Measurement of the neutron-induced capture-to-fission cross section ratio in $^{233}$U at LANSCE

2023 Annual NCSP Technical Program Review

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Motivation

- Th-U alternative to U-Pu fuel cycle due to its reduced amount of transuranium elements.
- $^{232}$Th is more abundant in nature than uranium.
- In the Th fuel cycle the $^{232}$Th, transmutes into the fissile isotope $^{233}$U.

\[ n + ^{232}_{90}Th \rightarrow ^{233}_{90}Th \beta^- \rightarrow ^{233}_{91}Pa \beta^- \rightarrow ^{233}_{92}U \]

- $^{233}$U(n,f) produces a large rate of emitted neutrons, enough to maintain the chain reaction.
- For this reason, the Th fuel cycle may be the basis of thermal breeder reactors, being also suitable to use in fast reactors.
- Chemical advantages from thorium vs uranium: higher melting point and thermal conductivity.

Illustration of the thorium fuel cycle.
Motivation

- Experimental $^{233}\text{U}(n,\gamma)$ cross section data in the literature are scarce and were measured decades ago.
- New report [1] suggests that a simultaneous measurement with capture would be useful.
- For $^{233}\text{U}$ fission is around one order of magnitude more likely than capture.
  - Good discrimination between gammas coming from capture and fission is required.
- New measurement proposed at LANL combining NEUANCE and DANCE.

Time-of-flight measurements

**p beam** (800MeV, 20Hz) → Spallation target

Proton bunch → Collimator → Flight path (20m)

Water moderator

White neutron bunch → "Stable" sample

Detector

**Neutron Energy:**

\[ E_n = m_n c^2 \left( \frac{1}{\sqrt{1 - \left( \frac{v}{c} \right)^2}} - 1 \right) \]

with:

\[ v = \frac{L}{T} \]

**Flux**

\[ \text{Flux}_{n} = 3 \times 10^5 \text{ n/s/cm}^2/\text{dec} \]
Detectors

**DANCE (Detector for Advanced Neutron Capture Experiments)**
- $4\pi$BaF$_2$ $\gamma$-ray calorimeter composed by 160 crystals with an inner cavity of 17 cm radius [2].
- Used to measure neutron capture cross section data on small quantities of radioactive isotopes.
- We can measure $E_n$, $E_{sum}$, $E_{cl}$, and $M_{cl}$, providing more information than with C6D6 detectors.

**NEUANCE (NEUtron detector array at dANCE)**
- Neutron detector array that consists in 21 stilbene crystals arranged in a cylindrical geometry around the beam pipe [3].
- Used to detect neutrons coming from fission and determine by coincidence with DANCE, the gammas coming from fission.
- NEUANCE detects neutrons with energies above 500 keV (fission neutrons have these energies), therefore **low energy scattered neutrons** that are below this threshold are discriminated.
- Possibility to use a thick target.
- NEUANCE can also detect gammas.

Fission tagging process

- Search for coincidences between the two detectors.
- The DANCE gammas in coincidence with the NEUANCE neutrons are tagged as fission gammas.
- The purpose of tagging is to define the shape of the fission $\gamma$-ray spectrum that can be subtracted from the total spectrum.
Background studies

- The background varies with the neutron energy, therefore it is subtracted per En bin.

\[ Q \text{ value peak} = 6.845 \text{ MeV} \]

Mcl=(4,5)

En = 300eV

Counts

$E_{\text{tot}} (\text{MeV})$

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Capture-to-fission ratio

The capture-to-fission ratio is given by:

\[ \alpha(E_n) \equiv \frac{\sigma_{\gamma}(E_n)}{\sigma_f(E_n)} \]
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$$C_i(E_n) = \varepsilon_i Y_i(E_n)$$
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\[ = \frac{k \sigma_\gamma(E_n)}{\sigma_f(E_n)} \]

Hence:

\[ \alpha(E_n) \equiv \frac{1}{k} \frac{C_\gamma(E_n)}{C_f(E_n)} \]
Capture-to-fission ratio

- Experimental advantages of the capture-to-fission ratio:
  - It is much simpler and more reliable to determine experimentally as many of the systematic questions:
    - Sample mass
    - Self-shielding
    - Neutron exposure
  will cancel out in an appropriately designed experiment.
Capture-to-fission ratio

- Normalization to ENDF/B-VIII.0 broadened cross section ratio in the neutron energy region suggested by the Luiz Leal and Marco Pigni (8.1-14.7) eV:

![Graph showing capture-to-fission ratio with different data sets compared.](image)
Capture-to-fission ratio

\[ \frac{\alpha}{E_n (\text{eV})} \]

This work
Berthomieux 1 (2007)
Berthomieux 2 (2007)
Weston (1968)
ENDF/B-VIII.0 broadened
ENDF/B-VIII.0

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Capture-to-fission ratio

- This work
- Berthomieux 2 (2007)
- Weston (1968)
- JEFF-3.3
- ENDF/B-VIII.0 broadened
- ENDF/B-VIII.0

- This work
- Weston (1968)
- JEFF-3.3
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- This work
- Hopkins (1962)
- JEFF-3.3
- JENDL-5
- ENDF/B-VIII.0
The capture cross section was calculated by multiplying the capture-to-fission ratio by the ENDF/B-VIII.0 fission cross section.

The broadened cross section was used in the Resolved Resonance Region.

Remember that this is not an independent measurement of the capture cross section.
Statistical Model Calculation

- Statistical model calculations were performed by I. Stetcu, T. Kawano and A. Lovell with the CoH3 code [4] from 1 keV to 5 MeV (Energy for which only the first fission chance is involved).
- This code combines the coupled-channels optical model and the statistical Hauser-Feshbach model calculations by performing the Engelbrecht-Weidenmüller transformation of the penetration matrix.
- Different values of the average $\gamma$-ray width have been tried by adjusting the M1 $\gamma$-ray strength function for the scissors mode.
- Mughabghab gives 40 meV.
- To reproduce the data from Hopkins it had to be reduced to 24 meV.
- A smaller value would be needed to reproduce this work.

Conclusions

- New measurement at LANSCE combining DANCE and NEUANCE at the end of 2020 and 2021.
- Two samples of 10 mg and 20 mg of $^{233}\text{U}$ have been prepared at LANL by Evelyn M. Bond (December 2020).
- Data analysis has been finished and results of the capture-to-fission ratio on $^{233}\text{U}$ in the neutron energy region from 0.7 eV to 250 keV have been provided.
- The focus was to provide data from 1-300 keV. We are providing data from 0.7 eV to 1 keV in addition.
- The result has been normalized to the ENDF/B-VIII.0 broadened capture-to-fission cross section ratio in the neutron energy region recommended by the Luiz Leal and Marco Pigni, between 8.1 and 14.7 eV.
- This is the first measurement of the capture-to-fission ratio between 2 - 30 keV.
- The data show some small differences in the RRR with the evaluation though the general trend is consistent.
- In the URR this data show a smaller capture-to-fission ratio than the evaluation from 10 to 150 keV.
Acknowledgements

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Los Alamos National Laboratory

Luiz Leal
Institut de Radioprotection et de Sûreté Nucléaire (France)

Marco T. Pigni
Oak Ridge National Laboratory
The 30 mg of $^{233}$U were supplied from Oak Ridge National Laboratory (ORNL).

Material composition:

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Atom (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{233}$U</td>
<td>99.9843</td>
</tr>
<tr>
<td>$^{234}$U</td>
<td>&lt;0.0002</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>0.0017</td>
</tr>
<tr>
<td>$^{236}$U</td>
<td>0.0004</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>0.0134</td>
</tr>
</tbody>
</table>

Two samples have been prepared by Evelyn M. Bond at LANL.

- 20 mg
- 10 mg
**PSD NEUANCE**

- Neutrons & gammas separation using the plot (long-short)/long vs long.

- Clear discrimination between fission neutrons and $\gamma$-rays.
DANCE calibrations

- Intrinsic radioactivity of BaF$_2$ used to calibrate the DANCE crystals.
- Using the Alpha-decay chain of the $^{226}$Ra present in the BaF$_2$.
  - $^{226}$Ra (4.8 MeV)
  - $^{222}$Rn (5.5 MeV)
  - $^{218}$Po (6.0 MeV)
  - $^{214}$Po (7.7 MeV)
NEUANCE calibrations

- Calibration using gamma sources:
  - $^{22}$Na (511 keV and 1274.537 keV).
  - $^{137}$Cs (661.657 keV).
  - $^{88}$Y (898.047 keV and 1836.090 keV).