Quantification of 242 Pu with a Microcalorimeter Gamma Spectrometer

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Abstract

We report observation of the 103-keV and 159-keV gamma ray signatures of 242 Pu using the SOFIA microcalorimeter gamma-ray spectrometer. This is the first observation of these gamma rays in a non-destructive measurement of an unprepared sample, and so represents an important advance in nuclear material accountancy. We extract the gamma emission probabilities at these two energies and compare with prior destructive analysis results. We also confirm that several public databases include an order-of-magnitude error in the emission probabilities at 103 keV, and report an improved centroid energy for this peak.

1 Introduction

Plutonium-242 is a challenge for nondestructive assay (NDA) because of its low specific activity and low gamma emission probability per decay. Neutron multiplicity methods are suitable for quantification but can only be used after the isotopic ratios have been determined [1]. For these ratios, one must depend on destructive measurements, correlation estimates (which can be inaccurate for high-burnup samples) [2], or direct measurement of the gamma ray signatures. The latter has never been achieved successfully except with carefully prepared, thin, unshielded laboratory samples. The only direct gamma rays from ^{242}Pu are at 45, 103, and 159 keV, and all are weak. The lowest energy line is of limited use because of potential attenuation within the sample and/or container. The two higher-energy lines are masked by gamma rays from $^{238-241}$Pu, ^{241}Am, and/or fluorescent X-rays, making them all but impossible to measure with high-purity germanium (HPGe) detectors except under very special circumstances.

Microcalorimetry (μCal) [3, 4, 5, 6] offers a potential method for NDA of ^{242}Pu. The energy resolution of μCal detectors is extraordinary, with a 70 eV full-width at half maximum (FWHM) now routinely achieved. Although efficiency is lower, the resolution allows for direct observation of the 103 and 159 keV signatures. In this demonstration, we use these signatures to quantify the gamma emission probability per decay of the 103-keV γ ray from five standards with no special sample preparation.

1.1 Prior Gamma Measurements of ^{242}Pu

In 1986, Vaninbroukx et al. [7] measured the γ emission probabilities for the ^{242}Pu using HPGe detectors with a FWHM resolution of 495-520 eV at 122 keV, which is near the best achievable for HPGe. Specially-prepared ultra-pure samples (99.85 Wt% ^{242}Pu) were mesured in a configuration designed for negligible photon attenuation. The ^{242}Pu peaks were not resolvable from interfering components from ^{241}Am, ^{241}Pu, and ^{240}Pu decay, whose contributions were computed and subtracted.

Table 1: γ-emission probabilities for ^{242}Pu lines. The top four references are the most recent measurements. The bottom seven are from publicly-available databases in rough order of their literature cutoff dates.

Reference	**103 keV**	**159 keV**
Schmorak 1972 [11]	8.1(9)E-5	5(2)E-6
Vaninbroukx 1986 [7]	2.63(9)E-5	2.98(20)E-6
Berlizov 2011 [8]	—	2.20(8)E-6
Wang 2012 [9]	2.79(8)E-5	2.25(8)E-6
Present Work	2.70(3)E-5	1.80(7)E-6
TORI[12]	7.80(80)E-5	4.50(150)E-6
LNBH[13]	2.53(12)E-5	2.98(20)E-6
ENDF/B-VIII[14]	2.55(10)E-5	3.00(20)E-6
ENSDF[15]	2.53(12)E-4*	2.20(8)E-6
JEFF-3.3[16]	2.55(10)E-5	3.00(20)E-6
NuDat 3.0[17]	2.53(12)E-4*	2.20(8)E-6
JENDL-5[18]	2.53(12)E-4*	2.20(8)E-6

* Reported values may include a transcription error.

In 2011, Berlizov et al. [8] performed measurements on a 99.7 Wt% ^{242}Pu sample with an HPGe detector in response to a suspected discrepancy. Their value for the γ_{159} emission probability was 35% smaller than Vaninbroukx et al.'s. No attempt was made to use the 45 and 103 keV lines. They considered the 159 keV line as "the only practical alternative" for quantitative analysis of ^{242}Pu, but no practical method was developed.

In 2012, Wang made measurements on a 99.97 Wt% ^{242}Pu sample evaporated only a thin foil [9]. An HPGe detector was used in coincidence with a Si(Sb) alpha detector; the α-γ coincidence mode allowed for a reduction of interferences, mostly from ^{241}Pu β-decay. The results are shown in Table 1 in comparison with Vaninbroukx et al., Berlizov et al., and the present work.

Results from several publicly-available databases are also shown in Table 1. ENSDF, NuDat 3.0, and JENDL-5 show an order-of-magnitude disagreement for the 103 keV emission probability; these values appear to be taken from a Kellett 2011 interpretation of data [10] from Schmorak 1972 [11] but with a transcription error in the exponent. LNBH does not suffer this error. Some databases include the results of Berlizov, but none include the results of Wang.

In 2016, Bates et al. successfully observed the 45 keV line with a metallic magnetic calorimeter (MMC, a type of microcalorimeter) [19]. The sample was 10.81 Wt% ^{242}Pu, prepared as a solution dried onto a thin foil, and measured through a low-attenuation window. A FWHM resolution of 140 eV was achieved, and a quantitative measurement of the ^{242}Pu concentration was reported in agreement with the declared value. This was a remarkable achievement, but the sample preparation and low-attenuation requirements are impractical for most NDA applications. Here we present NDA measurements which required no such sample preparation or considerations.

2 Relocation and Installation of the Instrument

For our measurements of the ^{242}Pu, we used the microcalorimeter array instrument, SOFIA (Spectrometer Optimized for Facility Integrated Applications) [20]. The instrument was designed to be re-locatable and this was our first experience with moving it to a different site. The instrument was moved from Los Alamos National Laboratory (LANL) Technical Area 35 Building 02 to LANL Technical Area 55 Building PF-4. Figure 1 is a photograph of the instrument and supporting hardware loaded for transport. Major components consist of (left to right in Figure 1) a compact milliKelvin cryostat housing the detector, electronics rack, and helium compressor.

SOFIA uses no liquid cryogens and requires only 220V, single phase power for the air-cooled helium pulse tube cryocooler. Suitable electrical power was readily available in the new location. Existing work authorization for gamma ray measurements in PF-4 was determined to cover operations as the instrument presents no unique hazards. No significant issues were encountered during the move and

installation, and performance was verified to be consistent with that observed in the TA-35 laboratory.

3 Characteristics of ^{242}Pu Samples

Five spectra were measured from two ^{242}Pu standards (STDB242C8 and STDB242C3) using the SOFIA instrument over the course of seven months. These items were 10.93 g of PuO_2 (item measured once) and 113.6 g of PUO_2 (item measured four times). At the time of measurement both items were 86.85 Wt% ^{242}Pu. They were packed in steel "food-pack" cans of unspecified wall thickness inside SAVY containers [21]. For measurement, the packaged item was simply placed in front of the instrument with no preparation, consistent with a routine NDA procedure, as shown in Figure 2. It is notable that all prior gamma measurements of ^{242}Pu involved bare or very lightly attenuated samples, and most involved ultra-pure (>99.5 Wt%) ^{242}Pu, but the purpose of these measurements was to demonstrate the utility of the μCal instrument for routine nondestructive quantification.

4 103-keV Peak Analysis

The spectra in this region are complex with overlapping features, but the energy resolution (70-eV FWHM) achieved with SOFIA is sufficient to extract the area of the ^{242}Pu peak. Figure 3 shows an illustrative example of the region between 102.5 and 104.5 keV which includes peaks from ^{241}Am, ^{242}Pu, ^{241}Pu, the Kα1 Pu x-ray, and ^{240}Pu. This region of interest (ROI) is fit using the software SAPPY [5, 22, 23, 24]. The ^{242}Pu peak and nearby cluster are fit very well. The high-energy side of the ^{241}Am 102.966 keV peak is fit poorly, suggesting that unexplored structure(s) are likely present. This is a topic for future investigation but has negligible effect on the ^{242}Pu peak analysis.

The nearby ^{241}Pu 103.68 keV peak is of practical interest for plutonium isotopic analysis. It arises directly from the ^{241}Pu α-decay (^{214}Pu $\rightarrow^{237}$U), unlike most signatures for this radionuclide, which involve the ^{241}Pu $\rightarrow^{241}$Am $\rightarrow^{237}$Np and/or ^{241}Pu $\rightarrow$ ^{237}U $\rightarrow$ ^{237}Np chains. The direct decay means that the signature is time-independent and therefore reliable for very freshly separated plutonium. Fluorescent Pu Kα1 X-rays at 103.74 keV interfere with this ^{241}Pu peak (as well as the ^{242}Pu), but SAPPY is able to isolate the individual components.

The ^{240}Pu peak at 104.23 keV is also of practical interest for improving precision of isotopic analysis. This peak is cleanly separated and its area is easy to extract. A drawback is interference from Sn Kα1 and Kα2 escape lines at 104.03 and 104.25 keV (too weak to be seen here but stronger for low-burnup samples). Software techniques to deconvolve the escape peaks will be necessary to take full advantage of this ^{240}Pu signature.

In this ROI, we focus on fitting the ^{242}Pu peak. This is the clearest observation of this peak from ^{242}Pu decay ever reported, and this is the first time it has been observed without special sample preparation. Vaninbroukx et al. and Berlizov et al. observed an unresolved multiplet including ^{241}Am, ^{240}Pu, ^{241}Pu, and ^{242}Pu components. Wang likely observed this peak via $\alpha - \gamma$ coincidence

Figure 1: SOFIA and supporting hardware loaded for transport.

Figure 2: Standard item being placed in its measurement configuration by co-author Eric Feissle.

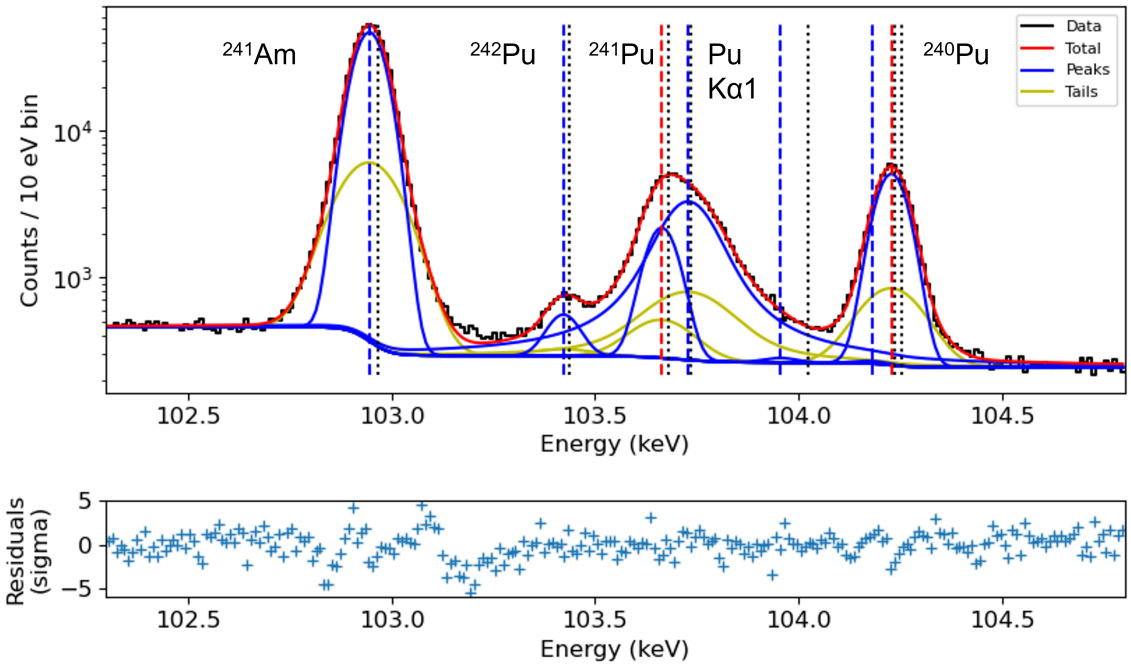


Figure 3: SAPPY Fits of the ROI from 102.5 keV to 104.5 keV. Notable peaks are labeled.

measurements but did not specifically report it, and Bates et al. did not report observations in this region likely due to limited efficiency.

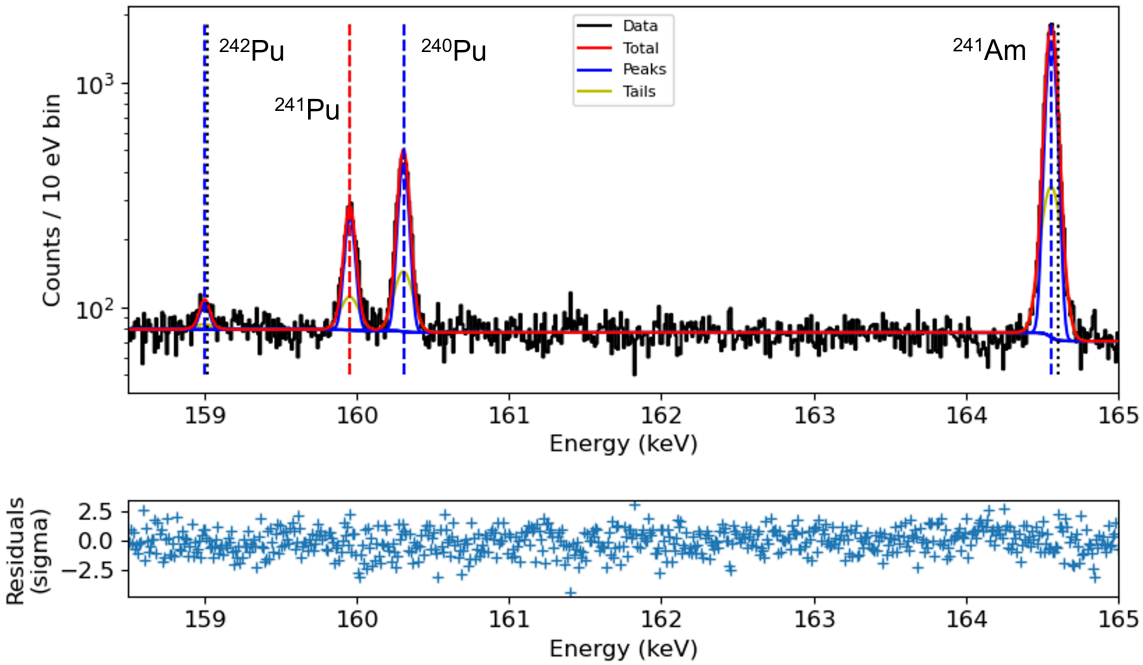
When fitting the ^{242}Pu peak, SAPPY estimates the peak centroid and area. During this process, the Lorentzian of the Pu Kα1 is set to 110.5 eV based on recent findings from Wessels et al. [25]. Based on independent fits for the five standard items, we found the centroid of the ^{242}Pu peak to be 103.440 keV $\pm$ .002 keV. This standard deviation reflects the statistical variance between the five standards rather than propagated uncertainty in the fit. This is more than 1 sigma lower in energy than the 103.499 $\pm$ 0.035 keV reported by Schmorak et al. [11], who discuss "impurities which made it more difficult to establish the peak centroids," so the difference is unsurprising. The actual centroid position is fortunate for quantification because the separation from interfering peaks is increased from what was expected.

Although SOFIA is able to measure relative centroid energies with a precision better than 0.5 eV, a limiting factor is the uncertainty in adopted centroids of nearby peaks needed for calibration, especially ^{241}Am. Our ^{242}Pu centroid shown above is interpolated using ^{241}Am at 102.966$\pm$0.005 keV [26] and Pu Kα1 at 103.7347$\pm$0.0006 keV [27]. A secondary limitation for centroid determination is nonlinearities that are potentially introduced during processing of the μCal data (see Yoho et al. 2020 [28]), which is a topic for future interest if this instrument will be used to improve nuclear data tables. Even with these limitations, our new value is considerably more reliable than the previously adopted value.

For each of the items, we calculate a gamma emission probability and propagate uncertainty. We do so by using the mass ratio of ^{242}Pu/^{240}Pu. This allows us to leverage the cleanly separated ^{240}Pu peak at 104.23 keV. Since the two peaks are within 1 keV, the uncertainty in the efficiency curve can be considered negligible. We found the weighted average emission probability to be $2.70(3)E-5$. The weighted average takes into consideration the propagated uncertainty of each run, which is largely driven by the uncertainty in the peak area estimation. Our estimate falls between the Vaninbroukx et al. and Wang estimations but offers an improvement in the uncertainty.

5 159-keV Peak Analysis

The 159-keV region includes cleanly-separated peaks from ^{242}Pu at 159.02 keV, ^{241}Pu at 159.96 keV, and ^{240}Pu at 160.31 keV as seen in Figure 4. Vaninbroukx et al., Berlizov et al., and Wang observed this region as an unresolved triplet, and Bates did not report observations in this region. We present



[b]

Figure 4: SAPPY Fits of the ROI from 159 keV to 165 keV. Notable peaks are labeled.

our preliminary estimation of the energy value and yield for the ^{242}Pu peak. Our measurement of the centroid energy is 159.016±0.031 keV and the weighted average of the gamma emission probability is $1.80(7)E-6$. This value falls several sigma below the previous works' estimations. Further work is needed to decrease the statistical uncertainty and resolve the discrepancy between the statistical and propagated uncertainties. We predict both of these issues are impacted by poor fits due to fewer counts in this ROI.

6 Efficiency Calculation

Relative efficiency calibration as a function of energy is necessary for accurate quantification, even though the extrapolation distances used here are less than 1 keV. The calibration involves the intrinsic detector efficiency, attenuation by the steel container, and self-attenuation within the PuO_2 samples. Intrinsic detector efficiency is determined using a separate ^{133}Ba source and includes four free parameters in the form $\exp(a_0 + a_1 \ln E + a_2 \ln^2 E + a_3 \ln^3 E)$ where E is γ-ray energy in keV. Two additional free parameters that account for container thickness and sample self-attenuation are found using two gamma ray peaks from ^{238}Pu, one from ^{240}Pu, three from ^{241}Pu, and five from ^{241}Am, using the test item itself as the source of radiation. The calibration assumes that the declared isotopic ratios are correct. Many other peaks are available but for simplicity, we avoided any that involve multiple decay channels (such as 208.00 keV, which has components from both ^{237}U and ^{241}Am). Net peak areas were extracted using SAPPY. An example of the relative efficiency curve calculated from one of the items is seen in Figure 5.

7 Conclusion

We have successfully demonstrated the utility of μCal for estimating the ^{242}Pu gamma emission probabilities per decay for the 103- and 159-keV peaks, thus supporting its viability for ^{242}Pu isotopic quantification. This meets a long-standing NDA challenge. We have observed, for the first time, nearly entirely resolved ^{242}Pu 103- and 159-keV gamma ray peaks in samples that were not specially prepared and this is the highest resolution observation ever reported by any method. We have found a more precise value for the 103 keV centroid, a more precise value for the gamma emission probability

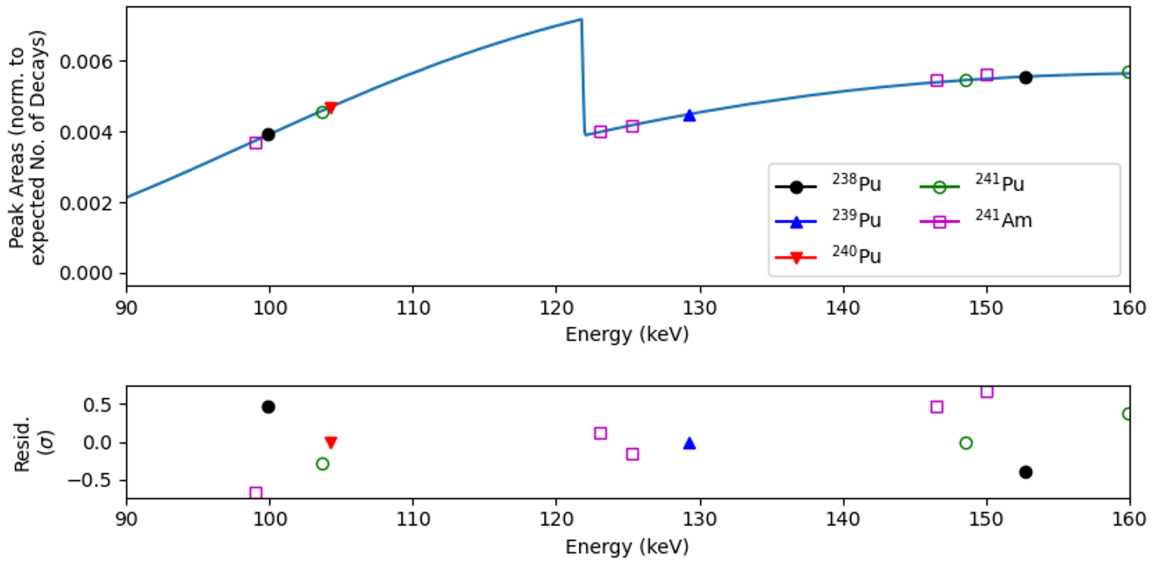


Figure 5: Efficiency calibration curve derived from five radionuclides within the sample.

for the 103 keV peak, and demonstrated the portability of the SOFIA instrument. We also found a new estimate for the 159-keV gamma emission probability, which was far lower than any other previously recorded estimates. Further investigation into the validity of this estimate is needed. Once validated, this peak may offer advantages for higher-mass samples, consistent with Vaninbroukx's findings.

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