Nuclear data needs for Plutonium accounting using microcalorimeters

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Pu Accounting During the Nuclear Fuel Cycle

Example: Rokkasho plant: 800,000 kg spent fuel $\rightarrow$ 8000 kg Pu

Safeguards target: account for all Pu to within $\sim$8 kg per year

$\rightarrow$ 0.1 % precision required

Mass spectrometry achieves this but ...

- Time-consuming, labor-intensive ($\sim$6000 measurements / year)
- Destructive

Want non-destructive, fast technique that approaches mass spec precision and accuracy
Two Gamma-Ray Spectroscopy Technologies: HPGe and TES Microcalorimeters

High-Purity Germanium Detectors
- Mature, many commercial solutions available
- High efficiency (energies up to 1+ MeV)
- Energy Resolution: Limited to ~500 eV at 100 keV

Low-Temperature TES Microcalorimeters
- Emerging technology, not yet commercial
- Good efficiency at 100 keV, falls off above 250 keV
- Excellent energy resolution: <= 80 eV at 100 keV
  → Reduces systematic errors due to peak overlaps

Irwin & Hilton, “Transition-Edge Sensors” in Cryogenic Particle Detection, Springer-Verlag, 2005
Low-Temperature Microcalorimeters: Opportunities and Challenges

- **Efficiency** comparable to energy dispersive detectors
- **Energy resolution** approaches many wavelength-dispersive instruments
- **Broadband** detection allows measurement of many spectral lines simultaneously

\[ \Delta E \sim \sqrt{4k_B T^2 C} \]

\[ \frac{E}{\Delta E} > 1000 \text{ from 1 keV to 5 MeV} \]

To take full advantage of new measurement capabilities requires better reference data!
Deployed and Planned Microcalorimeter Instruments

2019 - present: SOFIA spectrometer
- 256 pixels, portable
- Commissioned at LANL in 2019
- Measurements at PF-4
- Currently at ORNL

2022 - present: INL Spectrometer
- 384 pixels
- Delivered Aug 2022

Planned Instruments in FY23 & FY24
- 2 SOFIA-style instruments (IAEA, PF-4)
- 672-pixel system for PNNL
To measure Isotopic Composition ...
• Measure relative peak areas
• Account for detector efficiency
• Account for Branching Ratios
  → Fraction of decays that produce a particular gamma-ray energy

Plutonium Isotopic Analysis via Gamma-Ray Spectroscopy

Contributions to Uncertainty Budget

Key results of Hoover-2014
- Microcal results are dominated by uncertainties in BRs (~ 1 % limit)
- Can reduce total uncertainty to “tenths-of-percent level” for microcalorimeter isotopic analysis through better BR uncertainties
- HPGe also limited by BRs, but line energies are equally important

<table>
<thead>
<tr>
<th>Uncertainty Source</th>
<th>Microcal (%)</th>
<th>HPGe (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BRs</td>
<td>0.82 - 1.24</td>
<td></td>
</tr>
<tr>
<td>Half Life</td>
<td>0.13 – 0.19</td>
<td></td>
</tr>
<tr>
<td>Line Energies</td>
<td>0.02 – 0.15</td>
<td>0.18 – 3.1</td>
</tr>
<tr>
<td>X-Ray Linewidths</td>
<td>0.01 – 0.65</td>
<td>0.16 – 0.97</td>
</tr>
</tbody>
</table>

What Gamma Lines are We Talking About?

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy (keV)</th>
<th>Relative BR</th>
<th>BR Uncert (%)</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Am-241</td>
<td>98.97</td>
<td>1.0</td>
<td>2.0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>102.966</td>
<td>0.96</td>
<td>2.1</td>
<td></td>
</tr>
<tr>
<td></td>
<td>125.3</td>
<td>0.21</td>
<td>2.2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>146.55</td>
<td>0.02</td>
<td>2.4</td>
<td></td>
</tr>
<tr>
<td></td>
<td>164.61</td>
<td>0.003</td>
<td>3.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>169.56</td>
<td>0.009</td>
<td>2.3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>208.005</td>
<td>0.04</td>
<td>2.2</td>
<td></td>
</tr>
<tr>
<td>Pu-241</td>
<td>102.966</td>
<td>&lt; 0.001</td>
<td>14.1</td>
<td></td>
</tr>
<tr>
<td></td>
<td>103.68</td>
<td>0.20</td>
<td>0.5</td>
<td>[1]</td>
</tr>
<tr>
<td></td>
<td>148.567</td>
<td>0.36</td>
<td>0.66</td>
<td></td>
</tr>
<tr>
<td></td>
<td>159.96</td>
<td>0.013</td>
<td>1.1</td>
<td>[2]</td>
</tr>
<tr>
<td></td>
<td>164.610</td>
<td>0.09</td>
<td>1.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>208.005</td>
<td>1.0</td>
<td>1.4</td>
<td></td>
</tr>
</tbody>
</table>

[1]: Only 50 eV away from Pu Ka1
[2]: Not currently used, but would like to
[3]: Not currently used, but “anchor” BR (see later slide)
[4]: We have evidence the tabulated BR is wrong (see later slide)
**BRs for Np X-Ray Lines Might be Useful**

- Np x-rays are produced directly by decay of Am-241, and indirectly by decay of Pu-241.
- Some HPGe codes have used Np Ka1, but microcal currently uses no x-rays.

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<tr>
<td>Am-241</td>
<td>97.069</td>
<td>0.06</td>
<td>3.5</td>
<td>Np-Ka1</td>
</tr>
<tr>
<td></td>
<td>101.0563</td>
<td>0.09</td>
<td>3.3</td>
<td></td>
</tr>
<tr>
<td>Pu-241</td>
<td>97.069</td>
<td>0.73</td>
<td>2.6</td>
<td>Np-Ka1</td>
</tr>
<tr>
<td></td>
<td>101.0563</td>
<td>1.16</td>
<td>2.4</td>
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Am-241

- β⁻: 14.329 y
- α: 14.290 y
- α: 432.6 y
- β⁻: 6.75 d
Use well-characterized materials with known isotopic ratios to infer BRs

→ 2020 Paper by Yoho showing improvements in multiple branching ratios above the Pu K edge (121.8 keV)

We are also pursuing an analysis that spans the Pu K edge.

• Ex: New BR for $^{239}$Pu 98.78 keV
• Value 13% lower than ENSDF, with uncertainty 3x smaller

Yoho, M.D. et al, “Improved plutonium and americium photon branching ratios from microcalorimeter gamma spectroscopy”, NIM A (977) 2020

DOI: 10.1016/j.nima.2020.164307
Relative vs Absolute Branching Ratios

• Absolute measurement of BR challenging with microcal (quantum efficiency ~ 45%)
• Thus Yoho-2020 measured BRs relative to a set of “anchor” BRs
• Improvement in these would aid microcalorimeter determination of BRs

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<th>Energy (keV)</th>
<th>BR (%)</th>
<th>BR Uncert (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-239</td>
<td>129.3</td>
<td>6.31e-3</td>
<td>0.63</td>
</tr>
<tr>
<td>Pu-241</td>
<td>148.57</td>
<td>1.8975e-4</td>
<td>0.66</td>
</tr>
<tr>
<td>Pu-240</td>
<td>160.3</td>
<td>4.02e-4</td>
<td>1.00</td>
</tr>
<tr>
<td>Pu-239</td>
<td>195.68</td>
<td>1.07e-4</td>
<td>0.93</td>
</tr>
<tr>
<td>Pu-239</td>
<td>203.55</td>
<td>5.69e-4</td>
<td>0.53</td>
</tr>
</tbody>
</table>

Anchor BRs from Yoho 2020

New, larger data sets acquired in 2021 will allow reduction in uncertainty due to counting statistics
• Branching Ratios uncertainties limit isotopic analysis using gamma spectroscopy to ~ 1% accuracy

• We’ve identified top candidates where better values would be useful

• Highest priority are absolute measurements of the “anchor” BRs
Determining Pu Composition

\[ A_j = \sum_k I_k BR_{jk} \eta(E_j) \]

find best fit of 9 parameters over 15 equations

\[ A_j = \text{area of peak with energy } E_j \]

\[ I_k = \text{decay intensity of isotope } k \]

\[ BR_{jk} = \text{branching ratio for isotope } k \text{ to energy } E_j \]

\[ \eta(E_j) = \text{source-detector efficiency at energy } E_j \]


**TES Spectrometer Architecture**

- **TES microcalorimeter pixel**
- **Commercial Refrigerator:** Cryogen-free, reliable
- **TES microcalorimeter array subassembly**
- **Readout:** custom IF boards

**Readout:**
- Cryogenic Microwave SQUID Multiplexer circuit
- Commercial FPGA + DAC/ADC boards

Detectors view samples through a window in the cryostat. In this case the window faces down, but sideways-facing windows are also possible.