Measurement of actinides (n,xn $\gamma$) cross sections: Towards the evaluation

Introduction: general context

Gen IV reactor systems

New concepts of reactors
- fast reactors
- accelerator driven systems

New fuel cycle
- $^{238}\text{U} / ^{239}\text{Pu}$
- $^{232}\text{Th} / ^{233}\text{U}$

In reactor, \((n,xn)\) reactions \((x \geq 1)\) contribute to
- Energy loss mechanism
- Neutron multiplication
- Production of radioactive isotopes

Bibliography in data bases shows that improvement of the knowledge of \((n,xn)\) process is necessary.

Important needs of new nuclear data over a wide range of nuclei, energy and reactions

One of the challenges is measurement's accuracy

NEA Nuclear Data
High Priority Request List
Introduction: experimental method

How to study \((n,xn)\) reactions?

- Direct measurement of secondary neutrons
- Activation technique
- \textit{prompt} \(\gamma\)-ray spectroscopy

\textbf{Method:} detection of the \(\gamma\)-rays stemming from the \textit{decay of excited states} of nucleus created by the \((n,xn)\) reaction.

\((n,xn\ \gamma)\) cross section measurements -> \textit{total} \((n,xn)\) cross section:

Need of theoretical model...

\textbf{but}

\((n,xn\ \gamma)\) cross sections:
- can be measured using \textit{“white” neutron beam} with the TOF technique,
- provide \textit{exclusive measurements very restrictive} for testing models.

\textbf{Case of \(^{235}\text{U}\) and \(^{232}\text{Th}\)}
Experimental set-up

GELINA, IRMM, Geel, Belgium

White neutrons beam
few eV → 20 MeV

Energized e-
f = 800 Hz

Uranium Target

30m

Prompt γ spectroscopy

Pulsed Neutron Beam

Fission Chamber

HPGe planar detectors

Sample

150° 110° 150°
110°
Experimental set-up

FP16 – 30 m

- HPGe planar Detector
- Fission Chamber
- Shielding: lead castle
- BEAM
- Digital acquisition system
  TNT2 Card @IPHC

-> Electromagnetic field (from the accelerator) insulation
-> \( \gamma \) Background reduction
Data Analysis

TOF and γ spectra

$^{232}\text{Th}$

Radioactivity

$^{232}\text{Th}(n,n' \gamma)^{232}\text{Th}$

$^{232}\text{Th}(n,2n \gamma)^{231}\text{Th}$

$^{232}\text{Th}(n,3n \gamma)^{230}\text{Th}$

γ-flash
Data Analysis

TOF and $\gamma$ spectra

$\gamma$ flash

$E_n = 20$ MeV

$E_n = 6$ MeV

$E_n = 1$ MeV

radioactivity

$\gamma$ Energy (keV) $\Delta E = 1 - 6$ MeV

$\gamma$ Energy (keV) $\Delta E = 6 - 20$ MeV

$^{235}$U case
Data Analysis

Cross section calculation

$$\frac{d\sigma}{d\Omega}(\theta) = \frac{n_{\text{det}}}{N_{\text{at}} \cdot \phi_n \cdot \varepsilon \cdot t}$$

$n_{\text{det}}$: number of detected $\gamma$

$N_{\text{at}}$: number of atoms in the sample

$\phi_n$: neutron flux

$\varepsilon$: HPGe efficiency

$t$: measurement time

Differential cross section can be expressed by a finite sum of Legendre polynomials:

$$\frac{d\sigma}{d\Omega}(\theta) = \frac{\sigma_{\text{tot}}}{4\pi} \sum_{i=0}^{\infty} a_i P_i(\cos \theta)$$

Measurement at the polynomial nodes Gauss quadrature:

$$\sigma_{\text{tot}} = 4\pi \left[ w_1^* \frac{d\sigma}{d\Omega}\left(\theta_1^*\right) + w_2^* \frac{d\sigma}{d\Omega}\left(\theta_2^*\right) \right]$$

$w_1 = 0.3479$ for $\theta_1 = 30.56^\circ$ or $149.44^\circ$ and $w_2 = 0.6571$ for $\theta_2 = 70.12^\circ$ or $109.88^\circ$
Results: $^{235}\text{U}$

TALYS % Exp data
*(n,n'γ): good agreement in amplitude but the shape is not well reproduced
*(n,2nγ): quite good agreement in the shape but factor of 0.455 in amplitude

D.P. McNabb* exp data % Our exp data
*factor of 0.455 in amplitude but McNabb* data are normalized by an unknown factor

Results: $^{235}$U

**TALYS Considerations:**

**Fissionable isotopes:** How good is the TALYS fission cross section fit? And what about the total (n,n') and (n,2n) cross sections?
Results: $^{232}$Th

TALYS % Exp data

* Total cross sections:
  - Fission: ~ ok
  - $(n,n')$ $(n,2n)$: small underestimation of the code predictions
  - $(n,n' \gamma)$ cross sections for states in ground state band:
    - amplitude ok
    - overestimation above $E_n = 7$ MeV
Results: $^{232}$Th

Preliminary results (data from only 2 detectors): $\gamma$-transitions from few exciting bands to the ground state band for the $(n,n' \gamma)$ process compared to TALYS and another experiment.

TALYS % Exp data
* $(n,n' \gamma)$: agreement is less good than for $\gamma$-transition within the ground state band.

J.H. Dave* exp data % Our exp data
* agreement is very good up to $E_n=2$ MeV (high limit of the J.H. Dave* exp data).

Results: $^{232}$Th

$^{232}$Th(\(n,2n\gamma\))

E\(_{n}\) = 185.7 keV, L\(4 \rightarrow L0\)

\[ \text{TALYS} \% \text{Exp data} \]

*(\(n,2n\gamma\)*: shape is well reproduced but factor of 2.58 in amplitude

*(\(n,3n\gamma\)*: shape is well reproduced but factor of 0.72 in amplitude

Note: the uncertainties are rather large because of the combined effects of the lower cross sections and the low neutron incident flux.
@GELINA / FP16 – 30 m:  
* We have measured \((n,xn \gamma)\) reaction cross sections \((x \geq 1)\) on \(^{235}\text{U}\) and \(^{232}\text{Th}\)  
* **Uncertainties** reached: 5 – 7% (see J.C Thiry thesis 09/2010)

\(^{235}\text{U}:\)
* **New experimental data** in disagreement with the only published one \((n,2n \gamma)\)  
* Highlighted the **TALYS difficulties to predicted with confidence** \((n,xn \gamma)\) cross section (even if the total reaction cross section is well calculated)

\(^{232}\text{Th}:\)
* **New experimental data** in agreement with the published one \((n,n' \gamma)\)  
* Confirmed that, in TALYS, semi-classical exciton model simply fails to properly describe the spin distribution that accompanies the pre-equilibrium process \((E_n \geq 7 \text{ MeV})\).
Towards the evaluation?

What about uncertainties?

Accuracy:
Target accuracies are specified per system and per energy group when they are not met of the current (initial) uncertainties.

<table>
<thead>
<tr>
<th>Energy Range</th>
<th>Initial</th>
<th>ABTR</th>
<th>SFR</th>
<th>EFR</th>
<th>GFR</th>
<th>LFR</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.07-19.6 MeV</td>
<td>29</td>
<td>12</td>
<td>7</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2.23-6.07 MeV</td>
<td>20</td>
<td>5</td>
<td>4</td>
<td>2</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>1.35-2.23 MeV</td>
<td>21</td>
<td>4</td>
<td>5</td>
<td>4</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>0.498-1.35 MeV</td>
<td>12</td>
<td>7</td>
<td>6</td>
<td>5</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>67.4-183 keV</td>
<td>11</td>
<td>7</td>
<td>9</td>
<td>7</td>
<td>4</td>
<td></td>
</tr>
</tbody>
</table>

GEDEPON Workshop: Nuclear Data Evaluation, Issy-les-Moulineaux, France 29-30 November, 2010
Towards the evaluation?

What about uncertainties?

Cross section of a given $\gamma$ ray transition \([b]\)

\[
\frac{d\sigma}{d\Omega}(\theta) = \frac{n_{\text{det}}}{N_{\text{at}} \cdot \phi_n \cdot \varepsilon_\gamma \cdot t}
\]

Detected hits in a given ray:
- Good statistics
- Low background

Number of atoms in target

Incident neutron flux \([s^{-1} \cdot \text{cm}^{-2}]\)

Fission chamber characteristics

$\gamma$ detection efficiency
- Measurements
- Simulations

Acquisition time \([s]\)
Towards the evaluation?

What about uncertainties?

<table>
<thead>
<tr>
<th>parameters</th>
<th>Uncertainties min</th>
<th>Uncertainties max</th>
<th>comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_{\text{det}}$ (for one detector)</td>
<td>1%</td>
<td>15%</td>
<td>Long running time (235U: 1248h; 232Th: 375h)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Neutron flux dependence</td>
</tr>
<tr>
<td>$N_{\text{nat}}$</td>
<td>1.2% (232Th)</td>
<td>2.7% (235U)</td>
<td>Precise mass, thickness, … measurements</td>
</tr>
<tr>
<td>$\Phi_n$</td>
<td>2.3%</td>
<td>2.9%</td>
<td>Simulation + calibration measurements</td>
</tr>
<tr>
<td></td>
<td>1 MeV $&lt; E_n &lt; 2$ MeV</td>
<td>$E_n &lt; 1$ MeV, $E_n &gt; 2$ MeV</td>
<td></td>
</tr>
<tr>
<td>$\varepsilon_\gamma$</td>
<td>3%</td>
<td>6%</td>
<td>Simulation + calibration measurements</td>
</tr>
<tr>
<td></td>
<td>100 keV $&lt; E_\gamma &lt; 1$ MeV</td>
<td>$E_\gamma &lt; 100$ keV</td>
<td></td>
</tr>
</tbody>
</table>

$\Delta \sigma / \sigma$ vary from 5% to 7% in the 0.5 – 9 MeV neutron energy range. But can reach up to 20% in the high neutron energy range (where the neutron flux is low).
Towards the evaluation?

Is it possible to do better?

Neutron flux:
With this set-up, it’s certainly optimized

-> multi layers fission chamber (²³⁵U)?

-> another independent neutron flux measurements with an another reference cross section, H(n,p)?

-> ~1.5%?

HPGe detector efficiency:
some improvements are still possible at low γ energy

-> by carrying out optimization of the simulation (dead zones, multiple diffusions)

-> ~1%?

Total uncertainty in (n,n’) domain -> ~3%
Total inelastic scattering cross section is the sum of the cross section carried by all transitions that directly decay to the ground-state. In general case:

\[
\sigma_n'(E) = \sum_{i=1}^{E_x(L_i) \leq E} \sigma_{n',\gamma}(E, L_i \rightarrow L_{ki}) \frac{p(L_i \rightarrow g.s.)}{p_{\gamma}(L_i \rightarrow L_{ki})}
\]

- Requires a good knowledge of spectroscopic parameters
- Practically, the deduced inelastic cross section is a lower limit for the total inelastic cross section -> model prediction
From \((n,xn \gamma)\) to \((n,xn)\) cross sections?

232\(^{\text{Th}}\) case

We have measured several \((n,n' \gamma)\) cross sections and especially some more intense transitions to the ground state.

The deduced 232\(^{\text{Th}}\)(n,n’) cross section (from the measured \((n,n' \gamma)\) ones) is very close to the total 232\(^{\text{Th}}\) inelastic cross section.
From \((n,\gamma)\) to \((n,xn)\) cross sections?

\(^{235}\text{U}(n,2n)\) case

TALYS 1.0 (P. Romain, CEA/DAM)

We have measured only \(~50\%\) of the total cross section ...

Strong model dependence

-> What is the uncertainty???
Conclusions

**Accuracy business**: we will continue our investigations and efforts to provide the \( (n,xn\gamma) \) cross sections as accurate as possible !!!!

**Deduced total reaction cross section**: collaboration with theoreticians is essential

**Dear evaluators and experimentalists colleagues**: this workshop is the opportunity to discuss about our work so feel free to comment, to review…
Outlooks

@GELINA / FP16 – 30 m
* ANDES project (7ème PCRD)

$^{238}\text{U}(n,xn\gamma)$ reaction cross sections measurement
-> NEA Nuclear Data High Priority Request List (2 – 3%)

@SPIRAL2 / NFS
Day one experiment proposal:
"Comparison between activation and prompt spectroscopy as means of $(n,xn)$ cross section measurements"
(case of $^{90}\text{Zr}(n,3n)$)

@ theoretical aspects
* Collaboration with A.J. Koning (NRG, Petten)
* Collaboration with P. Romain (CEA, Bruyères le Châtel) for TALYS calculations
* Collaboration with M. Sin (University of Bucharest) for EMPIRE calculations

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