Nanocomposite Absorbers, Energy Thermalization, and Nuclear Decay Energy Spectroscopy

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A new category of measurements

- Radioactive material inside absorber
- Energy of all relevant decay products captured in single pulse
- Cryogenic microcalorimeters provide excellent energy resolution

Other groups working in this field:
- CEA
- KRISS
- KIP / Heidelberg
- INFN Genova, Milano-Bicocca

Path length of prominent actinide (Pu,Am) decay radiation in a gold absorber
Alpha decay: measure Q value

Q value

240Pu

236U

α particles

Relative mass – energy (MeV)

Counts per bin

Energy (keV)

Np-237
Pu-242
Pu-241
Pu-239
Pu-238
Pu-240
Pa-231
Th-229
Pu-240
U-232
Am-243
Th-228
Am-241
Pu-239
Pu-240
Pu-239
Pu-240
5000
5100
5200
5300
5400
5500
5600

Mark Croce – Los Alamos National Laboratory – LTD-16 – Slide 3
Beta decay: measure Q minus neutrino energy

\[ ^{241}\text{Pu} \rightarrow ^{241}\text{Am} + e + \overline{\nu}_e \]

\[ ^{163}\text{Ho} \rightarrow ^{163}\text{Dy} + \nu_e + \{\gamma, e, \text{phonons}\} \]
Transition-edge sensors

Microcalorimeter optimized for MeV energies

Microcalorimeter optimized for keV energies
A very simple deposition: dried drop of solution

013114-3 ($^{239}$Pu, $^{240}$Pu)

020414-4 ($^{239}$Pu, $^{240}$Pu)

Plutonium in 3M nitric acid, dried on gold foils, forms Pu nitrate crystals
Q-spectra from dried $^{239,240}$Pu nitrate: split peaks
As deposited

Split peaks, most counts in lower energy component

Plutonium nitrate as deposited from 3M nitric acid
Fracture crystals with 3 kneading cycles

Counts shift to higher energy component

(b)

Counts / 200 eV bin

Energy (keV)

5210 5220 5230 5240 5250 5260 5270

239 Pu

240 Pu
Fracture crystals with 6 kneading cycles

Peak split significantly reduced
Fracture crystals with 100 kneading cycles

Peak splitting and low energy tailing nearly eliminated

Lanthanum nitrate, stable surrogate for TEM

1.02 keV FWHM (Gaussian component of Bortels function fit)
Nanocomposite absorbers from simple mechanical alloying

Optimal energy thermalization achieved when radioactive material is distributed in grains that are small compared to the relevant radiation path lengths.
$^{240}\text{Pu}/^{239}\text{Pu}$ Quantitative Analysis

- See poster by Croce et al. for details
- Well-separated peaks resulting from optimized absorber structure enable accurate isotopic analysis

Pu Chemical Age

- See poster by Rabin et al. for details
- Simultaneously measure $^{241}$Pu beta and $^{241}$Am Q
- Ingrowth of $^{241}$Am daughter over time provides an “atomic clock” to determine time since purification using $^{241}$Am:$^{241}$Pu ratio
Particles

- First steps toward particle measurements with very simple sample preparation
- Collected Pu oxide material directly with gold foil from surface of CRM126A reference material
Particles

![Graph showing particle energy distributions for different isotopes.](image)

- $^{239}\text{Pu}$
- $^{240}\text{Pu}$
- $^{238}\text{Pu}$
- $^{241}\text{Am}$

Counts / 0.5 keV bin

Counts / 0.2 keV bin

Energy (keV)
Engineered nanomaterials

- Au nanofoam made by dealloying Ag from Au/Ag alloy
- Solutions are absorbed into nanofoam and deposits dry as nano-constrained crystals
Pu nitrate dried in Au nanofoam

- ~50-100 nm pores
- No split peaks, but significant low-energy tailing
- Need to reduce pore size
$^{163}$Ho electron capture spectroscopy for neutrino mass measurement

See talk by Kunde et al. for details

Unique challenges:
• Make and purify $^{163}$Ho
• Low energy decay: $Q < 3$ keV
$^{163}$Ho product solution, dried deposit

- This is highly purified material already: irradiated Dy target contains only about 1 ppm $^{163}$Ho
- But it’s still not pure $^{163}$Ho: major contaminants are organics, Na, Cl
- Need to develop methods to purify Ho, embed in absorber matrix
Hydrogen reduction/annealing of $^{\text{nat}}\text{HoCl}_3$ on Au foil

- Inspired by procedure from Barclay JLTP 1978
- 800°C in hydrogen atmosphere reduces HoCl$_3$
- Briefly raise temperature to 1100°C to melt Au, form Au:Ho alloy
- Oxidation problem: Ho ends up as oxide flakes on Au surface
163\(^\text{Ho}\) structure and composition tests

*Strong effect on spectral quality*

<table>
<thead>
<tr>
<th></th>
<th>Au foil absorber</th>
<th>Nanoporous Au absorber</th>
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</thead>
<tbody>
<tr>
<td><strong>As deposited</strong></td>
<td><img src="image1" alt="Graph for Au foil absorber" /></td>
<td><img src="image2" alt="Graph for Nanoporous Au absorber" /></td>
</tr>
<tr>
<td>Na, Cl, impurities</td>
<td><img src="image3" alt="Image for Au foil absorber" /></td>
<td><img src="image4" alt="Image for Nanoporous Au absorber" /></td>
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<tr>
<td><strong>Hydrogen annealed</strong></td>
<td><img src="image5" alt="Graph for Au foil absorber" /></td>
<td><img src="image6" alt="Graph for Nanoporous Au absorber" /></td>
</tr>
<tr>
<td>Na, Cl removed</td>
<td><img src="image7" alt="Image for Au foil absorber" /></td>
<td><img src="image8" alt="Image for Nanoporous Au absorber" /></td>
</tr>
</tbody>
</table>
Pd matrix: more reactive than Au

*Not yet tested as a microcalorimeter absorber*

Clusters of sub-20 nm Pd nanoparticles from combustion of $C_4H_{10}N_{20}Pd$ (PdBTA)

TEM from Tappan et al. 2010
Summary

- Nanocomposite absorbers, made with simple techniques, enable high resolution Q spectroscopy of alpha decaying isotopes
- Ongoing work to optimize absorber structure for other isotopes, includes modeling

See also: Croce poster, Rabin poster, Kunde talk