The LXXI International conference "NUCLEUS – 2021. Nuclear physics and elementary particle physics. Nuclear physics technologies
20 – 25 September Saint Petersburg, Russia

Advances in Modelling of fast neutron induced fission of ^{232}Th

Cristiana Oprea[1,3], Alexandru Mihul[2,3], Ioan Oprea[1]

[1]JINR, FLNP, Dept of Exp Nucl Phys, Dubna, RF

E-mail: istina@nf.jinr.ru

[2]CERN, Genève, CMS, Switzerland

[3]Romanian National Agency for Scientific Research

OUTLINE OF THE PRESENTATION

INTRODUCTION

ELEMENTS OF THEORY. COMPUTER CODES

RESULTS AND DISCUSSION

CONCLUSIONS AND PERSPECTIVES

INTRODUCTION

General motivation of using ^{232}Th

Properties of ^{232}Th – natural abundance 0.9998, spin and parity 0^+, time of life $1.405 \cdot 10^{10}$ y alpha decay
Rare decays: $b^- b^-$, spontaneous fission, cluster decay
Other main isotopes have trace concentration abundannce
Natural Main Isotopes A from 227 to 234
^{232}Th – fertile nucleus for ^{233}U as part Th-U fuel cycle

Isotope ^{233}U -> obtained as:
Fission product in neutron induced fission

Properties of ^{233}U – spin and parity $(5/2)^+$ time of life $16 \cdot 10^4$ y, alpha decay
Rare decays: spontaneous fission and cluster decay

Fission – > a real solution to the global energy challenge of the future

- Hydrocarbon based energy will be finished in a few decades
- Wind energy – still expensive and not effective
- Nuclear energy obtained by fission of ^{5}U is also limited

Thorium fuel cycle

Advantages over a Th - U cycle:

- Large reserves of Th -> may be the most appropriate energy solution
- Superior physical and nuclear properties
- Reduced Pu and actinide production
- Resistance to nuclear weapons proliferation for light water reactors (not for molten salt one)

Disadvantages

- Natural Th not contains fissile nuclei – need to be added $^{233,235}U$, Pu for criticality
- higher burnup for neutrons economy – hard to reach at LWR
- large periods of time for producing ^{233}U from ^{232}Th
- ^{233}U – large time of life – a radioactive isotope in the waste

Robert Hargraves, Ralph Moir, Liquid Fluoride Reactors, American Scientist, July/ 2010

In many countries have already started research programs on Th - U cycle

Goal and objectives

Fission process induced by neutrons up to 10 MeV energy on ^{232}Th was analyzed;

Experimental observables as cross sections, fragments mass distribution, yields of some nuclides of interest and average prompt neutrons multiplicity characterizing ^{233}U fission were theoretically evaluated by using TALYS-1.9 software;

This study represents a research proposal for neutron induced fission investigations and isotopes production at the new neutron source IREN, from FLNP - JINR

Fundamental researches

Fission - investigation of the configuration of fissionable system near scission point. It gives information on: measurements of anisotropy, emitted gamma rays, fission products ground states

Applicative researches

Fission – important for transmutation and nuclear energy projects, new generation of nuclear reactors

Isotopes and Isomers productions for a wide range of applications in medicine, electronics, engineering etc

CODES AND ELEMENTS OF THEORY

Evaluations by Talys

Codes for nuclear reaction mechanisms and nuclear structure calculations
Implemented compound, direct and pre-equilibrium processes
Wide databases of nuclear data - energy levels, density levels, spins, parity, optical potential parameters for many nuclei, and many others

Fission Induced by Neutron

Cross section -> compound nucleus process
Density levels – Constant temperature with Fermi gas model
Mass distribution of fission fragments and yields of isotope production – evaluated in the frame of Brosa model

A.J. Koning, S. Hilaire and M.C. Duijvestijn, TALYS-1.0, Proceedings of the International Conference on Nuclear Data for Science and Technology, April 22-27, 2007, Nice, France, editors O.Bersillon, F.Gunsing, E.Bauge, R.Jacqmin, S.Leray, EDP Sciences, 211 (2008)

Talys codes and elements of theory. I

Hauser – Feshbach Approach. XS

$$\sigma_{\alpha\beta} = \pi \lambda\!\!\!^{-}{}_{\alpha}^{2} \frac{T_\alpha T_\beta}{\sum_c T_c}$$

→ **Historically first HF expression**

$$\sigma_{\alpha\beta} = \pi \lambda\!\!\!^{-}{}_{\alpha}^{2} \frac{T_\alpha T_\beta}{\sum_c T_c} W_{\alpha\beta}$$

$W_{\alpha\beta} =$ **Widths Fluctuation Correction Factor (WFC)**

W. Hauser, H. Feshbach, Phys Rev 87 2 366 (1952)

WFC

- Indicates a correlation between the ingoing channel (incident) and outgoing channels
- At low energies (<1 MeV) WFC=1 - no correlation between *in* and *out* channels
- For neutron induced reactions with emission of charged particles this factor is slowly decreasing with energy for fast neutrons
- It is calculated by complicate procedures (ex Moldauer expression)

Talys codes and elements of theory. II

Fission XS for a given fission fragment (FF) mass

$$\sigma(A_{FF}) = \sum_{Z_{FS}, A_{FS}, E_x} \sigma_F(Z_{FS}, A_{FS}, E_x) Y(A_{FF}; Z_{FS}, A_{FS}, E_x)$$

A_{FF} = FF mass; $s_F(Z_{FS}, A_{FS}, E_x)$ = cross section of fissionable system (FS)
Y (A_{FF} ;Z_{FS}, A_{FS}, E_x) = relative yield of FF with mass A_{FF} coming from a FS with mass A_{FS} and charge Z_{FS}
Z_{FS}, A_{FS} = charge and mass of FS; E_x = excitation energy

XS Production of FF with given mass (A_{FF}) and charge (Z_{FF})

$$\sigma_{prod}(Z_{FF}, A_{FF}) = \sum_{Z_{FS}, A_{FS}, E_x} \sigma_F(Z_{FS}, A_{FS}, E_x) Y(A_{FF}; Z_{FS}, A_{FS}, E_x) Y(Z_{FF}; A_{FF}, Z_{FS}, A_{FS}, E_x)$$

Y (Z_{FF} ;A_{FF} ,Z_{FS}, A_{FS}, E_x) = relative yield of FF with charge Z_{FF} and mass A_{FF} coming from a FS with mass A_{FS} and charge Z_{FS}

is weighted by the product of yields with given mass and fixed charge

Talys codes and elements of theory. III

FF mass distribution

$$Y(A_{FF};Z_{FS},A_{FS},E_x)=\sum_{FM=SL,STI,STII} W_{FM}(Z_{FS},A_{FS},E_x)Y_{FM}(A_{FF};Z_{FS},A_{FS},E_x)$$

W_{FM} (Z_{FS}, A_{FS}, E_x) = weight of fission mode (FM);
Y_{FM} $(A_{FF}$;$Z_{FS}, A_{FS}, E_x)$ = mass distribution;
FM = SL = superlong; STI, II = standard I, II

going over the possible fission modes (FM) and the yield corresponding to a given mass A_{FF} and is pondered by the fission mode weight W_{FM}

FM weight

$$W_{CFM}(Z_{FS},A_{FS},E_X)=\frac{T^B_{f,CFM}}{T^B_{SL,CFM}+T^B_{STI,CFM}+T^B_{STII,CFM}}$$

ratio between the transmission coefficient corresponding to a fission mode and the sum of transmission coefficients on all modes

CFM = SL, STI, STII; $T_{f,\,CFM}$ = transmission coefficient (Hill – Wheeler);
B = second barrier

M. C. Duijvestijn, A. J. Koning, and F. -J. Hambsch, Phys Rev C **64**, 014607 (2001)
O. Bersillon, F. Gunsing, E. Bauge, R. Jacqmin, and S. Leray, EDP Sciences 211-214 (2008)

U. Brosa, S. Grossmann, A. Muller, Phys Rep **197**, 167-262 (1990)

Isomer Ratios

Isomer Ratios (IR) – extract info on spin distribution, dependence of level density on angular momentum, probabilities of radiation transitions between the levels

$$R = \frac{Y_m}{Y_g} = \frac{\int_{E_{th}}^{E_m} N_0 \phi(E_n) \sigma_{nf}^{m}(E_n) dE_n}{\int_{E_{th}}^{E_m} N_0 \phi(E_n) \sigma_{nf}^{g}(E_n) dE_n}$$

$\phi(E_n) =$ Incident neutrons flux ~1/E_n

$N_0 =$ Number of target nuclei

$Y_m, Y_g =$ Yields of isomer + ground states

$E_{th}, E_m =$ Threshold maximal energy

IR in fission

Yields of isomer and ground states are obtained using statistical approach proposed by Huizenga

Yields are proportional with the spin distribution of isomer and ground states

Spin Distribution

$$P(J) \sim (2J+1) Exp \frac{-J(J+1)}{2(\sigma+\lambda)^2}$$

J = spin

σ, λ = parameters

Yields for IR – calculated by own computer code using Huizenga approach

J. R. Huizenga, R. Vandenbosh, Phys. Rev. **120** (1960) 1305

J. R. Huizenga, R. Vandenbosh, Phys. Rev. **120** (1960) 1313

RESULTS. Isotopes Production. Talys input data I

Fission calculations
n+^{232}Th (incident channel) – double humped potential barrier was considered

First barrier
Height: 5.8 MeV; Width: 0.9 MeV
Type of axiality: axial symmetry

Second barrier
Height: 6.318 MeV; Width: 0.6 MeV
Type of axiality: left - right asymmetry

Fission model – experimental fission barrier chosen

Fission model yields – Brosa model

Level density model – Constant temperature Fermi gas model

Isotopes Production. XS. Talys input data II

Optical model parameters – n+^{232}Th incident channel

- For nuclear reaction calculations, for incident and emergent channels are used Wood – Saxon potential local parameters extrcted from experimental data

WS P components ->Volume, Surface, Spin-orbit with Real and Imaginary part

Volume	U[MeV]	r[fm]	a[fm]
Real	47.6	1.245	0.644
Imaginary	0.09	1.248	0.594
Surface			
Real	0	0	0
Imaginary	2.46	1.208	0.614
Spin-Orbit			
Real	6.470	1.080	0.570
Imaginary	0	0	0

In the evaluation

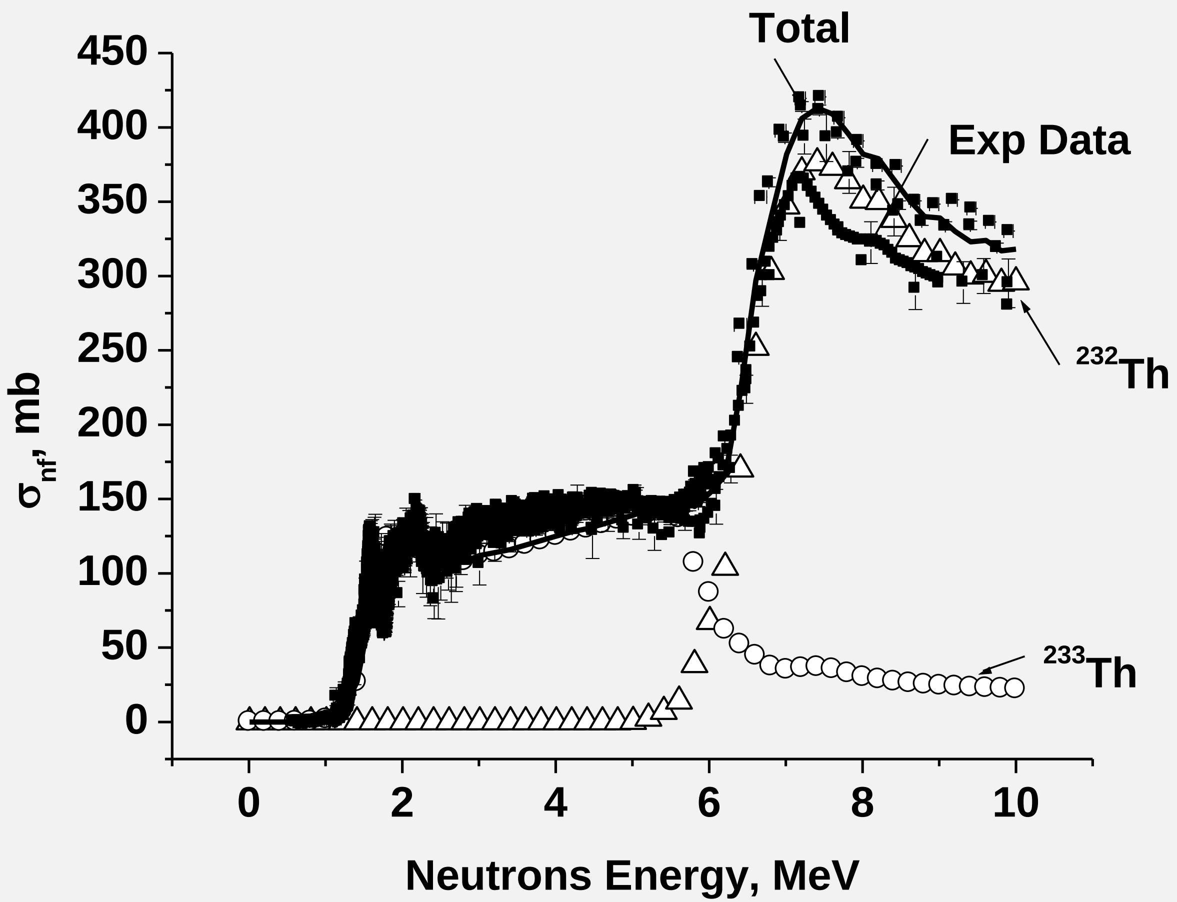
30 discrete levels for target nucleus

5 to 10 discrete levels for residual nucleus

5 excited rotation levels

All parameters can be varied as is necessary

Total Fission XS - ^{232}Th(n,f) – Theory + Experiment



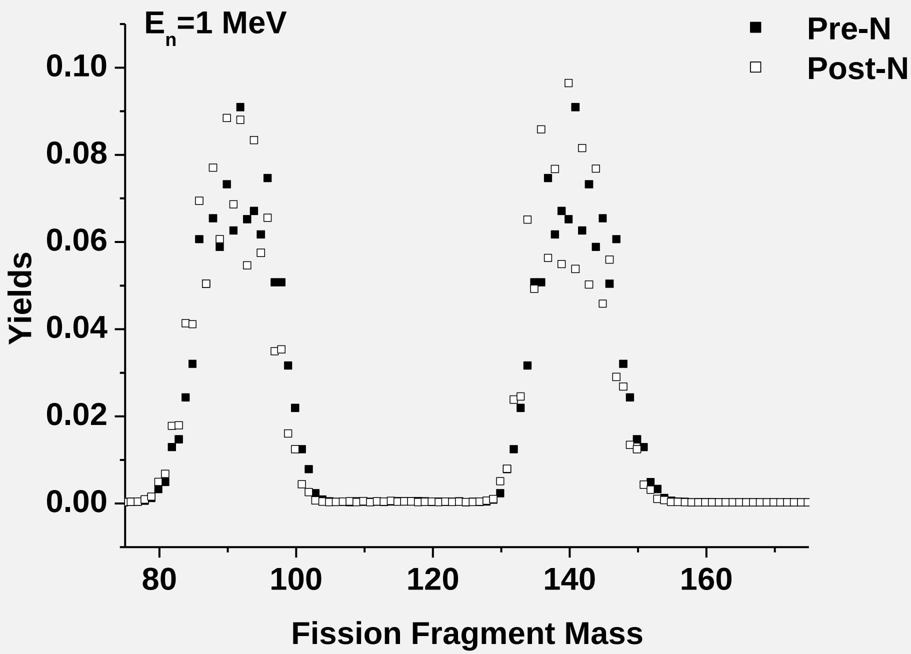
Experimental and Theoretical Data are compared in an energy interval from 0.01 MeV up to 10 MeV

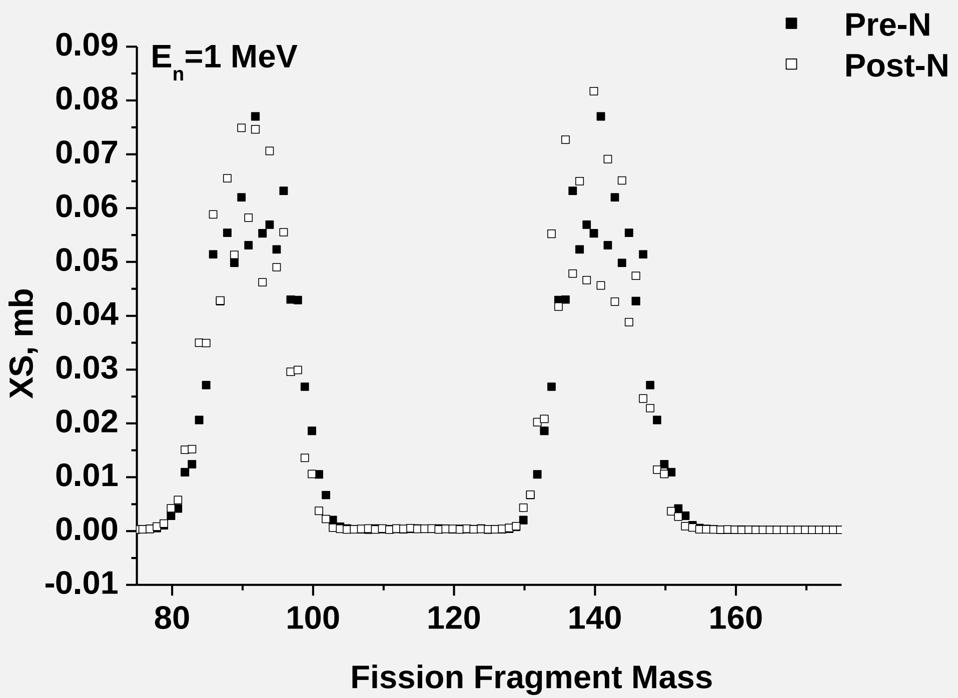
Total Fission XS – Contribution from $^{232, 233}$Th -> Fast neutrons fission well described

Experimental Data – EXFOR

Larson, Nuclear Science and Engineering 135:2 141-149 (2000)

Fission Fragment (FF) Mass Distribution (MD) – Yields + XS - I





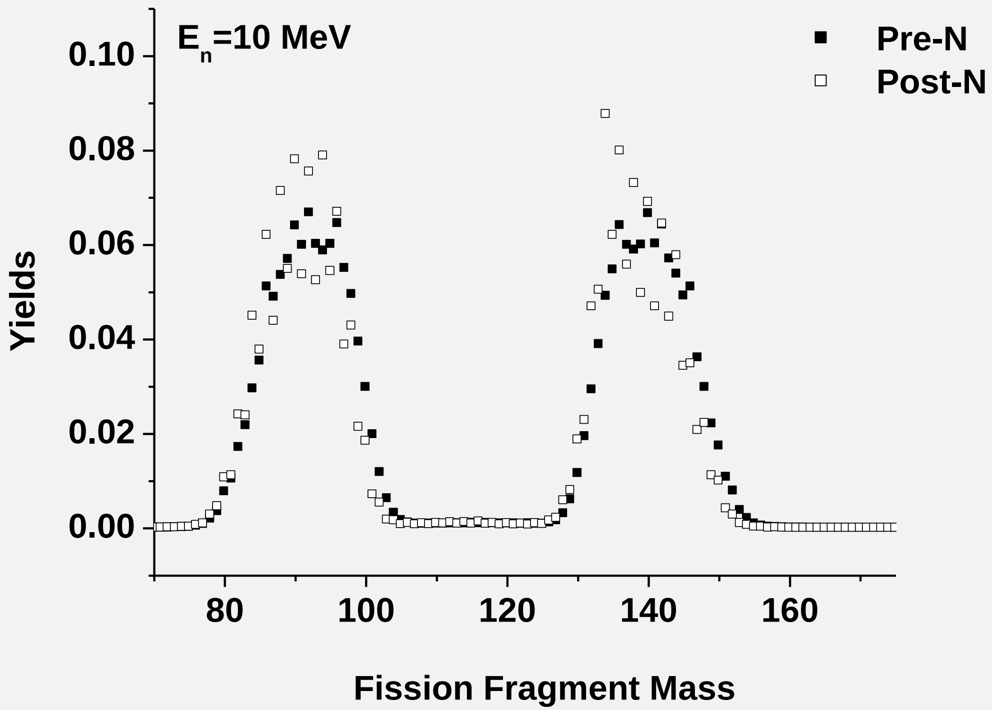
E_n = 1 MeV

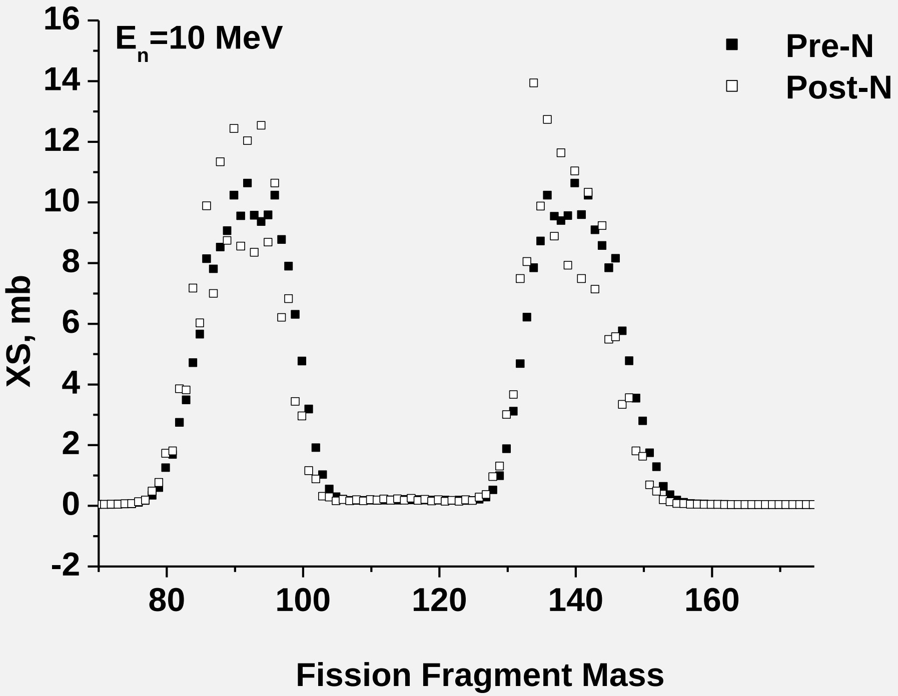
Yields an XS of FF-MD are evaluated using Brosa model

Yields and XS FF MD Dependence Before (Pre) and after Neutrons Emission (Post)

FF-MD is not very sensible to incident neutron energy up to 1 MeV

Fission Fragment (FF) Mass Distribution (MD)–Yields + XS - II





E_n = 10 MeV – Incident Neutrons Energy

Yields an XS of FF-MD are evaluated using Brosa model

Yields and XS FF MD Dependence Before (Pre) and after Neutrons Emission (Post)

XS is increasing with incident neutrons energy

Yields are not so sensitive to incident neutrons

FF-MD is slowly enlarging on FF mass

Prompt neutrons

AVERAGE PROMPT NEUTRONS MULTIPLICITY (APNM)

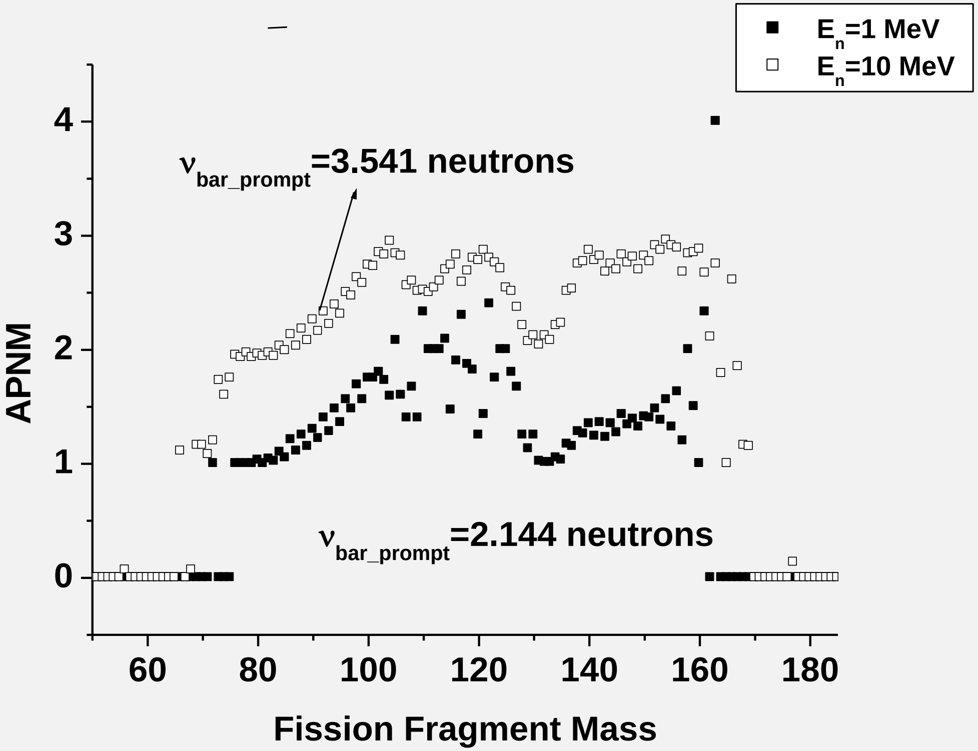
Number of Emitted Neutrons as Function of FF Mass is increasing with neutrons energy

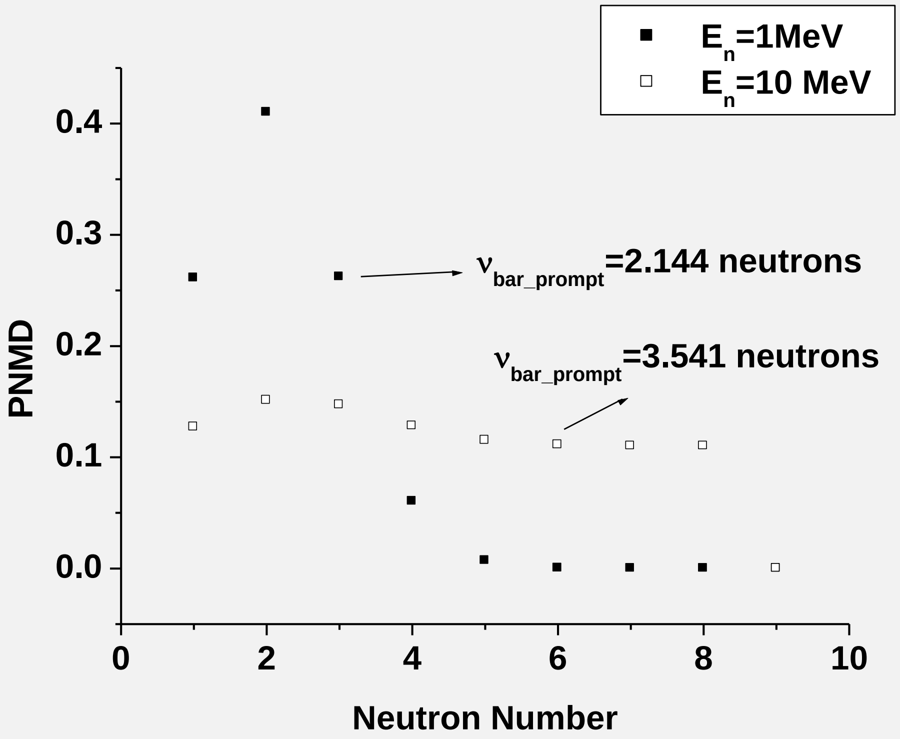
nu_{bar_pr} is also increasing

PROMPT NEUTRONS MULTIPLICITY DISTRIBUTION (PNMD)

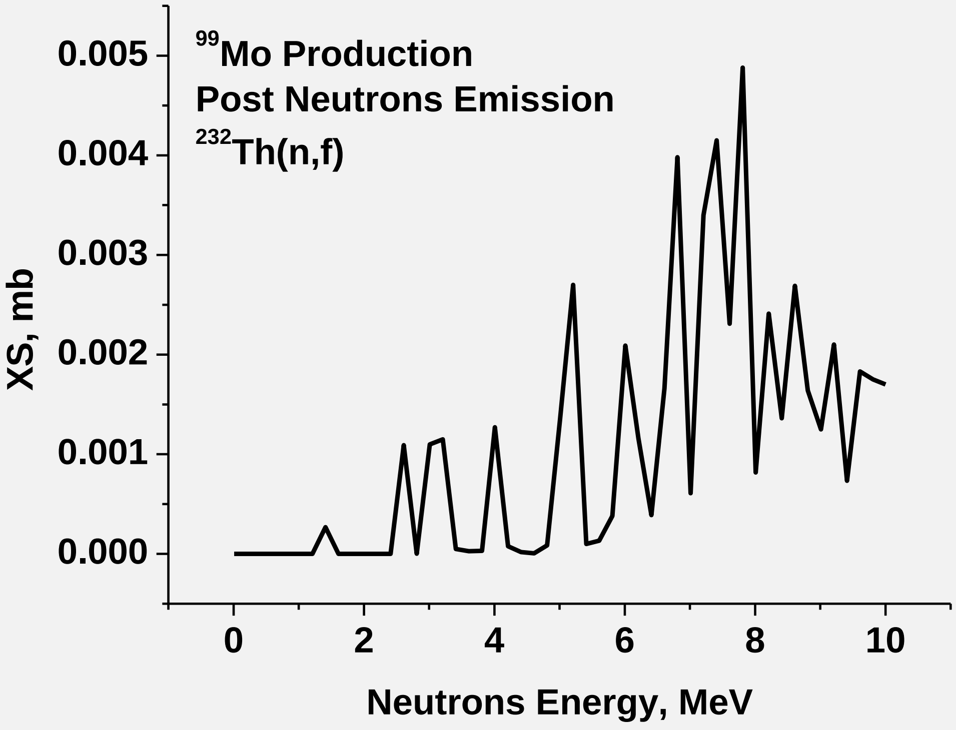
Increasing with neutrons incident energy but limited by excitation energy

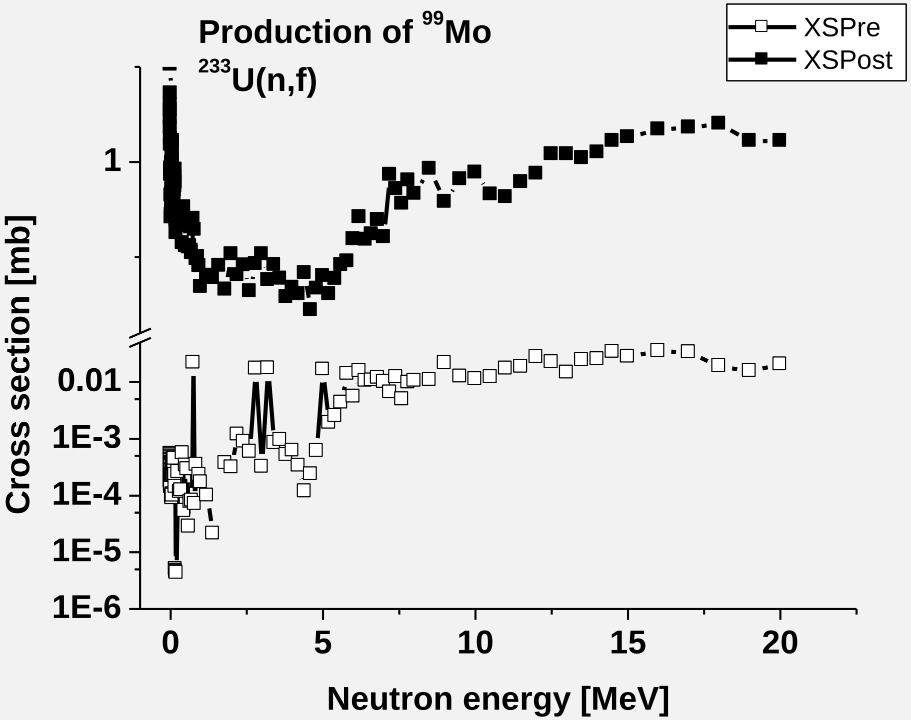
Prompt neutrons fission observables evaluated up to 10 MeV





Isotopes Production. ^{99}Mo - XS

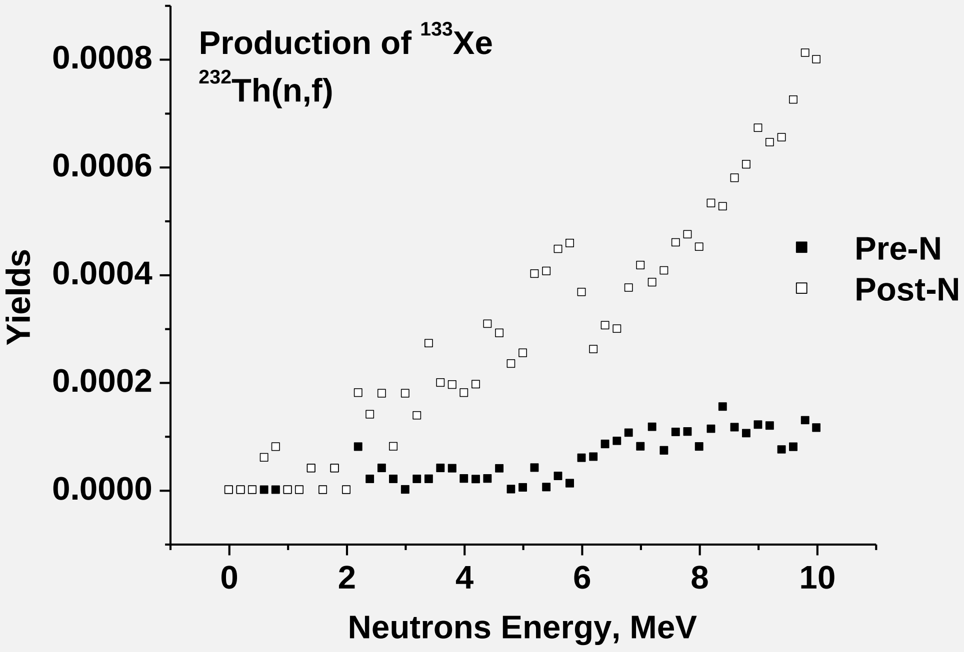


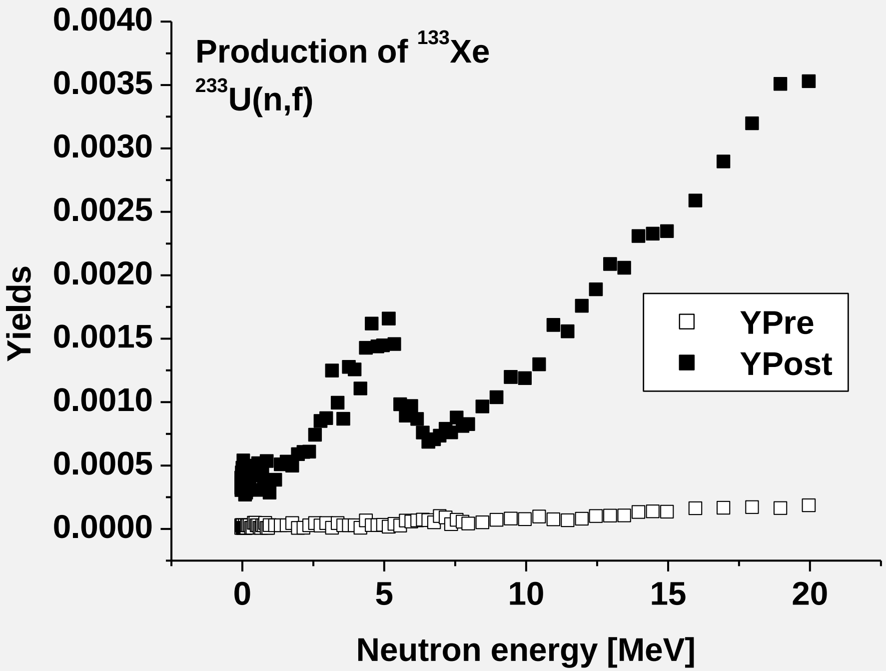


XS Production of ^{99}Mo in Fast Neutrons Induced Fission on ^{232}Th and ^{233}U
^{99}Mo isotope – high interest in oncology

^{99}Mo is produced in higher amount in neutron induced fission on ^{233}U
Yields of ^{99}Mo are not represented -> very low values
XS oscillations of ^{99}Mo are coming from Talys precision of calculation

Isotopes Production. ^{133}Xe - Yields





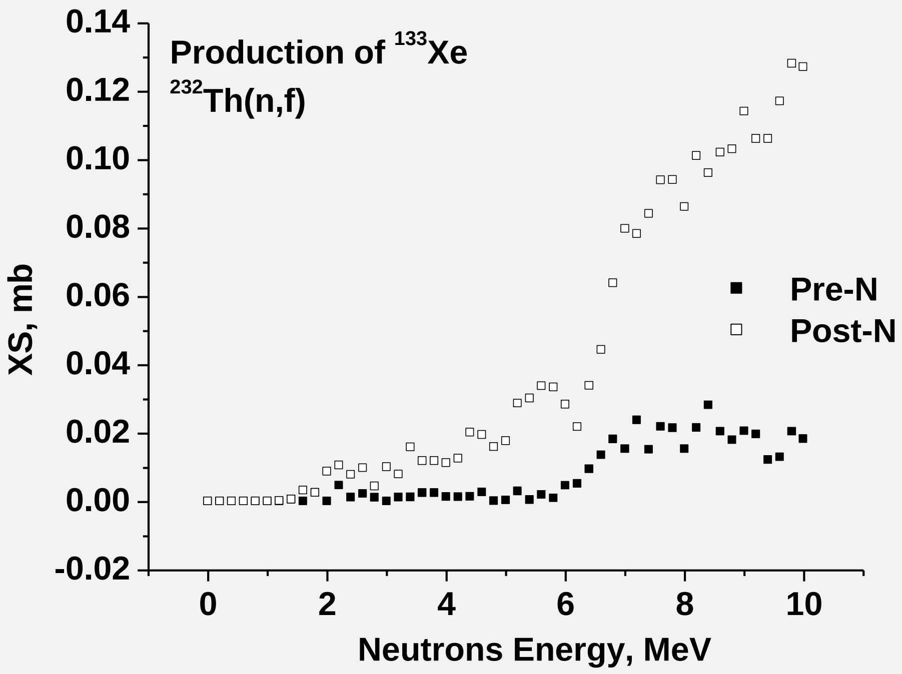
Relative yields of ^{133}Xe in Fast Neutrons Induced Fission on ^{232}Th and ^{233}U for Pre and Post Neutrons Emission

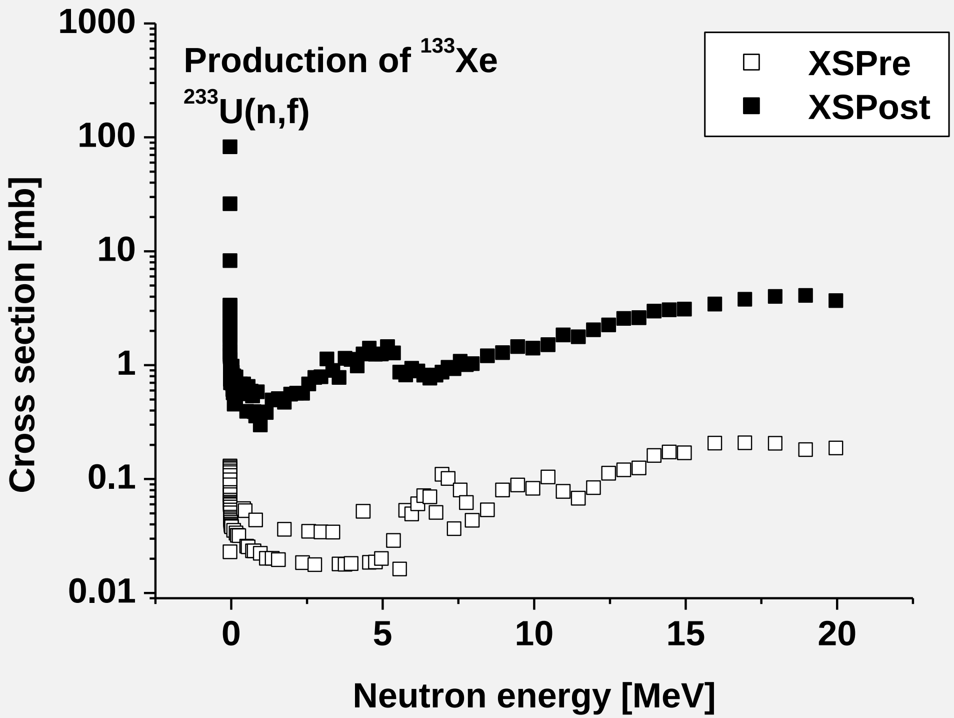
Talys Evaluations

^{133}Xe – of interest in medicine

- with a standard input these yields are not obtained because their values are lower than default Talys minimal value for XS and yields
- it is necessary to increase the precision of calculation in order to obtain evaluation which by default are neglected (by Talys)
- it is opening the possibility to investigate and predict isomer ratios obtained in fission

Isotopes Production of ^{133}Xe. XS





XS Production of ^{133}Xe - as function of incident energy

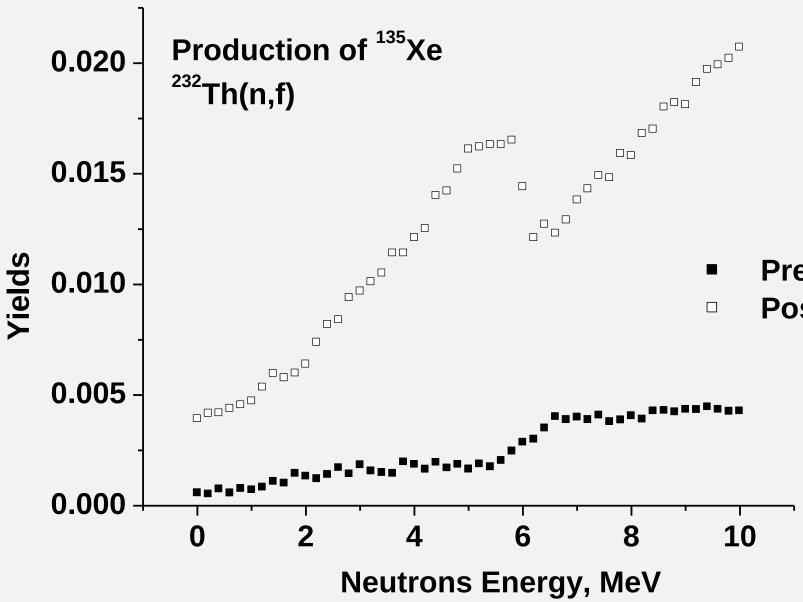
^{133}Xe – of interest in medicine

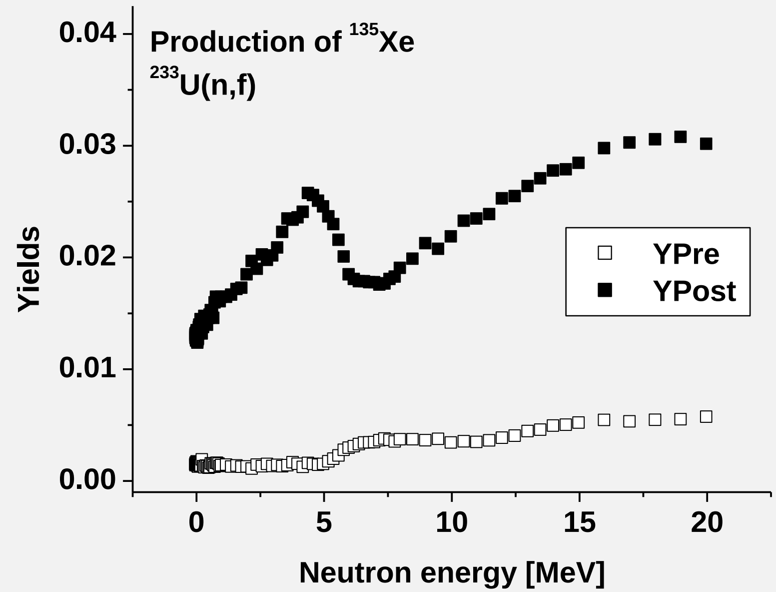
Talys calculation with increased precision

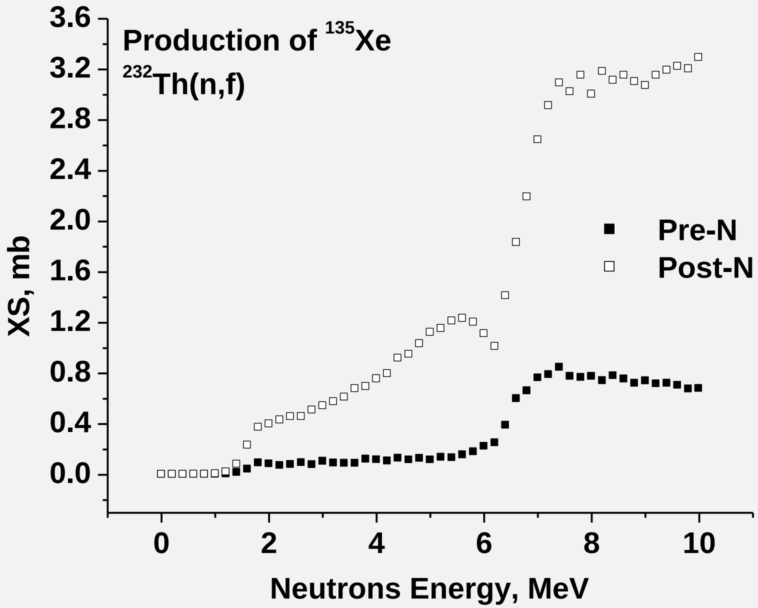
•XS for ^{133}Xe Production larger in the case of ^{233}U neutrons induced fission

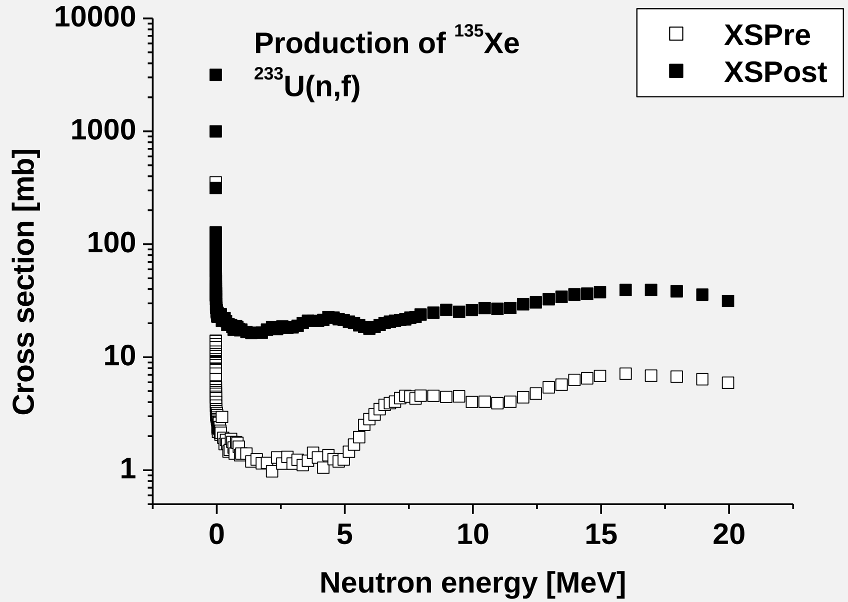
Isotopes Production. ^{135}Xe. Yields and XS

^{135}Xe – neutron absorber -> disturb fission chain reaction of reactor









Isomer Ratios. ^{133}Xe

For usual reactions like (n,p), (n,α), (n,γ) Talys calculates isomer and ground state XS production
For fission with a default run isomer and ground XS are not obtained

$^{133m,g}Xe$ Properties: spin, parity, time of life

Elem.	Ground (g)		Isomer (m)	
	J^{Π}	τ_g	J^{Π}	τ_m
^{133}Xe	$(3/2)^+$	5.24 d	$(11/2)^-$	2.19 d

Result – $^{233}U(n,f)$

$$R = 0.34 \pm 0.07$$

Result – $^{232}Th(n,f)$

$$R = 0.27 \pm 0.11$$

Integration: 0.5 – 10 MeV
coming from integrals which are in fact numerical evaluations sums with a given step (0.2 MeV)

Steps for IR calculations
- fission XS are taken from Talys for each energy
- Spins distributions are evaluated by statistical approach Huizenga
- parameters for spin distributions are taken from Talys database
- $1/E_n$ neutron flux was chosen
- Yields of isomer and ground state production are evaluated

CONCLUSIONS

- Observables of fast neutron induced fission on ^{232}Th were investigated
- Cross sections, mass distributions, dependence of average prompt neutron multiplicity on fission fragment mass, isotopes production were obtained for incident neutron energy up to 10 MeV
- Calculations were compared with existing experimental data
- XS well described for fast neutrons
- Evaluations were realized with Talys – an efficient tool of experimental data analysis

Perspectives

- New experimental data on neutron induced fission of ^{233}U are planned as necessary
- Project proposals for experiments at FLNP, FLNR JINR basic facilities
- Improvement of theoretical evaluations and computer simulations

THANK YOU VERY MUCH FOR YOUR ATTENTION! ☺