Direct Detection of Pu-242 with an MMC Gamma-Ray Detector

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Nuclear Safeguards: Pu-242 in Spent Fuel

Large reprocessing facilities (e.g. Rokkasho, Japan)
• 800 tons of spent fuel/year, ~8 tons of Pu/year in solution
• 1% error is 80 kg/year ⇒ 10 significant quantities of Pu

Accurate quantification of fissile material is essential.
Plutonium Isotope Analysis in Nuclear Safeguards

Typical Safeguards Measurements

1) Measure total neutron flux (even Pu isotopes) with neutron coincident counter
2) Quantify Pu isotope ratios (Pu-238 to Pu-242) with γ-spectroscopy or mass spectrometry
   ⇒ Infer amount of fissile Pu-239

• Pu-242 is currently determined through correlations (imprecise) or DA (costly)
• Error in Pu-242 sets error in total Pu, especially for high burn-up samples.

Pu-242 cannot be directly measured with Ge ⇒ Costly destructive analysis (DA) required.
Metallic Magnetic Calorimeters (MMCs)

MMC Operating Principle

Photon

\[ \mu B \geq kT \]

\( \{ \text{Au:Er} \} \)

MMC

SQUID

\( V_{out} \)

Ideal MMC:

\[ \Delta E_{\text{FWHM}} \approx 2.355 \sqrt{\frac{4kT^2C \cdot 2 \tau_{\text{rise}}}{\tau_{\text{decay}}}} \]

Real MMC:

- SQUID noise
- Er 1/f noise
- Magnetic Johnson noise

No resistive elements, no quiescent power dissipation \( \Rightarrow \) High resolution, scaling to arrays.
MMC Gamma Detectors

**New Geometry:**
Absorber on Au posts as bottlenecks

1) Fabricate MMC+wiring
2) AZ 125 nXT mold
3) Electroplate 150µm Au
4) Remove mold

Au posts reduce phonon loss into the substrate and improve resolution and dynamic range.
MMC Detector Tests at Heidelberg at 15 mK

- Detector: Heidelberg MaXs-200 MMC with posts
- Setup: 15 mK dilution refrigerator, pinhole collimator
- But: Still only 50% of optimum B-field

Energy resolution of 46 eV at 15 mK resolves details in Np X-ray lines.
MMC Detector Tests at LLNL at 35 mK

- Detector: Heidelberg MaXs-200 MMC with posts
- Setup: 35 mK in adiabatic demagnetization refrigerator
- Optimum B-field

Lower resolution in agreement with expected and measured noise at 35 mK.
Linearity and Reproducibility

Spectra can be added without loss in energy resolution.

- 2 pixels very similar
- Very reproducible
- Linear to ± a few eV at 35 mK

Source: WG Pu + $^{242}$Pu
Mixed Isotope Plutonium: MMC vs. Ge

Source: WG Pu + $^{242}$Pu

MMC enables direct detection of Pu-242.

Counts

Energy [keV]

10 $^6$

10 $^5$

10 $^4$

10 $^3$

10 $^2$

20 40 60 80 100 120

Energy [keV]

10 $^8$

10 $^7$

10 $^6$

10 $^5$

10 $^4$

10 $^3$

10 $^2$

20 40 60 80 100 120

Counts

Energy [keV]

10 $^6$

10 $^5$

10 $^4$

10 $^3$

10 $^2$

20 40 60 80 100 120

Energy [keV]
Non-Destructive Assay (NDA) of Pu Sample

\[
\frac{\text{Isotope}_1}{\text{Isotope}_2} = \frac{\text{Peak}_1 \times \text{Efficiency}_2 \times \text{Half Life}_1 \times \text{Br.Ratio}_2}{\text{Peak}_2 \times \text{Efficiency}_1 \times \text{Half Life}_2 \times \text{Br.Ratio}_1}
\]

Source of Error in NDA:
- Statistical errors, background subtraction
- **Relative detection efficiency**
- Nuclear data ($\tau_{1/2}$, branching ratio)

Efficiency = Self-absorption (in Pu source) 
\[\times \text{ Al holder / filter transmission}
\times \text{ Au MMC efficiency}
\]

\[
= \frac{1-e^{-\mu_{\text{Pu}} d_{\text{Pu}}}}{\mu_{\text{Pu}} d_{\text{Pu}}} \times e^{-\mu_{\text{Al}} d_{\text{Al}}} \times \left(1-e^{-\mu_{\text{Au}} d_{\text{Au}}} \right)
\]

NDA codes need to be updated for MMCs.
Error Analysis

Pu Isotope Concentration

<table>
<thead>
<tr>
<th>Isotope</th>
<th>DA (Mass spec.)</th>
<th>NDA (MMCs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-239</td>
<td>83.75 ± 0.13</td>
<td>83.67 ± 0.42</td>
</tr>
<tr>
<td>Pu-240</td>
<td>5.39 ± 0.01</td>
<td>5.22 ± 0.11</td>
</tr>
<tr>
<td>Pu-242</td>
<td>10.81 ± 0.02</td>
<td>11.11 ± 0.42</td>
</tr>
</tbody>
</table>

Relative error for Pu-242 of ±3.8 % exceeds accuracy of correlation functions with Ge detectors, but cannot compete with mass spec.

Error Contributions

<table>
<thead>
<tr>
<th>Error Source</th>
<th>Magnitude [% RSD]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-242 peak area</td>
<td>3.28 %</td>
</tr>
<tr>
<td>Pu-242 br. ratio</td>
<td>1.67 %</td>
</tr>
<tr>
<td>Pu-239 br. ratio</td>
<td>0.68 %</td>
</tr>
<tr>
<td>Pu-242 $\tau_{1/2}$</td>
<td>0.47 %</td>
</tr>
<tr>
<td>Pu-242 efficiency</td>
<td>0.25 %</td>
</tr>
<tr>
<td>Pu-239 efficiency</td>
<td>0.23 %</td>
</tr>
<tr>
<td>Pu-239 peak area</td>
<td>0.16 %</td>
</tr>
<tr>
<td>Pu-240 br. ratio</td>
<td>0.11 %</td>
</tr>
<tr>
<td>All other</td>
<td>&lt;0.10 %</td>
</tr>
<tr>
<td><strong>Total:</strong></td>
<td><strong>3.8 %</strong></td>
</tr>
</tbody>
</table>

Precision still limited by counting statistics ⇒ Can be improved with arrays.
MMC Gamma Detector Summary

Au:Er MMC γ-Detectors with posts:
- 46 eV FWHM at 15 mK, 91 eV at 35 mK
- Very linear and reproducible response

Non-Destructive Isotope Analysis:
- Direct Detection of Pu-242
- ~4% accuracy with 24 detector-days
- Limited by statistics ⇒ Arrays