Uranium gamma data needs for microcalorimetry at the IAEA NML

Geon-Bo Kim  Lawrence Livermore National Laboratory  kim90@llnl.gov
Mark Croce  Los Alamos National Laboratory
Dan Becker  University of Colorado & NIST

Feb-28, 2023
Microcalorimeters

Micro-calorimeters:
• Measure micro-heat produced by radiation absorption using quantum sensors
• Excellent energy resolution of O(10 eV) for ~100 keV gamma-rays
• Mature, and accelerator or user facility deployable

\[ \Delta T = \frac{E}{C} \]

Absorber (Heat Capacity C)

Thermometer

Heat Bath \( T_b < 0.1 \text{ K} \)

\( \alpha, \beta, \gamma, n \) (Energy E)

Magnetic Micro-Calorimeter (MMC)

Transition Edge Sensor (TES)
Microcalorimeter Applications (related to nuclear data)

- Quantification of radionuclides
- High Energy Resolution (x10 better than HPGe)
- Operation range: 1 keV – 300 keV
- TRL 7-9


• 16-pixel gamma microcalorimeter array
Microcalorimeter Applications (related to nuclear data)

- For β spectroscopy or absolute decay counting
- Radionuclide quantification with 100% detection efficiency
- Fission Total Kinetic Energy measurement
- Half-life measurements for nuclear dating and chronology
- TRL 3-6

Energy absorber

\[ \Delta T = \frac{E}{C} \]

Heat Bath \( T_b < 0.1 \text{ K} \)

Decay Energy Spectroscopy (Source-embedded 4π detection)

- Sm-145 EC decay
- Sm-146 alpha

\[ ^{239}\text{Pu} \]
Gamma Microcalorimeter Performance

High precision γ-spectroscopy of U-233

ΔEFWHM = 38 eV @ 60 keV (x 10 better than planar HPGe)
Microcalorimeters at the IAEA Nuclear Material Laboratory

SOFIA Gamma Spectrometer (NIST/LANL)

Planned deployment of Microcal. Systems at IAEA NML in 2025

New capabilities and needs of nuclear data by deployment of microcalorimeters

Source: D. Calma/IAEA
Uranium Enrichment Measurement at the IAEA NML

- Measure $^{235}$U Enrichment level by $^{235}$U/$^{238}$U ratio (one of the most important measurements)
- Currently there is no method for direct measurement of $^{238}$U
- Daughters are used instead, however this does not work for samples at non-equilibrium.

### Energy Levels and Comments

<table>
<thead>
<tr>
<th>$E_γ$</th>
<th>$E$(level)</th>
<th>$I_γ$</th>
<th>Mult.</th>
<th>$α$</th>
</tr>
</thead>
<tbody>
<tr>
<td>49.55</td>
<td>49.55</td>
<td>0.064</td>
<td>E2</td>
<td>326.4</td>
</tr>
<tr>
<td>113.5</td>
<td>163.0</td>
<td>0.0102</td>
<td>[E2]</td>
<td>6.63</td>
</tr>
</tbody>
</table>

$γ^{(234}\text{Th)}$

Current method using $^{234}\text{Th}$

- $^{234}\text{Th}$ decay ($^{238}\text{U}$)
  - $92.80(2)$ keV
  - $92.38(1)$ keV
  - $93.35$ keV

- $^{234}\text{Th} Kα1$ ($^{235}\text{U}$)
  - $92.13(20)$% $^{92.80}$ keV
  - $4.7(5)$% $^{93.35}$ keV

### Decay Constants

- $E_γ$: from 1973Ta25 (semi). Other: 1956Al30 ($(α/α)$(ec)). $E_γ=0.059%$ was measured by 1990Ro40.
- Total $l$(ec)=23 3 per 100 $α$ decays ($^{1952}\text{Du12, 1952Zn01, 1956Al30}$).
- Mult.: $α=359 65$ from $I\alpha/I_γ$ and the measured half-life of 0.37 ns 3 for the 49.55-keV level rule out other multipolarities.
- $E_γ$: from 1984Ro21.
New Capability with Microcalorimeters – Direct $^{238}$U Measurement

- Gamma-rays from U decays are populated in the 50 keV region, where can be precisely measured by microcal.
- 50 keV region will provide direct U enrichment analysis as well as minor U isotope compositions.
Uranium Gamma Data Needs Improvement

- 50 keV from $^{238}\text{U}$ can be used for direct measurement of U enrichment.
- $^{236}\text{U}$ interference need to be understood.
- Simple measurement series can significantly improve this decay data.
**Gamma Data Improvement with Yb-169 Calibration Source**

**Yb-169 is the most accurate** calibration source for low energy gamma.

Yb-169 was produced at the LBNL 88” Cyclotron
Summary: Microcalorimeter and Nuclear Data

1) Nuclear Data needs for Microcalorimeter Operations (IAEA NML, very high impact)
   - U/Pu Decay Gamma Intensities & Energies. **Significant impacts** to international nuclear safeguards.
   - U/Pu Half-lives (also needed for chronology)
   - $^{235}$U Alpha Branching Ratios

2) Microcalorimeter for Nuclear Data Improvements
   - Decay data (half-life, $\alpha\beta\gamma$ energies and BRs)
   - Fission yield
   - Reaction cross-section
### Gamma Data Needs for Microcal at the IAEA NML

<table>
<thead>
<tr>
<th>$^{235}\text{U}$ $\rightarrow$ $^{231}\text{Th}$ $\rightarrow$ $^{234}\text{Pa}$</th>
<th>$^{238}\text{U}$ $\rightarrow$ $^{234}\text{Th}$ $\rightarrow$ $^{234}\text{Pa}$ $\rightarrow$ $^{234}\text{U}$</th>
<th>$^{232}\text{U}$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Energy [keV]</strong></td>
<td><strong>Intensity [%]</strong></td>
<td><strong>Energy [keV]</strong></td>
</tr>
<tr>
<td>51.21(5)</td>
<td>0.034(7)</td>
<td>49.55(6)</td>
</tr>
<tr>
<td>54.25(5)</td>
<td>0.015(15)</td>
<td>113.5(1)</td>
</tr>
<tr>
<td>$^{231}\text{Th}$</td>
<td></td>
<td>$^{234}\text{Th}$</td>
</tr>
<tr>
<td>53.2(8)</td>
<td>0.48(?)</td>
<td>92.38(1)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>92.80(2)</td>
</tr>
<tr>
<td>$^{236}\text{U}$</td>
<td></td>
<td>$^{234}\text{U}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Energy [keV]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>53.20(2)</td>
</tr>
<tr>
<td>$^{236}\text{U}$</td>
<td></td>
<td>$^{238}\text{U}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Energy [keV]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>49.46(10)</td>
</tr>
</tbody>
</table>

- Huge uncertainties on intensity (10% - 100%)
Microcalorimeters for 92 keV Region

- $^{234}$Th decay ($^{238}$U)
  - 92.80(2) keV
  - 2.10(20)%

- $^{234}$Th Kα1 ($^{235}$U)
  - 92.38(1) keV
  - 2.13(20)%
  - 93.35 keV
  - 4.7(5)%

U Enrichment measurements will be limited by gamma intensity data (~10% uncertainty)