Partial Neutron Capture Cross Section Determination of $^{237}\text{Np}$, $^{242}\text{Pu}$ and $^{241}\text{Am}$ using Cold Neutron Beams

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How should we answer without knowing what is inside?
Prompt Gamma Activation Analysis

Non-destructive assay for the characterization of nuclear waste samples

neutrons

photons

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Identification and Quantification

- $^{27}\text{Al}$, $^{56}\text{Fe}$, ...
- $^{14}\text{N}$, $^{16}\text{O}$, ...
- Minor actinides: $^{237}\text{Np}$, ....

Counts vs. Energy [MeV] graphs are shown, indicating the detection of various elements in different energy ranges.
Motivation and Principle

\[ \sigma_{\gamma,Np237} = 141.7 \pm 5.4 \text{ b} \]
\[ \sigma_{\gamma,Np237} = 182.2 \pm 4.5 \text{ b} \]

Neutron Capture Data of Minor Actinides are inconsistent
But important for designing facilities
Sample Design

- Safe containment for actinides
- Transparency for neutrons
- Transparency for photons
- Avoid elements that change the neutron beams temperature
- As few elements as possible to reduce interference
Sample Preparation

Preparation of $^{242}$Pu and $^{237}$Np samples in quartz vials

$^{242}$Pu oxide powder filled into a quartz vial going to be sealed

The vial sealed in a teflon bag while measuring the decay gamma spectrum on a low energy Ge detector
Neutron Radiography

Neutron Radiographic Images of the quartzvial samples taken at the Budapest facility

$^{237}$NpO$_2$ + Au $^{237}$NpO$_2$

$^{242}$PuO$_2$ + Au $^{242}$PuO$_2$

Au powder could not be mixed with the Actinide oxides
Pressing Pellets

- Geometry is much better defined
- Homogenous mixture with a gold flux monitor not feasible
- Using a 3mm thick Gold foil as a separate flux monitor
Preparation of $^{242}\text{PuO}_2$ and $^{237}\text{NpO}_2$ samples as pellets mounted between Suprasil© quartz slabs (4 x 4 x 0.02 cm)

Glovebox at Forschungszentrum Jülich where the sample preparation took place

Examply one readily prepared 237NpO2 sample. For flux measurement an equaly sized Gold foil (3μm) was placed under the pellet.
Neutron Radiography and $^{241}$Am sample

Radiographic image of most massive $^{237}$NpO$_2$ and Gold sample.

Preparation of $^{241}$Am nitrate sample. A 12$\mu$l droplet of solution is dried on the surface of a thin gold foil.

Samples can be regarded as thin!
PGAA facility at Budapest Research Reactor

\[ \phi_n \approx 7 \times 10^7 \frac{n}{cm^2 s} \]

Cold neutron beam

Sample

Compton suppressor BGO

HPGe detector

Lead collimator

\(^{6}\text{Li}\) plastic
Measurement

Samples are placed at an angle of 30° with the neutron beam in the sample chamber in front of the detector.
PGAA facility at FRM-II

\[ \phi_n \approx 2.5 \cdot 10^{10} \frac{n}{cm^2 \ s} \]

Cold neutron beam

Sample

HPGe detector

Compton suppressor

BGO

Mitglied in der Helmholtz-Gemeinschaft

C

ompton

BGO

HPGe

detector

S

ample

PGAA

facility

FRM-II

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Measurement

Samples are placed at an angle of 45° with the neutron beam in the sample chamber in front of the detector.
Spectra Comparision

Resonance region is avoided using the cold neutron beams
$^{237}\text{Np}$ Prompt Gamma Spectrum

$^{237}\text{NpO}_2$ (blue) measured during a 7200 s irradiation at the Garching facility.

$^{242}\text{PuO}_2$ (blue) measured during a 7200 s irradiation at the Garching facility.

More than 900 resolvable peaks and an pronounced continuum part
237Np Prompt Gamma Spectrum

First identification using background measurement and Budapest PGAA data
First identification using background measurement and Budapest PGAA data
\(^{242}\text{Pu} \) Prompt Gamma Spectrum

\(^{242}\text{PuO}_2\) (blue) and blank sample spectrum (red) measured during a 54000 s irradiation at the Budapest facility.

More than 800 resolvable peaks and an pronounced continuum part
Evaluation of $^{237}$Np Peaks

Correct peak areas for the attenuation in the sample itself

Correction factor:

$$\kappa_{sc}(E_{\gamma}) = \frac{P_r}{P_c}$$

Real peak area without self-attenuation

Peak area with self-attenuation

<table>
<thead>
<tr>
<th>Energy [keV]</th>
<th>$\kappa_{sc}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>49.47</td>
<td>1.27</td>
</tr>
<tr>
<td>182.84</td>
<td>1.05</td>
</tr>
<tr>
<td>5352.03</td>
<td>1.00</td>
</tr>
</tbody>
</table>

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Evaluation of $^{237}\text{Np}$ peaks

2

Calculate the relative intensities for $^{237}\text{Np}$ lines

3

Calculate gamma ray production cross sections using the known cross section of a comparator such as gold
Please treat all results as preliminary

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy [keV]</th>
<th>$\sigma_\gamma$ [b]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{242}$Pu</td>
<td>$288.53 \pm 0.03$</td>
<td>$7.67 \pm 0.31$</td>
</tr>
<tr>
<td>$^{242}$Pu</td>
<td>$4335.05 \pm 0.17$</td>
<td>$0.06 \pm 0.01$</td>
</tr>
<tr>
<td>$^{237}$Np</td>
<td>$183.39 \pm 0.03$</td>
<td>$20.83 \pm 0.84$</td>
</tr>
<tr>
<td>$^{237}$Np</td>
<td>$5354.40 \pm 0.05$</td>
<td>$0.47 \pm 0.01$</td>
</tr>
</tbody>
</table>

- 97 prompt $\gamma$-ray peaks of $^{237}$Np identified
- Up to 100 prompt $\gamma$-ray peaks of $^{242}$Pu
- Only 3 prompt $\gamma$-ray peaks of $^{241}$Am identified
- Most intense peaks are at same energy as in literature
- Slight differences in the energy
- **Significant differences in relative intensities**
Significant Deviations

Mean: 0.19 RMS: 2.41
Mean: 3.30 RMS: 1.26 → systematic effect
Thermal Capture Cross Section of $^{237}$Np

Garching (2012)
Budapest (2012)

Letourneau et al. (2010)
Esch et al. (2008)
Harada et al. (2006)
Katoh et al. (2003)
Kobayashi et al. (1994)
Jurova et al. (1984)
Weston et al. (1981)
Eberle et al. (1971)
Schumann et al. (1969)
Tattersall et al. (1960)
Smith et al. (1957)

Thermal Cross Sections Calculated using the 1028 keV decay peak of $^{238}$Np
Thermal Capture Cross Section of $^{242}$Pu

Thermal Cross Sectins Calculated using the prompt peak of $^{242}$Pu at 288 keV $P_\gamma$ from ENDF database has 20% uncertainty
Thermal cross sections calculated using the $^{242}$Pu X-Ray emission at 99 and 103 keV after $^{242g}$Am decay to $^{242}$Pu
Conclusion & Outlook

Conclusion:
- Measurement of the $^{237}\text{Np}$, $^{242}\text{Pu}$ and $^{241}\text{Am}$ prompt gamma spectrum have been performed in the energy range from 40keV to 12 MeV
- Partial $\gamma$-ray production cross sections have been evaluated
- Thermal capture cross sections have been calculated

Outlook:
- Preparation of samples containing other actinides and relevant isotopes in nuclear waste characterization, such as $^{239}\text{Pu}$, $^{243}\text{Am}$, $^{85}\text{Kr}$, $^{226}\text{Ra}$
- Prompt Gamma with fast neutrons at Garching $(n,2n)$, $(n,p)$, $(n,\alpha)$ reactions
Thermal Capture Cross Section of $^{241}\text{Am}$

Thermal cross sections calculated using the $^{242}\text{Pu}$ X-Ray emission at 99 and 103 keV after $^{242\text{g}}\text{Am}$ decay to $^{242}\text{Pu}$
Evaluation of $^{237}$Np Peaks

Correct peak areas for the absorption in the Quartz

Correction via Lambert-Beer law:

$$\frac{P_c}{P_m} = \kappa_{abs} = e^{\mu_{Al}(E_\gamma)x}$$

-$x = d \cos(\theta) \approx 0.29 \text{ mm}$

Detector

Neutron beam
1st Generation Sample

Preparation of $^{237}$Np samples for measurement at external neutron beams. Pellets mounted between Al sheets.

$^{237}$Np oxide powder is filled into a dye for the pill press

The $^{237}$Np pill mounted in a deepening in an Al foil is going to be enclosed with another foil