Neutron-induced fission cross section of $^{240,242}\text{Pu}$ at the Van de Graaff accelerator (EC-JRC-IRMM)

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Outline

1. Introduction
2. JRC-IRMM
3. Experimental setup
4. Data acquisition and treatment
5. Measurements
6. Summary
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What is a cross section?

\[
\text{neutron} + \text{isotope} \rightarrow 2 \text{ fission fragments} + \text{neutrons}
\]

\[
\sigma_{\text{isotope}}(E_n) = 10^{-24} \text{ cm}^2 \text{ (barn)}
\]
For the longer term, more innovative nuclear energy technologies and fuel cycles, known collectively as Generation IV systems, are being developed through international co-operation. The most important initiative to co-ordinate research and development (R&D) efforts on advanced reactors and fuel cycles is the Generation IV International Forum (GIF). Formed in 2001, GIF brings together the major countries involved, including Canada, China, France, Japan, the Republic of Korea, the Russian Federation, South Africa, Switzerland and the United States, plus Euratom. The aim is to develop systems that offer improved sustainability, economics, safety and reliability, proliferation resistance and physical protection.

Six conceptual nuclear energy systems were selected for collaborative R&D, comprising the sodium-cooled fast reactor (SFR), the very-high-temperature reactor (VHTR), the supercritical-water-cooled reactor (SCWR), the gas-cooled fast reactor (GFR), the lead-cooled fast reactor (LFR), and the molten salt reactor (MSR). Each of these has reached a different stage of development, depending on the R&D efforts that have been made in the past and the level of commitment each has received from participating countries.

The most mature Generation IV concepts are the SFR and VHTR, which are based on proven technology. These are the leading candidates for large-scale demonstration projects, the first of which could be in operation in the 2020s. The R&D on sodium-cooled reactors draws on a long experience of operation of various prototypes, in the United States, the United Kingdom, France, the Russian Federation and Japan. Many of those are now shut down, but new sodium fast reactors (which are not considered as Generation IV), are being built in the Russian Federation, India and China. Other reactor concepts may require smaller scale prototypes before full-scale demonstration. The first commercial Generation IV systems are not expected to be available before the 2030s, with their full introduction unlikely before the 2040s. Hence, Generation IV reactors are not expected to be a major part of installed nuclear capacity until well after 2050. Figure 9.5 shows the successive generations of nuclear reactors, including their deployment timeline.
Fast neutron reactors

1 Energy spectra 0.5 MeV to 20 MeV

At thermal energies

235U → fissile
238U → fertile

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Fast neutron reactors

2 Highly enriched $^{238}\text{U}$ to function

$^{238}\text{U}$ captures a neutron
↓
one neutron less
↓
Production of minor actinides and plutonium
High Priority request list

Table 32. Summary of Highest Priority Target Accuracies for Fast Reactors

<table>
<thead>
<tr>
<th></th>
<th>Energy Range</th>
<th>Current Accuracy (%)</th>
<th>Target Accuracy (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U238</td>
<td>$\sigma_{\text{fis}}$ 6.07 + 0.498 MeV</td>
<td>10 + 20</td>
<td>2 + 3</td>
</tr>
<tr>
<td></td>
<td>$\sigma_{\text{capt}}$ 24.8 + 2.04 keV</td>
<td>3 + 9</td>
<td>1.5 + 2</td>
</tr>
<tr>
<td>Pu241</td>
<td>$\sigma_{\text{fis}}$ 1.35 MeV ± 454 eV</td>
<td>8 + 20</td>
<td>2 + 3 (SFR, GFR, LFR)</td>
</tr>
<tr>
<td></td>
<td>$\sigma_{\text{fis}}$ 5 + 8 (ABTR, EFR)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu239</td>
<td>$\sigma_{\text{capt}}$ 498 + 2.04 keV</td>
<td>7 + 15</td>
<td>4 + 7</td>
</tr>
<tr>
<td>Pu240</td>
<td>$\sigma_{\text{fis}}$ 1.35 + 0.498 MeV</td>
<td>6</td>
<td>1.5 + 2</td>
</tr>
<tr>
<td></td>
<td>$\sigma_{\text{fis}}$ 1.35 + 0.498 MeV</td>
<td>4</td>
<td>1 + 3</td>
</tr>
<tr>
<td>Pu242</td>
<td>$\sigma_{\text{fis}}$ 2.23 + 0.498 MeV</td>
<td>19 + 21</td>
<td>3 + 5</td>
</tr>
<tr>
<td>Pu238</td>
<td>$\sigma_{\text{fis}}$ 1.35 + 0.183 MeV</td>
<td>17</td>
<td>3 + 5</td>
</tr>
<tr>
<td>Am242m</td>
<td>$\sigma_{\text{fis}}$ 1.35 MeV ± 67.4 keV</td>
<td>17</td>
<td>3 + 4</td>
</tr>
<tr>
<td>Am241</td>
<td>$\sigma_{\text{fis}}$ 6.07 + 2.23 MeV</td>
<td>12</td>
<td>3</td>
</tr>
<tr>
<td>Cm244</td>
<td>$\sigma_{\text{fis}}$ 1.35 + 0.498 MeV</td>
<td>50</td>
<td>5</td>
</tr>
<tr>
<td>Cm245</td>
<td>$\sigma_{\text{fis}}$ 183 + 67.4 keV</td>
<td>47</td>
<td>7</td>
</tr>
<tr>
<td>Fe56</td>
<td>$\sigma_{\text{fis}}$ 2.23 + 0.498 MeV</td>
<td>16 + 25</td>
<td>3 + 6</td>
</tr>
<tr>
<td>Na23</td>
<td>$\sigma_{\text{fis}}$ 1.35 + 0.498 MeV</td>
<td>28</td>
<td>4 + 10</td>
</tr>
<tr>
<td>Pb206</td>
<td>$\sigma_{\text{fis}}$ 2.23 + 1.35 MeV</td>
<td>14</td>
<td>3</td>
</tr>
<tr>
<td>Pb207</td>
<td>$\sigma_{\text{fis}}$ 1.35 + 0.498 MeV</td>
<td>11</td>
<td>3</td>
</tr>
<tr>
<td>Si28</td>
<td>$\sigma_{\text{fis}}$ 6.07 + 1.35 MeV</td>
<td>14 + 50</td>
<td>3 + 6</td>
</tr>
<tr>
<td></td>
<td>$\sigma_{\text{capt}}$ 19.6 + 6.07 MeV</td>
<td>53</td>
<td>6</td>
</tr>
</tbody>
</table>
ANDES (Accurate Nuclear Data for nuclear Energy Sustainability):

- 3-year project (started mid 2010)
- 20 partners from 15 countries in Europe (CIEMAT, JRC-IRMM, n_TOF-CERN, CNRS, ...)
- WP1: Measurements for advance reactor systems
ANDES (Accurate Nuclear Data for nuclear Energy Sustainability):

- 3-year project (started mid 2010)
- 20 partners from 15 countries in Europe (CIEMAT, JRC-IRMM, n_TOF-CERN, CNRS, ...)
- WP1: Measurements for advance reactor systems
  \[ \sigma_{240,242\text{Pu}}(E_n) \rightarrow \text{JRC-IRMM, n\_TOF, CNRS} \]
- 3 different experiments on \( \sigma_{240,242\text{Pu}}(E_n) \)
- relative to different standards (\(^{235}\text{U},^{237}\text{Np},^{238}\text{U},^{1}\text{H}\))
How to measure cross sections
How to measure cross sections

ABSOLUTE

- High accuracy on the neutron flux determination
How to measure cross sections

**ABSOLUTE**
- High accuracy on the neutron flux determination

**RELATIVE**
- Back-to-back configuration
- No need of measuring the flux
- High accurate reference XS ($^{235}$U, $^{238}$U, $^{237}$Np, ...)

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Fission cross section measurements of $^{240,242}$Pu
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- The Van de Graaff accelerator
- The GELINA accelerator

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The Van de Graaff facility at JRC-IRMM

- 7 MV belt-driven, electrostatic and vertical accelerator
- Protons, deuterons or alpha particles
- 2 target halls
- Quasi-monoenergetic neutron beams
- Very high neutron energy resolution
- High neutron flux
- From 100 keV up to 20.5 MeV
- Reactions available: $^7\text{Li}(p,n)^7\text{Be}$, $^7\text{Li}(d,n)^3\text{He}$, $^3\text{He}$, $^7\text{Li}(d,n)^4\text{He}$
The Van de Graaff facility at JRC-IRMM

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Twin Frisch-Grid Ionization Chamber

NEUTRON BEAM

1.0 kV
0.0 kV
-1.5 kV
0.0 kV
1.0 kV

ANODE 1
GRID 1
CATHODE
GRID 2
ANODE 2

6 mm
6 mm

D= 31 mm
D= 31 mm
Experimental setup

Twin Frisch-Grid Ionization Chamber + Digitizer

NEUTRON BEAM

1.0 kV
0.0 kV
-1.5 kV

237\text{Np}, 238\text{U}, 240,242\text{Pu}

90\% \text{Ar} + 10\% \text{CH}_4
1052 \text{mbar}

HV 1.0 kV

ANODE
GRID
CATHODE

PA

Waveform digitizer
12 BIT 100 MHz

PC

Timing filter amplifier
Constant fraction discr.
TRIGGER
Experimental setup

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### Sample description

<table>
<thead>
<tr>
<th></th>
<th>$^{240}$Pu</th>
<th>$^{242}$Pu</th>
<th>$^{237}$Np</th>
<th>$^{238}$U</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Method</strong></td>
<td>electrodeposition</td>
<td>electrodeposition</td>
<td>vacuum deposition</td>
<td>vacuum deposition</td>
</tr>
<tr>
<td><strong>Mass ($\mu g$)</strong></td>
<td>92.9 (0.4%)</td>
<td>671 (0.9%)</td>
<td>391.3 (0.3%)</td>
<td>614 (0.5%)</td>
</tr>
<tr>
<td><strong>Diameter (mm)</strong></td>
<td>29.95 (0.1%)</td>
<td>29.95 (0.1%)</td>
<td>12.7</td>
<td>30</td>
</tr>
<tr>
<td><strong>Areal density ($\mu g/cm^2$)</strong></td>
<td>13.19 (0.4%)</td>
<td>95.3 (0.8%)</td>
<td>308.9</td>
<td>86.9</td>
</tr>
<tr>
<td><strong>Backings</strong></td>
<td>Aluminum</td>
<td>Aluminum</td>
<td>Stainless steel</td>
<td>Aluminum</td>
</tr>
<tr>
<td><strong>$\alpha$-activity (MBq)</strong></td>
<td>0.780 (0.4%)</td>
<td>0.095 (0.3%)</td>
<td>0.001 (0.1%)</td>
<td>$7 \cdot 10^{-6}$ (0.5%)</td>
</tr>
<tr>
<td>$%^{238}$Pu</td>
<td>0.0733</td>
<td>0.0027</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$%^{239}$Pu</td>
<td>0.0144</td>
<td>0.0044</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$%^{240}$Pu</td>
<td>99.8915</td>
<td>0.0192</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$%^{241}$Pu</td>
<td>0.0041</td>
<td>0.0081</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$%^{242}$Pu</td>
<td>0.02027</td>
<td>99.9652</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$%^{244}$Pu</td>
<td>0.00005</td>
<td>0.0004</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

All samples made at JRC-IRMM
Outline

1 Introduction

2 JRC-IRMM

3 Experimental setup

4 Data acquisition and treatment

5 Measurements

6 Summary
Data acquisition and treatment

Ideal signals collected

Real signals detected ($^{240}$Pu)
Data acquisition and treatment

Ideal signals collected

Real signals detected (\(^{240}\text{Pu}\))

\[ \text{\(\alpha\)-particle pile-up} \]
Data acquisition and treatment

Ideal signals collected

Real signals detected ($^{240}\text{Pu}$)

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Fission cross section measurements of $^{240,242}\text{Pu}$
Improvement of the signals detected: P10 as counting gas

![Graphs showing fission fragments and α particles for 242Pu and 240Pu](image)

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Fission cross section measurements of $^{240,242}$Pu
Improve the signals detected: CH$_4$ as counting gas

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Spontaneous fission measurements

\[ T_{1/2, SF} = \left( \frac{\%_{Pu}^j}{A_j} \right) \left( \frac{1}{\left( \frac{C_{SF}}{t \cdot \epsilon_j \cdot \ln 2 \cdot m_{Pu} \cdot N_A} - \sum_{i}^{n} \frac{\%_{Pu}^i}{A_i \cdot T_{1/2, SF}(i)} \right)} \right) \]
Spontaneous fission measurements

(a) $T_{1/2,\text{sf}} (10^{11}\text{ yr})$

- This experiment
- Literature values

(b) $T_{1/2,\text{sf}} (10^{10}\text{ yr})$

- This experiment
- Literature values

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Fission cross section measurements

\[ \sigma_{0,\text{Pu}}(E_i) = \left[ \frac{m_{\text{ref}} \cdot A_{\text{Pu}}}{m_{\text{Pu}} \cdot A_{\text{ref}} \cdot \%^{i}\text{Pu}} \cdot \frac{C_{\text{Pu}}/\epsilon_{\text{Pu}} - C_{\text{SF}}}{C_{\text{ref}}/\epsilon_{\text{ref}}} - \frac{A_{\text{Pu}}}{\%^{i}\text{Pu}} \cdot \sum_i \frac{\sigma_{i^{i}\text{Pu}}}{A_{i^{i}\text{Pu}} \sigma_{\text{ref}}} \right] \cdot \left( \frac{\Phi_0^{\text{ref}}}{\Phi_0^{\text{Pu}}} \sigma_0^{\text{ref}} + \frac{\Phi_1^{\text{ref}}}{\Phi_0^{\text{Pu}}} \sigma_1^{\text{ref}} \right) - \frac{\Phi_1^{\text{Pu}}}{\Phi_0^{\text{Pu}}} \sigma_1^{\text{Pu}} \]

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Fission cross section measurements of $^{240,242}\text{Pu}$
Fission cross section measurements: Preliminary results

\[ \sigma_f (\text{barn}) \]

\[ E_n (\text{MeV}) \]

Pu\(^{240}\)(n,f) and Pu\(^{242}\)(n,f) fission cross sections compared with different nuclides and libraries.
The $^{237}$Np evaluation

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Fission cross section measurements of $^{240,242}$Pu

---

Jiacoletti 1972
Paradela 2010
ENDF/B-VII.1
JEFF 3.1
Fission cross section measurements: Preliminary results

\[
\sigma_f (\text{barn}) = \begin{cases} 
0 & \text{for } E_n = 0 \\
0.0 & \text{for } E_n = 0.5 \\
0.5 & \text{for } E_n = 1.0 \\
1.0 & \text{for } E_n = 1.5 \\
1.5 & \text{for } E_n = 2.0 \\
2.0 & \text{for } E_n = 2.5 \\
\end{cases}
\]

\[
E_n (\text{MeV}) 
\]

Pu(n,f) from
- ENDF - $^{237}$Np norm
- ENDF - $^{238}$U norm
- Paradela - $^{237}$Np norm
- Tovesson - 2009
- Laptev - 2004
- Meadows - 1981
- ENDF/B-VII.1
- JEFF 3.1

Pu(n,f) from
- ENDF - $^{237}$Np norm
- ENDF - $^{238}$U norm
- Paradela - $^{237}$Np norm
- Tovesson - 2009
- Weigmann - 1984
- Meadows - 1978
- Auchampaugh - 1971
- ENDF/B-VII.1
- JEFF 3.1

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Why?
Nuclear data needs for GEN-IV nuclear power plants $\rightarrow$ stringent uncertainties
Summary

Why?
Nuclear data needs for GEN-IV nuclear power plants $\rightarrow$ stringent uncertainties

How?
New cross section data relative to other isotopes than $^{235}$U
Need of digital electronics with high $\alpha$-active targets (e.g. $^{240}$Pu)
Summary

Why?
Nuclear data needs for GEN-IV nuclear power plants → stringent uncertainties

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New cross section data relative to other isotopes than $^{235}$U
Need of digital electronics with high $\alpha$-active targets (e.g. $^{240}$Pu)

Where?
VdG accelerator → fast neutron energies
Summary

**Why?**
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**How?**
New cross section data relative to other isotopes than $^{235}$U
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**Where?**
VdG accelerator → fast neutron energies

**What?**
Detailed study of the efficiency → fission fragment loss within sample
New spontaneous fission half-lives for $^{240,242}$Pu → uncertainties <1.3%
New relevant cross section data on $^{240,242}$Pu
Summary

Why?
Nuclear data needs for GEN-IV nuclear power plants → stringent uncertainties

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Where?
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New relevant cross section data on $^{240,242}$Pu

Future needs
Investigation of the $^{237}$Np cross section
Thank you for your attention!

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