(n,xn γ) reaction cross section measurements for (n,xn) reaction studies

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Gen IV reactor systems

New concepts of reactors
- fast reactors
- accelerator driven systems

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

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

In reactor, $(n,\text{x}n)$ 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 $(n,\text{x}n)$ process is necessary.
Introduction: importance of (n,xn) cross section knowledge

**Example of \(^{238}\text{U}(n,n')\)**

**State of the art:**
- **40 measurements** in EXFOR from 1956 to 2009
- **8 total cross section** measurements

=> **large discrepancies**

between experimental data and between evaluated cross sections

=> \(^{238}\text{U}(n,n')\) XS uncertainty = 20 %

**Precise requirement from NEA and CEA/Cadarache**

*Salvatores et al. ; A.Santamarina et al.*

Current uncertainties on \(^{238}\text{U}(n,n')\) impact the accuracy of the \(k_{\text{eff}}\) of the power and of the \(\beta_{\text{eff}}\) calculations of large core reactors (PWR, FR).

**Target accuracy** on \(^{238}\text{U}(n,n')\):

- \(\rightarrow\) PWR: \(\pm 10\%\)
- \(\rightarrow\) SFR: \(\pm 5\%\)
Introduction: experimental method

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

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

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

Example of \(^{238}\text{U}(n,n'\,\gamma)\)
D. Bernard et al.
For small reactor core:
\(k_{\text{eff}}\) \(\rightarrow 50\%\) of its sensitivity from first inelastic threshold
For large reactor core:
radial power \(\rightarrow\) \textbf{sensitivity to the first inelastic levels}

From \((n,xn\,\gamma)\) cross section measurements \textbf{to total} \((n,xn)\) cross section:
Need of structure parameters and theoretical model...
Introduction: IPHC / IRMM / IFIN-HH experimental project

\[(n, xn \gamma)\] reaction cross sections measurements

IPHC (France) / IRMM (Belgium) / IFIN-HH (Romania) collaboration

⇒ development of an experimental set-up GRAPhEME
dedicated to the precise measurement
of the \[(n, xn \gamma)\] reaction cross sections on actinides
@ GELINA facility (IRMM-Belgium)

2005 – 2010: \(^{235}\text{U}\) campaign
2009 – 2010: \(^{232}\text{Th}\) campaign
2009 – 2012: nat,182,183,184,186\(^{W}\) campaign
2011 – 2012: \(^{238}\text{U}\) campaign

Collaboration with theoreticians and evaluators to improve the quality
and the description of our measured cross sections
Experimental set-up @ GELINA: GRAPHEME

**GELINA, IRMM, Geel, Belgium**

- White neutrons beam
- few eV ➔ 20 MeV

- $f = 800 \text{ Hz}$
- $30 \text{m}$
- $e^-$
- Uranium Target

**Prompt $\gamma$ spectroscopy**

- Pulsed Neutron Beam
- Fission Chamber
- HPGe planar detectors
- Sample

Experimental set-up @ GELINA:

- **GRAPHEME**
Experimental set-up @ GELINA : GRAPHEME

FP16 - 30 m

- GRAPhEME
- 4 HPGe planar detectors
- Noise insulation (electromagnetic field from the accelerator)
- and γ background reduction
- Shielding: Pb-Cd-Cu castle

Fission Chamber (²³⁵U)

TNT2 Card @IPHC
Data Analysis

TOF and $\gamma$ spectra

Radioactivity

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

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

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

TOF spectrum

$^{235}\text{U case}$

γ flash

$E_n = 20\text{ MeV}$

$E_n = 6\text{ MeV}$

$E_n = 1\text{ MeV}$

$E_n = 0.1\text{ MeV}$

radioactivity

Time of Flight (μs)
Data Analysis

TOF and $\gamma$ spectra

$^{235}$U case

$\text{(n,n')}$ energy gate spectrum

$\text{(n,2n)}$ energy gate spectrum

Radioactivity spectrum

Counts

Time of Flight (μs)

Energy (keV)

Radioactivity spectrum

Counts

Time of Flight (μs)

Energy (keV)
Data Analysis

Cross section calculation

\[ \frac{d\sigma}{d\Omega}^{(n,x\gamma)}(\theta) = \frac{n_{\text{det}}}{N_{at} \cdot \phi_n \cdot \varepsilon \cdot t} \]

\( n_{\text{det}} \): number of detected \( \gamma \)
\( N_{at} \): 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 \textbf{sum of Legendre polynomials}:

\[ \frac{d\sigma}{d\Omega}^{(n,x\gamma)}(\theta) = \frac{\sigma_{\text{tot}}}{4\pi} \sum_{i=0}^{\infty} a_i P_i(\cos \theta) \]

Measurement at the \textbf{polynomial nodes} \textbf{Gauss quadrature}:

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

\( w_1 = 0.3479 \) for \( \theta_1 = 30.56^\circ \) or \( 149.44^\circ \) and
\( w_2 = 0.6571 \) for \( \theta_2 = 70.12^\circ \) or \( 109.88^\circ \)
Data Analysis

What about uncertainties?

\[ \frac{\Delta \sigma}{\sigma} : 5\% \text{ to } 7\% \text{ for } E_n = 0.5 - 9 \text{ MeV} \]

But can reach up to 20\% for \( E_n > 9 \text{ MeV} \)

(where the neutron flux is low)

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

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

Detected hits in a given ray:
- peak identification (possible contamination)
- Good statistics
- Low background

Number of atoms in target
- Sample composition
- Precise measurements (size, weight)

Acquisition time [s]
- Long time measurements (stability)

\( \gamma \) detection efficiency
- Measurements
- Simulations (attenuation, oxidation)

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

Fission chamber characteristics

But can reach up to 20\% for \( E_n > 9 \text{ MeV} \)

\( \Delta \sigma/\sigma \): 5\% to 7\% for \( E_n = 0.5 - 9 \text{ MeV} \)

Data Analysis

\( \Delta \sigma/\sigma \): 5\% to 7\% for \( E_n = 0.5 - 9 \text{ MeV} \)

But can reach up to 20\% for \( E_n > 9 \text{ MeV} \)

(where the neutron flux is low)
Results

\[ ^{235}\text{U}(n,xn\gamma) \]

**Beam time:** 1466 hours  
**Sample:** enrichment \(^{235}\text{U} 93.2\%\)  
mass 37.43 g  
diameter 12.00 cm  
thickness 0.21 mm

**Bibliography:**  
Very few measurements in EXFOR:  
4 \(\sigma(n,n')\) measurement (1961-1969)  
1 \(\sigma(n,n'\gamma)\) meas. (2000 Younes et al.)

2 \(\sigma(n,2n)\) measurement (1972-1980)  
1 \(\sigma(n,2n\gamma)\) meas. (2000 Younes et al.)

+ \(\sigma(n,xn\gamma)\) in A.L. Hutcheson Thesis (2008)

\(-\rightarrow\) Compare to TALYS calculations (P. Romain, CEA/DAM, FRANCE)
Results

\[ ^{235}\text{U}(n,xn\gamma) * \]

\begin{itemize}
  \item \textbf{exp data} \% Our \textbf{exp data} \\
  \item * discrepancies with Hutcheson data. \\
  \item * agreement with Younes data for the 244 keV \( \gamma \) transition but discrepancies at high neutron energies for the 2 other (n,2n\( \gamma \)) transitions.
\end{itemize}

\begin{itemize}
  \item \textbf{TALYS} \% \textbf{Exp data} \\
  \item * pheno-cgmr is the best parameterization.
  \item * \((n,n'\gamma)\): shape and amplitude are not well reproduced.
  \item * \((n,2n\gamma)\): quite good agreement in the shape but factor 1.5 to 1.9 in amplitude.
\end{itemize}

\*J.C Thiry et al. paper submitted soon
Results

\[ ^{238}\text{U}(n,\alpha \gamma) : \text{preliminary} \]

**Beam time:** 1200 hours

**Sample:** purity natU 99.9 %
- mass 10.62 g
- diameter 7.02 cm
- thickness 0.18 mm

About 30 \((n,\alpha \gamma)\) transitions “spotted”
- 20 preliminary \(\gamma\) transitions in \(^{238}\text{U}\)
- including 5 going to the ground state.
- \(\rightarrow\) Compare to TALYS calculations
  (P.Romain, CEA/DAM, FRANCE)

**Bibliography:**
- lot of total cross section measurements in EXFOR but:
  - \(4 \sigma(n,n' \gamma)\) meas. (1976 Voss et al., 1979 Olsen et al., 2004 Fotiades et al., 2009 Hutcheson et al.)
  - \(2 \sigma(n,2n \gamma)\) meas. (2009 Hutcheson et al., 2004 Fotiades et al.)

**NEA Nuclear Data High Priority Request List**

**\(^{238}\text{U} \text{level scheme}**
**Results**

**$^{238}\text{U}(n,n',\gamma)$ : preliminary**

**N.B.** we are able to measure the de-excitation of the first level in $^{238}\text{U}$

- **exp data**
  - Our exp data
  - Fotiades data slightly higher than our data but good agreement in shape.

- **TALYS**
  - Exp data
  - Shape is well reproduced except in some case when a direct component appears: the relative proportion of the two components are not well calculated.
  - In amplitude, discrepancies depend on the $\gamma$-transition.
Results

$^{232}\text{Th}(n,xn\gamma)$: very preliminary

**Beam time:** 375 hours  
**Sample:** enrichment $^{232}\text{Th}$ 99.5%  
mass 11.99 g  
surface 36.46 cm$^2$  
thickness 0.30 mm

12 $\gamma$ transitions in $^{232}\text{Th}$  
1 $\gamma$ transition in $^{231}\text{Th}$  
-> Compare to TALYS calculations  
(A. Koning, NRG, The Netherland)

**Bibliography:**
Several measurements in EXFOR:  
3 $\sigma(n,n')$ measurement (1962-1983)  
12 $\sigma(n,n')$ level production measurement (1962-2001)  
1 $\sigma(n,n'\gamma)$ meas. (1985 Dave et al.)  
21 $\sigma(n,2n)$ measurement (1956-2011)  
0 $\sigma(n,2n\gamma)$ meas.
Results

\(^{232}\text{Th}(n, n' \gamma)\): very preliminary

N.B. we are able to measure the de-excitation of the first level in \(^{232}\text{Th}\).

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

TALYS % Exp data
* amplitude is well reproduced for states in ground state band but overestimation above \(E_n = 7\) MeV.
* for other \(\gamma\)-transitions the agreement is less good.
Results

\[ ^{186,184,183,182}\text{W}(n,xn \gamma) : very preliminary \]

**Beam time:** 300 hours \(^{186,184,183}\text{W} \)
500 hours \(^{182}\text{W} \)

**Sample:** enrichment \(^{186,184,182}\text{W} \sim 94.5 \% \)
\(^{183}\text{W} \sim 83.75 \% \)

mass \sim 45 to 49 g
diameter \sim 6.6 to 7.1 cm
thickness \sim 0.13 to 1.30 mm

**Bibliography:**
Few measurements in EXFOR:

\(^{182}\text{W} : 2 \sigma(n,n') \) measurement (1967 – 1999)
7 \(\sigma(n,2n)\) measurement (1959 – 1997)

\(^{183}\text{W} : 2 \sigma(n,n') \) measurement (1982 – 1996)
1 \(\sigma(n,2n)\) measurement (1980)

\(^{184}\text{W} : 3 \sigma(n,n') \) measurement (1967 – 2003)
4 \(\sigma(n,2n)\) measurement (1966 – 1982)

\(^{186}\text{W} : 3 \sigma(n,n') \) measurement (1967 – 1996)
14 \(\sigma(n,2n)\) measurement (1959 – 1999)

\(^{182}\text{W} \) sample
27 \(\gamma\) transitions in \(^{182}\text{W} \)
4 \(\gamma\) transitions in \(^{181}\text{W} \)

\(^{183}\text{W} \) sample
17 \(\gamma\) transitions in \(^{183}\text{W} \)
5 \(\gamma\) transitions in \(^{182}\text{W} \)

\(^{184}\text{W} \) sample
15 \(\gamma\) transitions in \(^{184}\text{W} \)
4 \(\gamma\) transitions in \(^{183}\text{W} \)

\(^{186}\text{W} \) sample
15 \(\gamma\) transitions in \(^{186}\text{W} \)
3 \(\gamma\) transitions in \(^{185}\text{W} \)

-> Compare to TALYS calculations
(P.Romain, CEA/DAM, FRANCE)
Results

$^{186,184,183,182}$W(n,xn $\gamma$) : very preliminary

$^{184}$W(n,n$'$ $\gamma$)

Preliminary conclusions

TALYS % Exp data
* in most cases the shape is well reproduced but the discrepancies for the amplitude are different for each $\gamma$ transition.
* branching ratio data bases play an important role.
From \((n,xn\gamma)\) to \((n,xn)\) cross sections?

**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:

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

- 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 \((n,x_n \gamma)\) to \((n,x_n)\) cross sections?

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

We have measured only \(~20\%\) of the total cross section ...

*Strong model dependence*
From \((n,xn\gamma)\) to \((n,xn)\) cross sections? Discussion

**What did we learn?**

**Experimental point of view:**
- **control** and **minimize all source of error**
- **matrix covariance calculation**
- measurement of a **maximum of \(\gamma\)-transitions**
- efforts have to be done to measure the \(\gamma\)-transitions to the ground state

**Theoreticians and evaluators point of view:**
- for fissionable nuclei, \(\sigma(n,f)\) must be **well described**
- nucleus **structure, branching ratios and internal conversion coefficients** play an **important role**
- another approach than the **exciton** model (TALYS-1.2) has to be **tested** to model the **pre-equilibrium reactions**
Conclusion

**Covariance matrix**

\[ \sigma(n,xn,\gamma) \]

\[ ^{235}\text{U}, ^{238}\text{U}, ^{232}\text{Th}, 186,184,183,182\text{W} \]

**Precise** \[ \sigma(n,xn,\gamma) \]

**Highly radioactive targets**

\[ ^{233}\text{U} \text{ (}^{232}\text{Th cycle)} \]

**Segmented HPGe**

**Branching ratio**

**Conversion electron measurements**

**Collaboration with evaluators:**
- Quality of experimental data

**Theoreticians:**
- Quality of model predictions
The authors thank the team of the GELINA facility for the preparation of the neutron beam and for their strong support day after day....