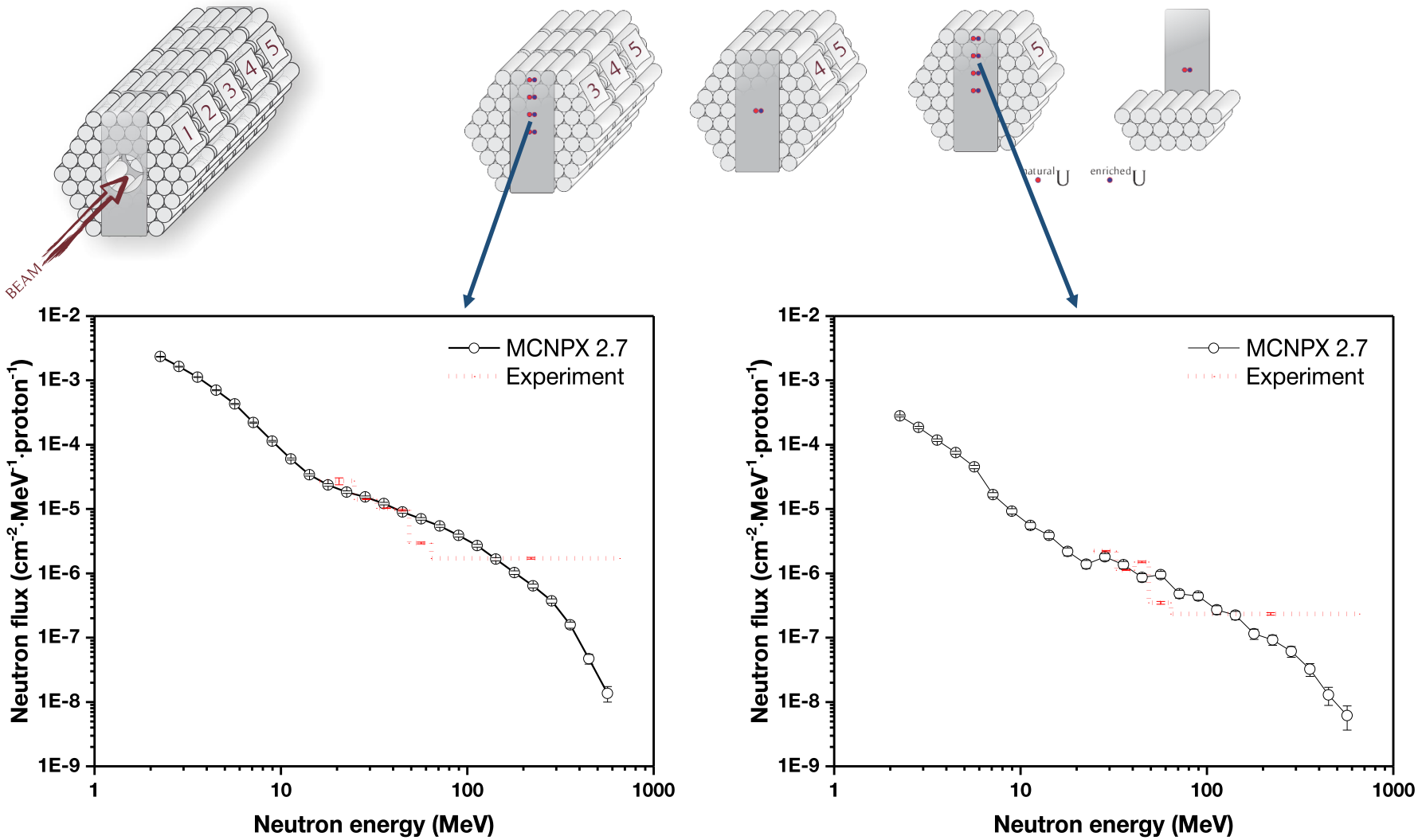






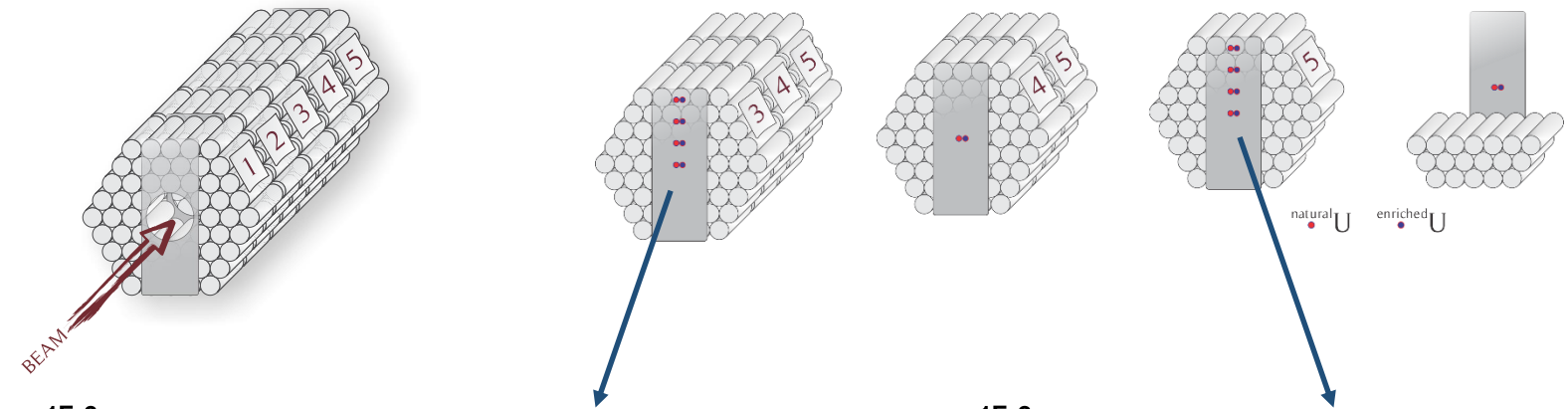
- **EMPIRE is not suitable for our purposes**

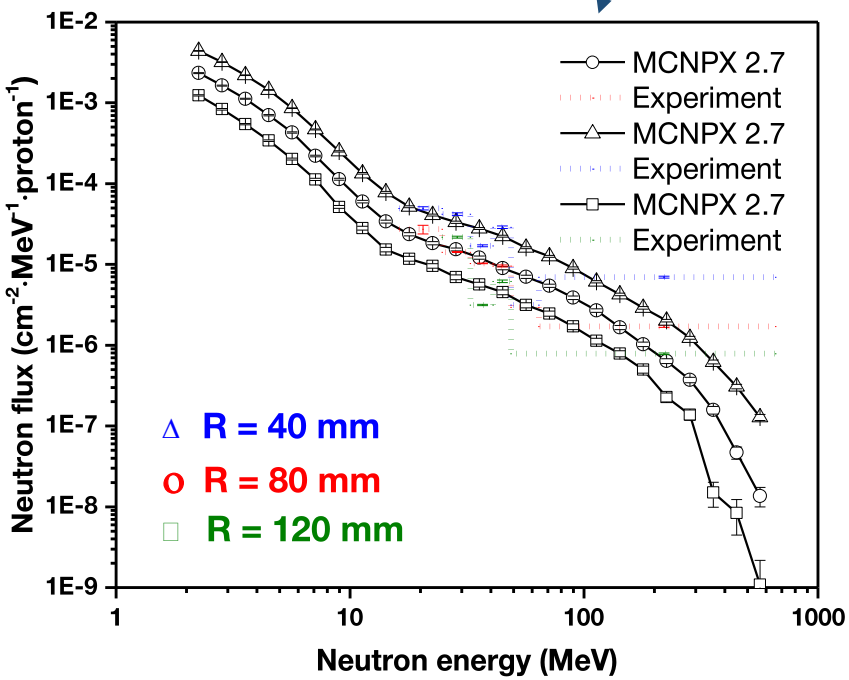
Spallation neutron spectra

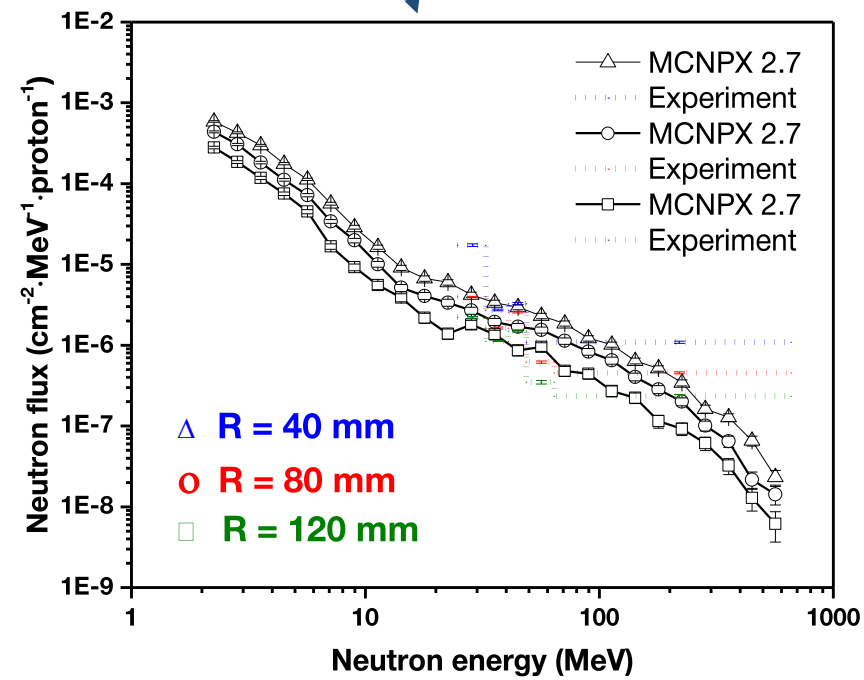


- **MCNPX 2.7 and experiment with TALYS 1.6 data**

Spallation neutron spectra







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

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

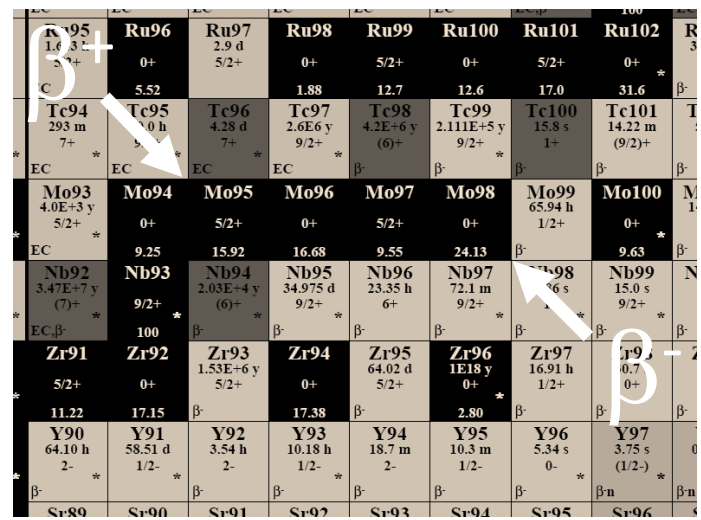
- **using methods of direct kinematics at JINR Dubna (this experiment) to obtain new experimental data and validate nuclear physics models at the beams of relativistic energies - deuterons up to 3.5 AGeV**

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

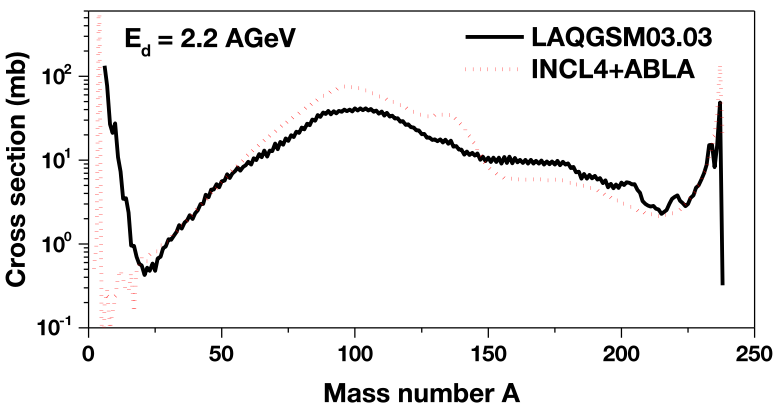
- Cross sections obtained for a large number of neutron-rich nuclei, including some metastable states of produced residues, as well as some neutron-deficit nuclei and products of quasi-elastic reactions
- New experimental data compared to the results of simulations employing physics models available 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

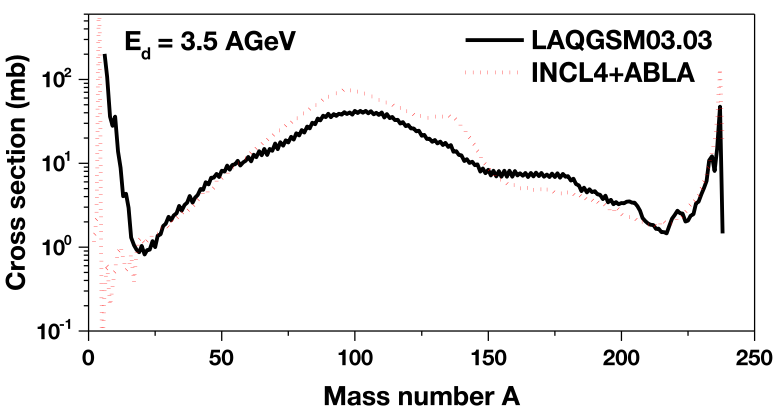


Isotope	^{69m}Zn	^{85m}Kr	^{106m}Ag	^{117m}Cd	^{120m}Sb
$T_{1/2}$	13.76 h	4.48 h	8.28 d	3.36 h	5.76 d

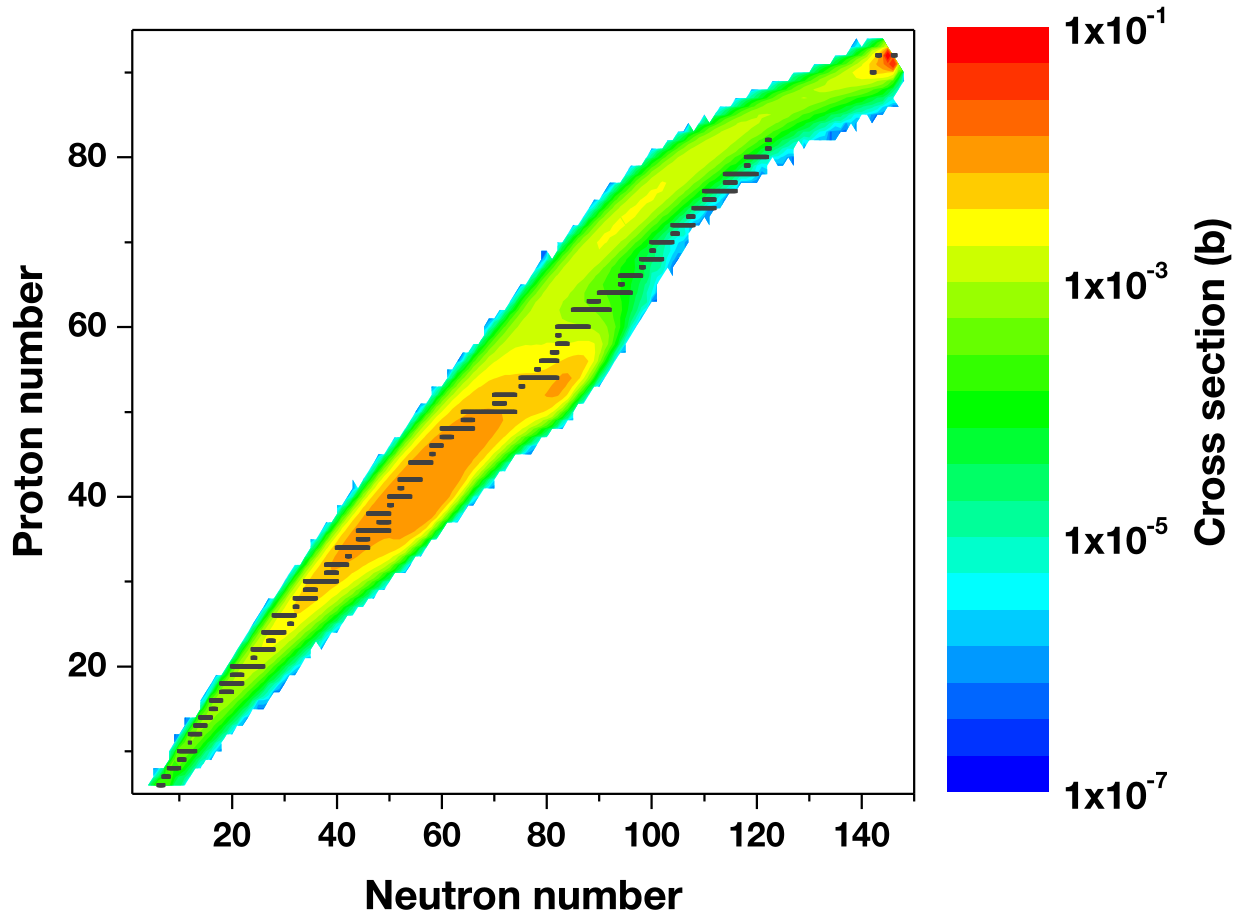
Cross section determination

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



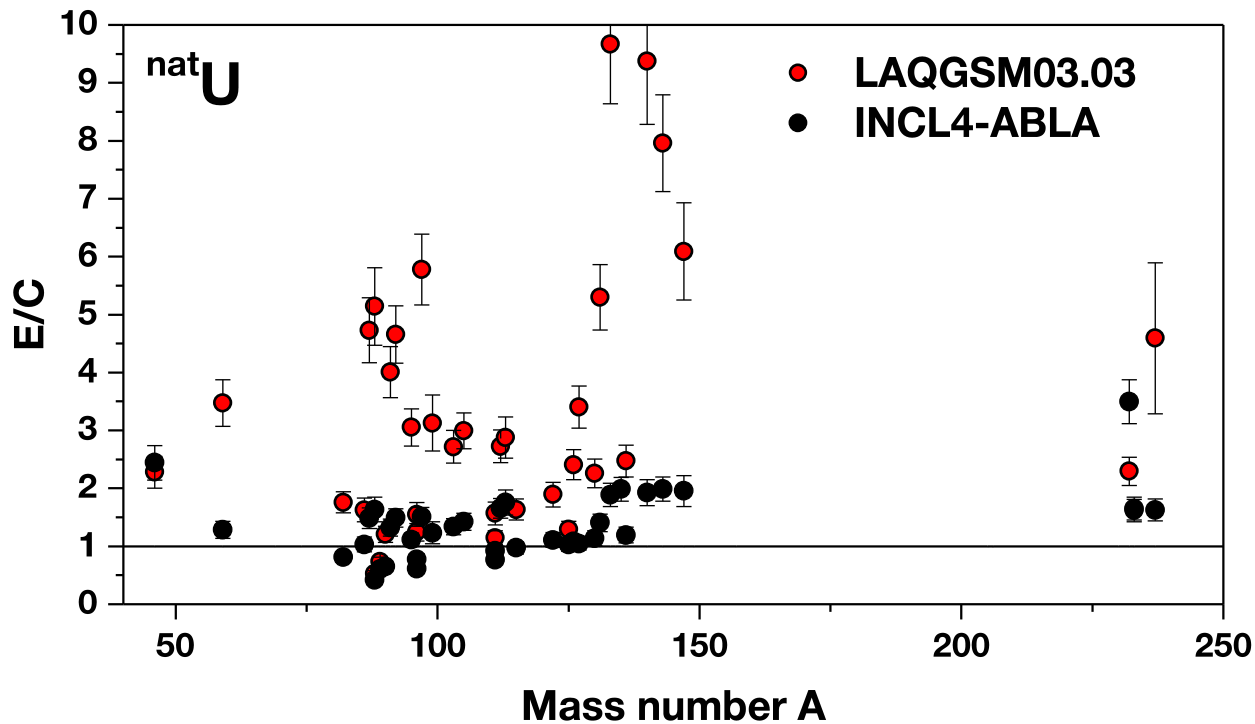


Residual nuclei cross section in ^{nat}U irradiated with 2.2 AGeV deuterons. INCL4/ABLA used.



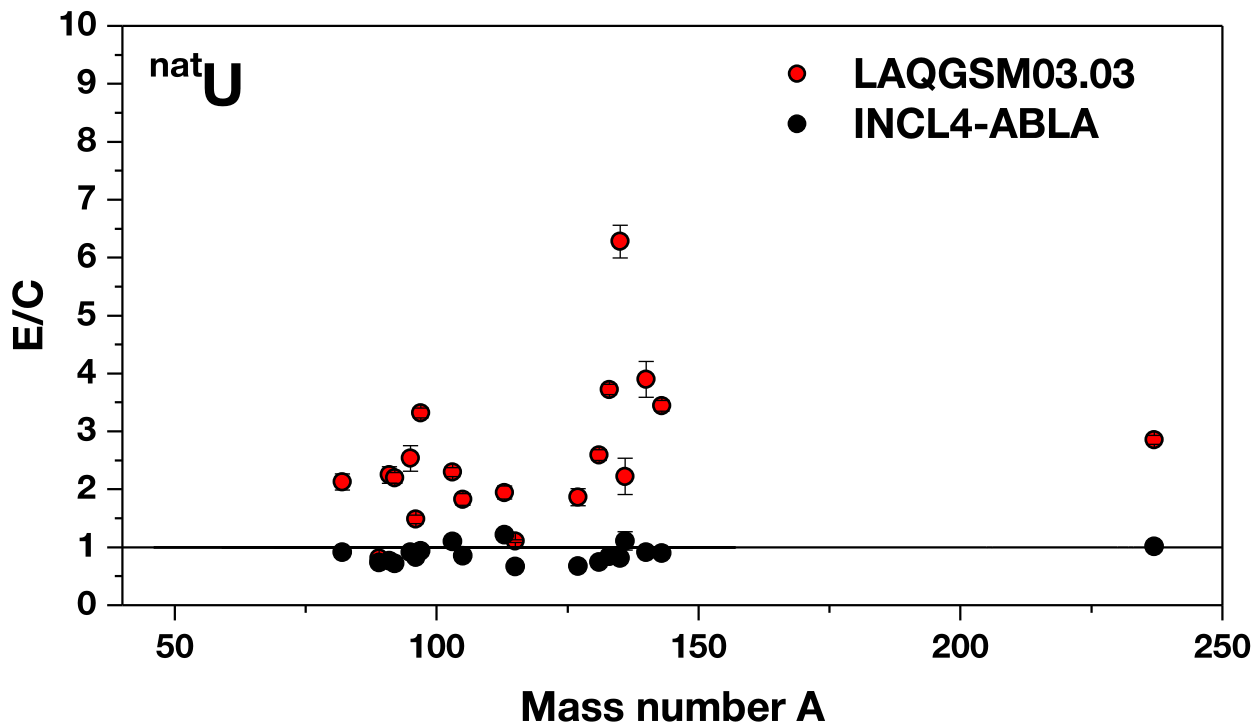
Cross section determination

- **Comparison between experiment and simulation employing INCL4/ABLA and LAQGSM03.03 at the deuteron beam energy 2.2 AGeV**



Cross section determination

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

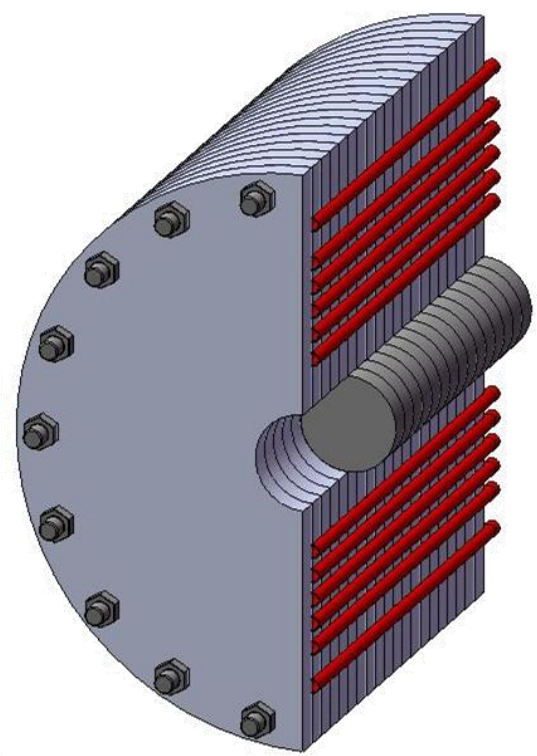


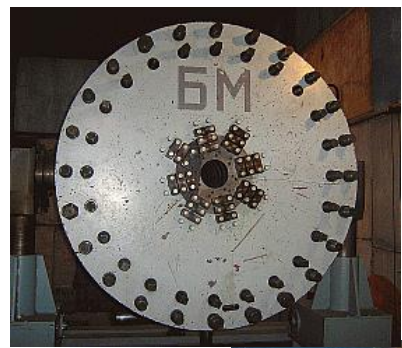
Conclusion

- At relativistic energies of deuteron beams, the INCL/ABLA physics models perform well in production of fission residues in thin U targets
- 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 spallation neutron spectrum as a function of beam energy, which was experimentally confirmed at the massive U target (at energies up to 4 AGeV)

Spallation target BURAN

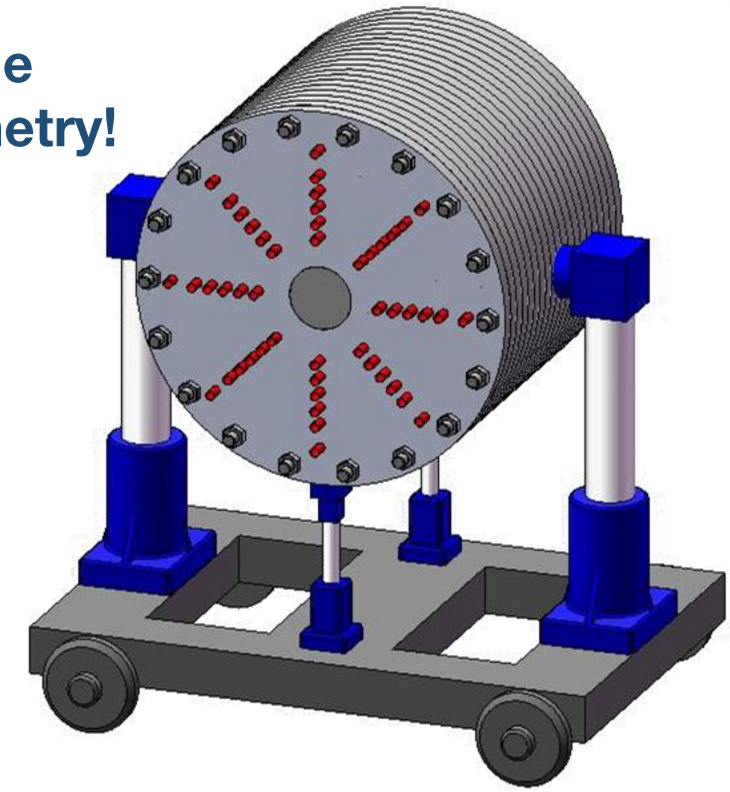
- 20 tons of depleted uranium (central part repl.)
- Simple geometry (diam. 1.2 m, length 1 m)





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.