Nuclear data needs for Plutonium accounting using microcalorimeters

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Pu Accounting During the Nuclear Fuel Cycle

Rokkasho

Savannah River H-Canyon

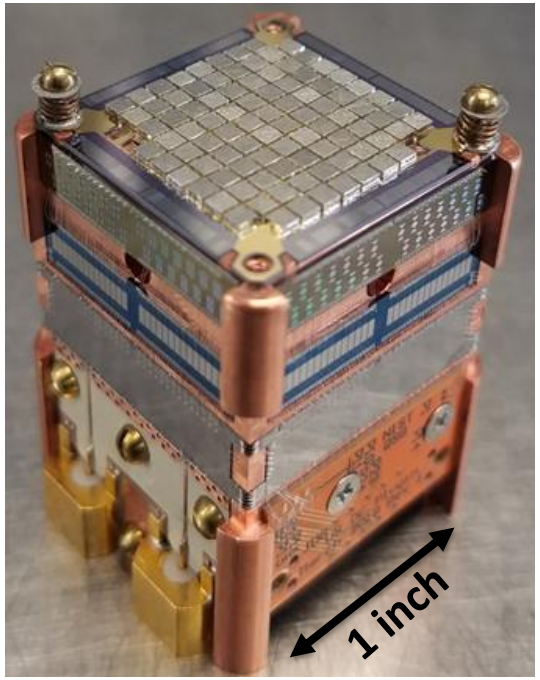
- Example: Rokkasho plant: 800,000 kg spent fuel → 8000 kg Pu
- Safeguards target: account for all Pu to within ~8 kg per year

  → 0.1 % precision required
- Mass spectrometry achieves this but …
  - Time-consuming, labor-intensive (~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



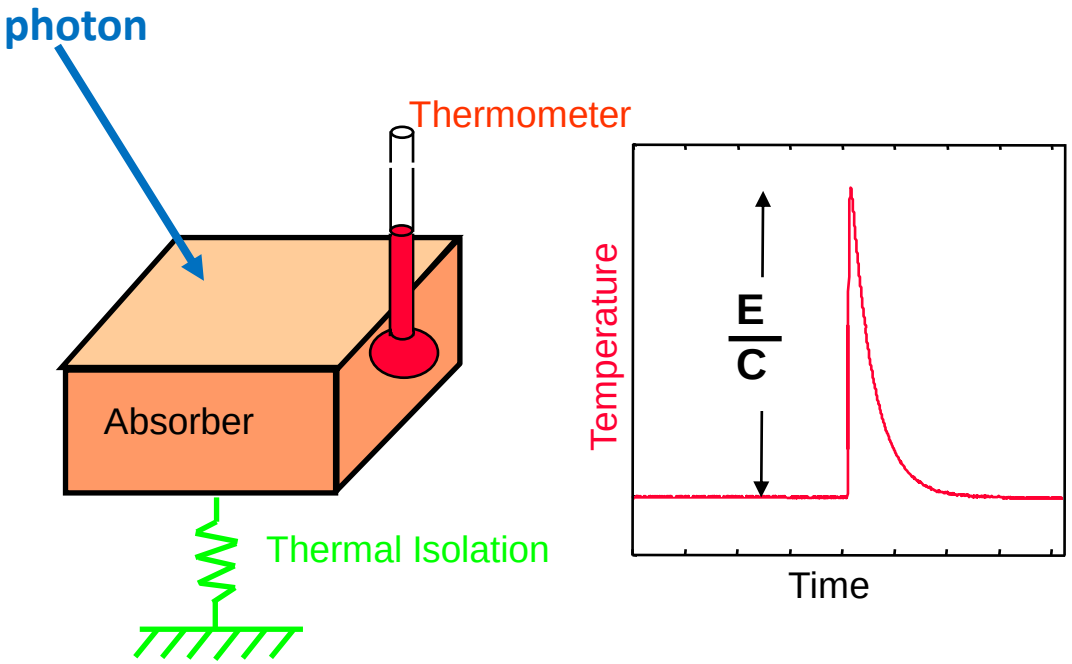
Low-Temperature TES Microcalorimeters

- 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



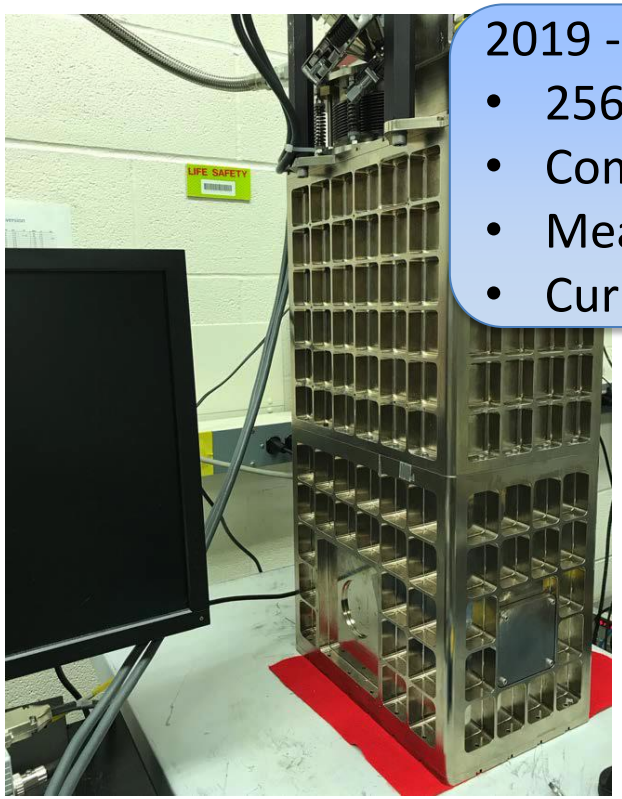
- **Efficiency** comparable to energy dispersive detectors
- **Energy resolution** approaches many wavelength-dispersive instruments
- **Broadband** detection allows measurement of many spectral lines simultaneously

$$\Delta E \sim \sqrt{4\, k_B T^2 C}$$

E/ΔE > 1000 from 1 keV to 5 MeV

To take full advantage of new measurement capabilities requires better reference data!

Deployed and Planned Microcalorimeter Instruments

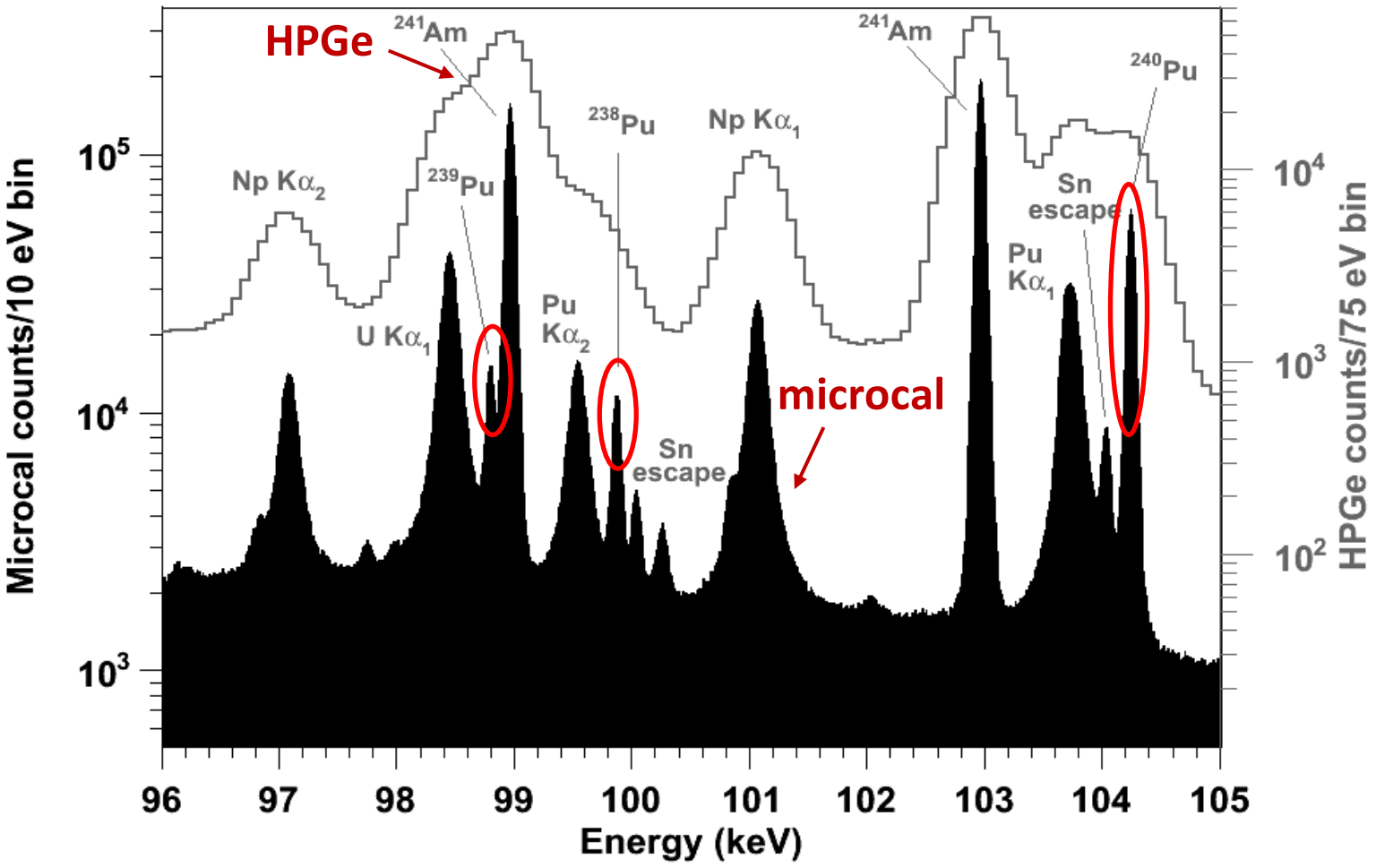


2019 - present: SOFIA spectrometer
- 256 pixels, portable
- Commissioned at LANL in 2019
- Measurements at PF-4
- Currently at ORNL

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

Plutonium Isotopic Analysis via Gamma-Ray Spectroscopy



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

Hoover et. al., IEEE Trans. Nuc. Sci. 60 (2013)

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

Uncertainty Source	Microcal (%)	HPGe (%)
BRs	**0.82 - 1.24**	
Half Life	0.13 – 0.19	
Line Energies	0.02 – 0.15	**0.18 – 3.1**
X-Ray Linewidths	0.01 – 0.65	**0.16 – 0.97**

Hoover, A.S., et al., "Uncertainty of Plutonium Isotopic Measurements with Microcalorimeter and High-Purity Germanium Detectors", IEEE-TNS, 2014

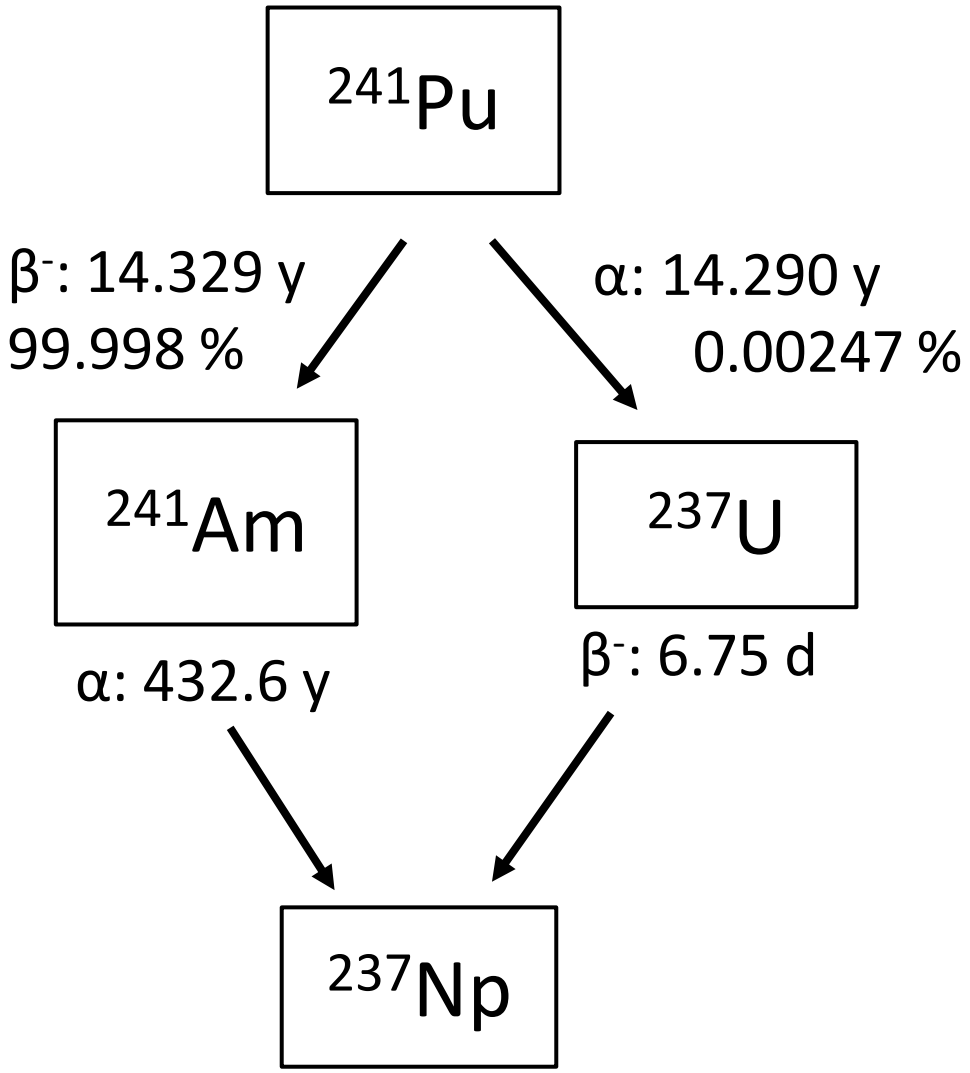
What Gamma Lines are We Talking About?

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

Isotope	Energy (keV)	Relative BR	BR Uncert (%)	Notes
Pu-238	99.853	1.0	1.1	
	152.72	0.13	0.75	
Pu-239	98.78	0.2	4.6	[2] [4]
	115.4	0.07	10.8	[2]
	116.3	0.09	1.9	[2]
	129.296	1.0	0.63	
	195.68	1.07e-4	0.93	[3]
	203.550	0.09	0.53	
Pu-240	104.234	1.0	1.3	
	160.308	0.06	1.0	

[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



Isotope	Energy (keV)	Relative BR	BR Uncert (%)	Notes
Am-241	97.069	0.06	3.5	Np-Ka1
	101.0563	0.09	3.3	Np-Ka1
Pu-241	97.069	0.73	2.6	Np-Ka1
	101.0563	1.16	2.4	Np-Ka2

- 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

Yoho 2020: Better Branching Ratios From Microcal Spectra

Table 6
Comparison of NNDC [17] branching ratios (BRs) to those of this work. ^{241}Pu BRs at 164 keV and 208 keV assume secular equilibrium with ^{237}U. Relative % uncertainties (μ) represent 67% confidence intervals. BR units are in γ/decay × 100.

Energy [keV]	Isotope	NNDC BR	μ_{BR} [%]	This work BR	μ_{BR} [%]	μ_{BR} Agreement
125.21	^{239}Pu	5.63×10^{-5}	2.7	5.51×10^{-5}	13	−0.2
125.3	^{241}Am	4.08×10^{-3}	2.5	4.08×10^{-3}	1.0	0.0
144.201	^{239}Pu	2.83×10^{-4}	2.1	2.87×10^{-4}	1.0	0.6
146.094	^{239}Pu	1.19×10^{-4}	2.5	1.22×10^{-4}	1.4	0.7
146.55	^{241}Am	4.61×10^{-4}	2.6	4.75×10^{-4}	0.75	1.2
150.04	^{241}Am	7.40×10^{-5}	3.0	7.76×10^{-5}	1.3	1.5
152.72	^{238}Pu	9.29×10^{-4}	0.75	9.46×10^{-4}	0.78	1.7
159.955	^{241}Pu	6.68×10^{-6}	1.1	6.87×10^{-6}	2.0	1.2
160.19	^{239}Pu	6.20×10^{-6}	19	5.82×10^{-7}	331	−2.5
161.45	^{239}Pu	1.23×10^{-4}	1.6	1.20×10^{-4}	1.6	−1.1
161.54	^{241}Am	1.50×10^{-6}	20.0	3.52×10^{-6}	19.9	2.7
164.61	^{241}Pu	4.56×10^{-5}	1.6	4.46×10^{-5}	2.0	−0.9
164.69	^{241}Am	6.67×10^{-5}	3.7	7.78×10^{-5}	4.9	2.4
169.56	^{241}Am	1.73×10^{-4}	2.3	1.72×10^{-4}	0.9	−0.3
171.393	^{239}Pu	1.10×10^{-4}	1.8	1.12×10^{-4}	1.4	0.9
175.07	^{241}Am	1.82×10^{-5}	5.5	1.85×10^{-5}	2.8	0.3
188.23	^{239}Pu	1.09×10^{-5}	10	8.63×10^{-6}	10.8	−1.6
189.36	^{239}Pu	8.30×10^{-5}	1.2	7.91×10^{-5}	1.4	−2.6
191.96	^{241}Am	2.16×10^{-5}	4.6	2.01×10^{-5}	2.8	−1.3
208.005	^{241}Pu	5.19×10^{-4}	1.4	5.34×10^{-4}	1.9	1.2
208.01	^{241}Am	7.91×10^{-4}	2.4	8.08×10^{-4}	5.4	0.4

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.

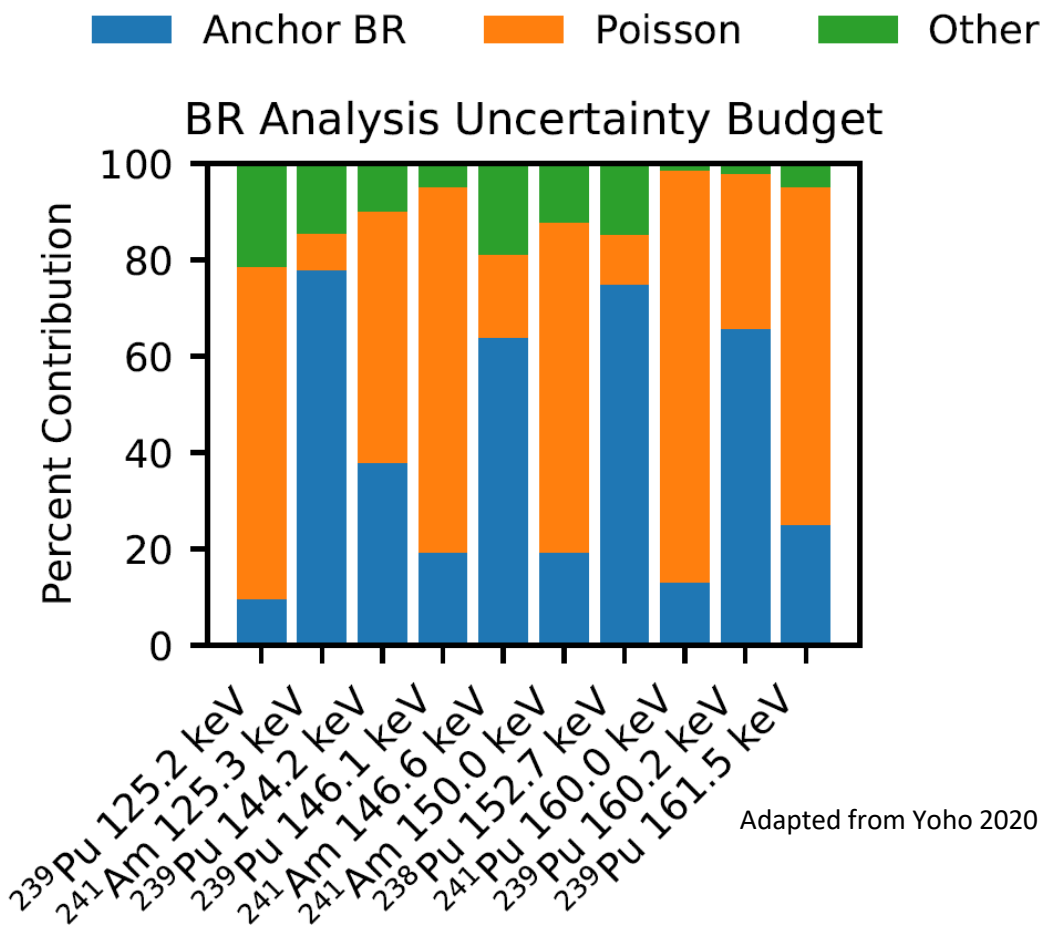
- Ex: New BR for ^{239}Pu 98.78 keV
- Value 13% lower than ENSDF, with uncertainty 3x smaller

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



Adapted from Yoho 2020

Anchor BRs from Yoho 2020

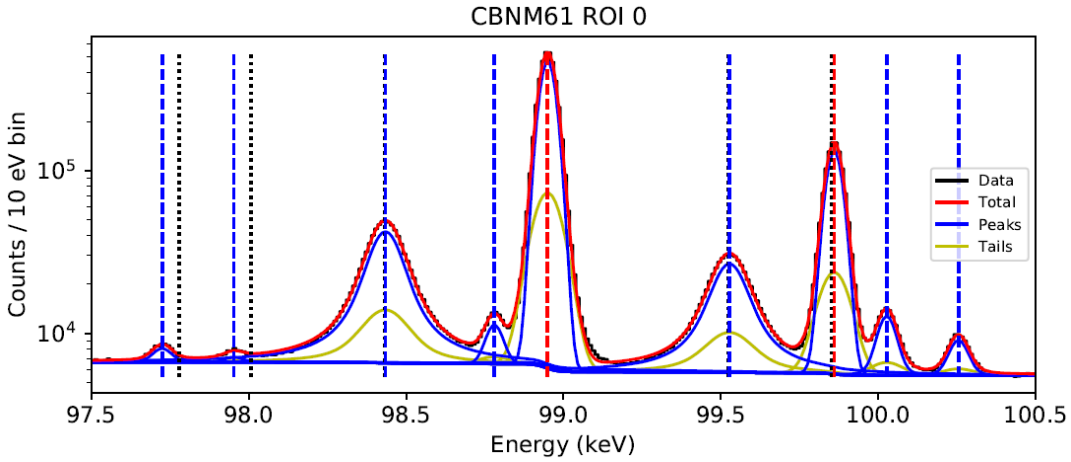
Isotope	Energy (keV)	BR (%)	BR Uncert (%)
Pu-239	129.3	6.31e-3	0.63
Pu-241	148.57	1.8975e-4	0.66
Pu-240	160.3	4.02e-4	1.00
Pu-239	195.68	1.07e-4	0.93
Pu-239	203.55	5.69e-4	0.53

New, larger data sets acquired in 2021 will allow reduction in uncertainty due to counting statistics

Summary – Gamma Spectroscopy

- 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



Regions of Interest

Peak Areas

Fit Using Efficiency Model

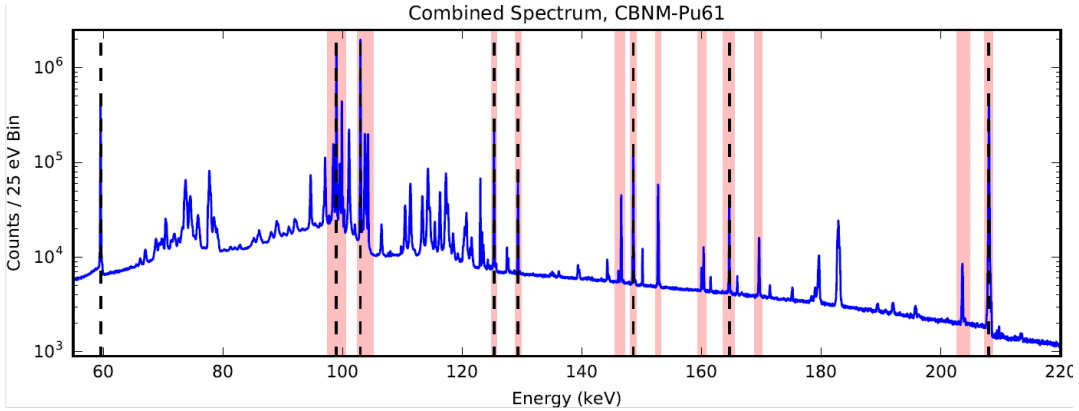
Efficiency Curve

Isotopic Ratios

Branching Ratios

Other Nuclear Data

Detector Efficiency Model



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

find best fit of 9 parameters over 15 equations

A_j = area of peak with energy E_j

I_k = decay intensity of isotope k

BR_{jk} = branching ratio for isotope k to energy E_j

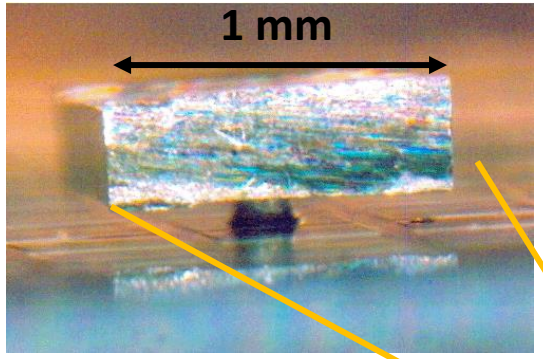
$\eta(E_j)$ = source-detector efficiency at energy E_j

Hoover et. al. "Determination of Plutonium Isotopic Content by Microcalorimeter Gamma-Ray Spectroscopy", *IEEE Trans. Nucl. Sci.* 60 (2013) http://dx.doi.org/10.1109/TNS.2013.2249091

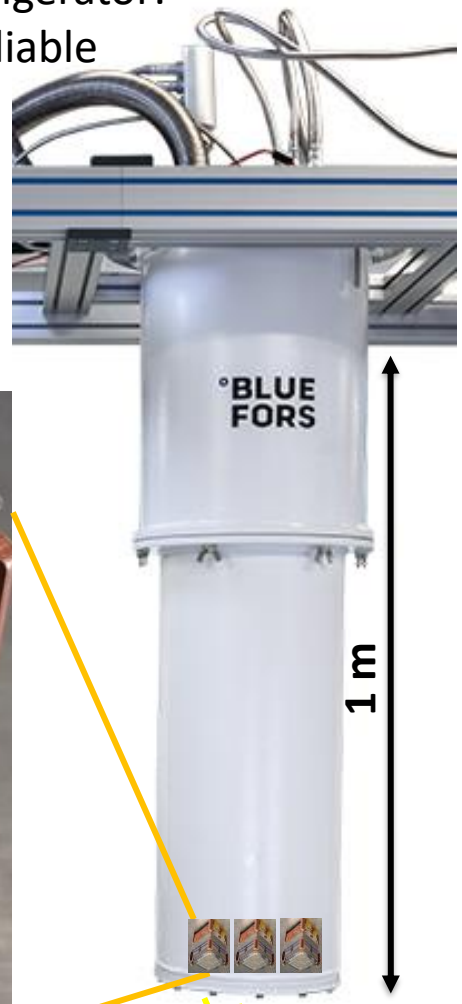
Becker et. al. "Advances in Analysis of Microcalorimeter Gamma-Ray Spectra", *IEEE Trans. Nucl. Sci.* 66 (2019) http://dx.doi.org/10.1109/TNS.2019.2953650

TES Spectrometer Architecture

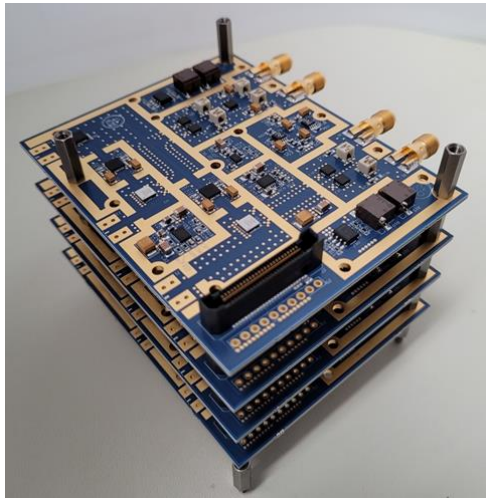
TES microcalorimeter pixel



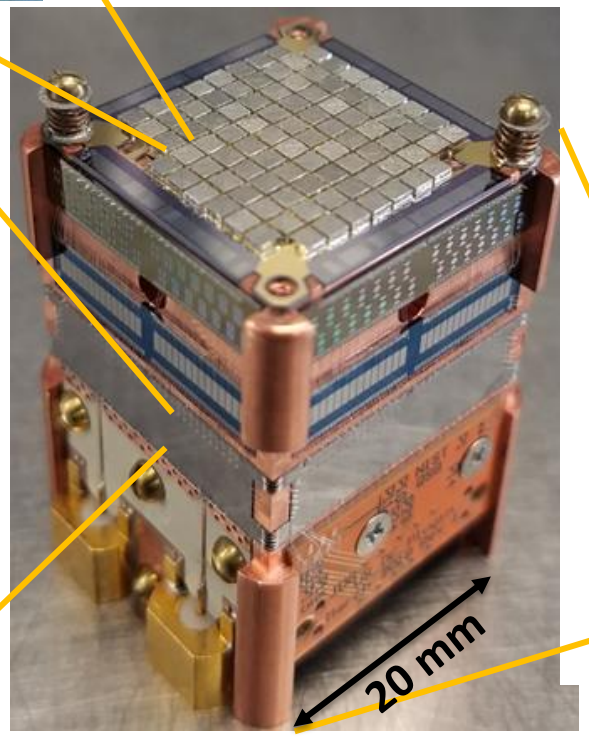
Commercial Refrigerator:
Cryogen-free, reliable

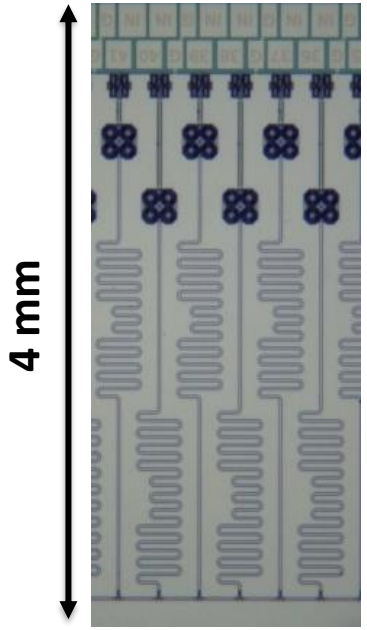


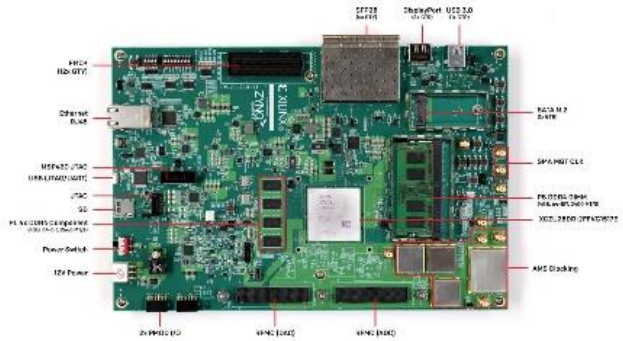
Readout: custom IF boards



TES microcalorimeter array subassembly







Readout: commercial FPGA + DAC/ADC boards

Readout: cryogenic Microwave SQUID Multiplexer circuit

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