Detailed Modelling of $^{233}$U(n,f)

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OUTLINE OF THE PRESENTATION

INTRODUCTION

ELEMENTS OF THEORY. COMPUTER CODES

RESULTS AND DISCUSSION

CONCLUSIONS AND PERSPECTIVES
INTRODUCTION

General motivation of using $^{233}$U

Isotope $^{233}$U -> obtained as:
1) Fission product in neutron induced fission reactions
2) Bred from $^{232}$Th as fertile nucleus as part of Th – U fuel cycle, neutron irradiation of $^{232}$Th

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

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

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 U fuel 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 $^{233,235}\text{U}$, Pu for criticality
- higher burnup for neutrons economy – hard to reach at LWR
- large periods of time for producing $^{233}\text{U}$ from $^{232}\text{Th}$
- $^{233}\text{U}$ – 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 20 MeV energy on $^{233}$U 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

Talys codes and elements of theory. I

Hauser – Feshbach Approach. XS

\[ \sigma_{\alpha\beta} = \pi A_\alpha^2 \frac{T_\alpha T_\beta}{\sum_c T_c} \]

Historically first HF expression

\[ W_{\alpha\beta} = \text{Widths Fluctuation Correction Factor (WFC)} \]

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

**FM weight**

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

- \( CFM = SL, STI, STII; T^B_{f,CFM} = \) transmission coefficient (Hill – Wheeler);
- \( B = \) second barrier

M. C. Duijvestijn, A. J. Koning, and F. -J. Hambsch, Phys Rev C 64, 014607 (2001)
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

\[ R = \frac{Y_m}{Y_g} = \frac{E_m}{E_{th}} \int N_0 \phi(E_n) \sigma_{nf}^m(E_n) dE_n \]

\[ \phi(E_n) = \text{Incident neutrons flux } \sim 1/E_n \]

\[ N_0 = \text{Number of target nuclei} \]

\[ Y_m, Y_g = \text{Yields of isomer + ground states} \]

\[ E_{th}, E_m = \text{Threshold maximal energy} \]

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

\[ P(J) \sim (2J + 1) Exp \left( -\frac{J(J+1)}{2(\sigma + \lambda)^2} \right) \]

\[ J = \text{spin} \]

\[ \sigma, \lambda = \text{parameters} \]

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

**Fission calculations**
n\(^{+233}\text{U}\) (incident channel) – double humped potential barrier was considered

**First barrier**
Height: 4.35 MeV; Width: 0.8 MeV
Type of axiality: axial symmetry

**Second barrier**
Height: 5.5 MeV; Width: 0.8 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
**Optical model parameters** – n$^{+233}$U incident channel

- For nuclear reaction calculations, for incident and emergent channels are defined local parameters based on experimental data and Wood – Saxon Potential

<table>
<thead>
<tr>
<th></th>
<th>U[MeV]</th>
<th>r[fm]</th>
<th>a[fm$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Central</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Real</td>
<td>46.75</td>
<td>1.245</td>
<td>0.660</td>
</tr>
<tr>
<td>Imaginary</td>
<td>0.09</td>
<td>1.248</td>
<td>0.594</td>
</tr>
<tr>
<td><strong>Surface</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Real</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Imaginary</td>
<td>2.46</td>
<td>1.208</td>
<td>0.614</td>
</tr>
<tr>
<td><strong>Spin-Orbit</strong></td>
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<td></td>
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</tr>
<tr>
<td>Real</td>
<td>5.680</td>
<td>1.121</td>
<td>0.590</td>
</tr>
<tr>
<td>Imaginary</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

**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
Experimental and Theoretical Data are compared in a large energy interval from slow neutrons up to 20 MeV.

- Low energy part, resonant part – Talys gives only average values because databases of neutrons resonance parameters are not implemented.

- Fast neutrons region well described.

Experimental Data – EXFOR

M. Calvini et all, Phys Rev C 80 044604

Experimental and Theoretical Data - Slow Neutrons 0.025 eV – 6E-4 MeV
- New versions of Talys allow to treat slow neutron region using space level, D₀ and other parameters together with extrapolations methods
- The results obtained must be treated carefully and in many cases this treatment gives approximated results
- in this case the slow neutron energy well described
Cross Section. Incident fast neutrons

Left Figure
- Contribution of fissile nuclei formed \((n,xn)\) and capture processes
- Represented XS for \(^{231}\text{U},^{232}\text{U},^{233}\text{U},^{234}\text{U}\)
- Neglected \(^{228,229,230}\text{Th},^{231,232,233}\text{Pa}\)
- first chance fission, second chance fission, etc.

Possible to observe the threshold
For \(^{232}\text{U}\) and \(^{233}\text{U}\) – characteristic shape of Giant Resonance and Compound processes
\(^{231}\text{U}\) – possible at higher energies to have the same shape like \(^{232,233}\text{U}\)
\(^{234}\text{U}\) – formed by capture

Right Figure
- Fast neutron XS – Exp Data described well by Talys
- Contributions of fissile nuclei are reflected in the total XS
Yields of FF mass, distribution and isotopes production are evaluated using Brosa model.

Mass Distribution (MD) and XS Fragment Mass Dependence Before (Pre) and after Neutrons Emission (Post)

Low Energy Part

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

Mass dependence of XS is more sensible to neutron incident energy after 1 MeV
Fission Fragment (FF) Mass Distribution (MD). Thermal point

Applying the approach described in Talys documentation the MD in thermal point \( (E_n=0.0253 \text{ eV}) \) was obtained

Procedure description:
- minimal energy in Talys 1e-6 MeV
- by using a number of values higher than 1e-6 MeV in the low energy part
- \( 1/v \) law

Upper figure
Relative yields as function of FF mass

Lower figure
XS as function of FF mass

In both cases Pre and Post neutrons were considered
MD for some energy values

- Pre and Post Neutrons Emission

- The relative yields are not sensible at the maximum point

- With the increasing of incident neutrons energy the distribution is enlarging which means that more isotopes can be observed / produced.

- With the increasing of the energy MD becomes more symmetric
Prompt neutrons. Thermal Point

\[ ^{233}\text{U}(n,f) \]

**Average Prompt Neutrons Multiplicity**

- Thermal point
- Post and Pre Neutrons Emission
- Obtained by extrapolation
- Results can be considered only qualitatively as for APNM the dependence on fission XS is more complicated
- For incident slow neutrons up to 0.001 MeV the average number of emitted neutrons is changing very slowly

For slow neutrons the average number of emitted prompt neutrons is practically constant

\[ \nu_{prompt} = 2.78 \]
Prompt neutrons. APNM. PNMD. Energy Range 1E-3 – 20 MeV

- For Pre and Post Neutrons Emission
- Number of Emitted Neutrons as Function of FF Mass is increasing in both cases
- Because fission at equal masses has very low probability APNM has low value also

PNMD – increasing with neutrons incident energy but limited by excitation energy
Isotopes Production. $^{99}\text{Mo}$, $^{131}\text{I}$, $^{133}\text{Xe}$. Relative Yields

Relative yields of $^{99}\text{Mo}$, $^{131}\text{I}$ and $^{133}\text{Xe}$

- 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)
- evaluated by own computer code
- it is opening the possibility to investigate and predict isomer ratios obtained in fission
Isotopes Production. $^{99}$Mo, $^{131}$I, $^{133}$Xe. XS

Production of $^{99}$Mo as function of incident energy
- XS for $^{99}$Mo are very low - difficult to separate especially due to the short isotope time of life – requires higher precision than $^{131}$I and $^{133}$Xe
- For $^{131}$I, $^{133}$Xe – High XS for slow neutrons
- possible explanation – these nuclei are closer to stable magic nuclei
Isotopes Production. $^{135}$Xe. Yields and XS

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

- $^{135}$Xe major fission product
- Yields and XS are with order of magnitude higher then $^{99}$Mo, $^{131}$I and $^{133}$Xe
- Obtained with default Talys precision together with variation of parameters of nuclear potential and level density.
- Analogue XS were obtained for a large number of isotopes
### Isotopes Production. Thermal Point

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$Y_{\text{pre}}$</th>
<th>$Y_{\text{post}}$</th>
<th>XS$_{\text{pre}}$[mb]</th>
<th>XS$_{\text{pro}}$[mb]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{92}\text{Mo}$</td>
<td>$7.48 \cdot 10^{-13}$</td>
<td>$3.64 \cdot 10^{-4}$</td>
<td>$1.85 \cdot 10^{-6}$</td>
<td>$87.21$</td>
</tr>
<tr>
<td>$^{131}\text{I}$</td>
<td>$1.96 \cdot 10^{-5}$</td>
<td>$3.96 \cdot 10^{-4}$</td>
<td>$4.71$</td>
<td>$94.49$</td>
</tr>
<tr>
<td>$^{133}\text{Xe}$</td>
<td>$2.96 \cdot 10^{-7}$</td>
<td>$3.38 \cdot 10^{-4}$</td>
<td>$7.08 \cdot 10^{-2}$</td>
<td>$80.98$</td>
</tr>
<tr>
<td>$^{135}\text{Xe}$</td>
<td>$1.44 \cdot 10^{-3}$</td>
<td>$1.29 \cdot 10^{-2}$</td>
<td>$3.46 \cdot 10^{-2}$</td>
<td>$3103$</td>
</tr>
</tbody>
</table>

- Isotopes of interest for thermal neutrons
- Results obtained by extrapolating of theoretical data in accordance with $1/v$ law
- Yields and XS isotopes production is proportional with fission XS
- Lowest production $^{99}\text{Mo}$
- Largest – $^{135}\text{Xe}$ – Yields (major fission product)
Isomer Ratios. $^{133}$Xe

For usual reactions like $(n,p)$, $(n,\alpha)$, $(n,\gamma)$ 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

<table>
<thead>
<tr>
<th>Elem.</th>
<th>Ground (g)</th>
<th>Isomer (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$J^\Pi$</td>
<td>$\tau_g$</td>
</tr>
<tr>
<td>$^{133}$Xe</td>
<td>(3/2)$^+$</td>
<td>5.24 d</td>
</tr>
</tbody>
</table>

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.

Result

$$R = 0.34 \pm 0.07$$

Integration: 0.5 – 20 MeV

Error – 0.07

is coming from integrals which are in fact numerical evaluations sums with a given step.
CONCLUSIONS

• Observables of neutron induced fission process on $^{233}$U was investigated
• Cross sections and their uncertainties, mass distributions, dependence of average prompt neutron multiplicity on fission fragment mass, isotopes production were obtained for incident neutron energy starting from slow up to 20 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! 😊