SAPPY: A New Software Pipeline for Analysis of Gamma-Ray Spectra Produced by Microcalorimeter and High-Purity Germanium Detectors

D.T. Becker[1], D.A. Bennett[2], M.H. Carpenter[3], M.P. Croce[3], M.W. Keller[2], K.E. Koehler[3], D.G. McNeel[3], D.J. Mercer[3], K.A. Schreiber[3], D.R. Schmidt[2], E.S. Teti[3], J.N. Ullom[1,2], D.T. Vo[3], and A.L. Wessels[2]

[1]University of Colorado, Boulder
[2]National Institute of Standards and Technology
[3]Los Alamos National Laboratory

Abstract

Gamma-ray spectroscopy using cryogenic microcalorimeters has matured significantly in the last few years with the deployment of spectrometers at Los Alamos National Laboratory, Oak Ridge National Laboratory, and Idaho National Laboratory, with additional instruments planned for delivery over the next few years. The goal of these spectrometers is to take advantage of the excellent energy resolution provided by TES microcalorimeters to close the gap in accuracy between non-destructive and destructive assay, with applications in nuclear materials control and accounting. A critical part of these efforts has been in the data processing pipeline. We have developed SAPPY (the Spectral Analysis Program in Python) to extract isotopic information from gamma-ray spectra produced by both microcalorimeter arrays and High-Purity Germanium (HPGe) detectors. SAPPY analyzes microcalorimeter and HPGe spectra using a common code base, using common fundamental nuclear data and consistent algorithms for peak fitting, efficiency curve fitting, and extraction of isotopic contents. The software also directly incorporates uncertainties in nuclear data such as gamma-ray emission probabilities in a consistent way. Using consistent nuclear data and algorithms for both microcalorimeters and HPGe allow direct comparisons of the accuracy of these technologies for performing isotopic analysis.

1   Introduction

Over the last 10 years, cryogenic microcalorimeter spectrometers have emerged as cutting-edge tools for improved Nuclear Material Accountancy and Control (NMAC) based on gamma-ray spectroscopy [1, 2, 3]. Gamma-ray spectrometers based on cryogenic microcalorimeters combine unmatched energy resolution with broadband spectral response, providing 5x – 10x better spectral resolution than widely used High Purity Germanium (HPGe) detectors, and are thus able to resolve closely spaced spectral features and improve peak-to-background ratios in NMAC measurements (compare Figure 2 and Figure 3). The resolution advantage of microcalorimeters over other gamma-ray detectors is intrinsic; their ~ 100 mK operating temperature suppresses electrical and thermal noise and provides access to beneficial quantum phenomena such as superconductivity.

These spectrometers are now being deployed to national lab settings to make measurements. The 256-detector Spectrometer Optimized for Facility Operations (SOFIA) spectrometer was deployed Los Alamos National Laboratory in 2021 [3], is currently located at Oak Ridge National Laboratory, and is actively being used to make measurements [4]. A 384-pixel spectrometer was deployed to

Idaho National Laboratory in 2022, and a 672-pixel instrument is scheduled for deployment to Pacific Northwest National Laboratory in 2024. Additional instruments using the same form-factor as SOFIA are also being assembled for delivery in 2024.

In order to accurately extract isotopic information from gamma-ray spectra, specialized analysis software is required. For spectra generator using HPGe detectors, multiple software packages are available and have been under development for decades, such as MGA [5] and FRAM [6]. However, these software packages have drawbacks for analyzing microcalorimeter spectra. The excellent energy resolution of microcalorimeters allows x-ray lines to be distinguished from gamma-ray lines, and reduces or eliminates the necessity to fix line locations when dealing with overlapping peaks. Due to the small size of microcalorimeter absorbers, microcalorimeter exhibit clear "escape" peaks which must be accounted for. Additionally, existing software does not account for uncertainties of reference data such as gamma-ray emission probabilities.

We therefore have developed a new software package, the Spectral Analysis Program in Python, or SAPPY. This paper provide a general overview of SAPPY, leaving a more detailed description of its operation for future work.

2   SAPPY Overview

SAPPY emerged from prior work in [7], which described the first software package to be capable of analyzing both HPGe and microcalorimeter data, as well as accounting for uncertainties in reference data. SAPPY initially began as a re-implementation of that work in the Python programming language, and continues this heritage of supporting both HPGe and microcalorimeter spectra while also accounting for uncertainties in reference data. Using consistent nuclear data and algorithms for both microcalorimeters and HPGe allow direct comparisons of the accuracy of these technologies for performing isotopic analysis. SAPPY also includes additional algorithmic improvements and advances, some of which were described in [8], while others are described below.

The general operation of SAPPY is depicted in Figure 1. Data from a spectrometer is first gathered and a binned spectrum is produced. SAPPY then breaks the spectrum into a set of "Regions of Interest" (ROIs). The number and endpoints of these ROIs is user-configurable. As independent fit is then performed within each ROI to extract estimates for the areas of each peak (see Section 3). The peaks to be fit and various properties of the peaks are also set via user configuration. While operating for the extraction of Pu isotopic ratios, a typical configuration results in 15 peak areas across the 5 isotopes that SAPPY currently supports (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am). While ^{242}Pu is currently not included in this list, recent work demonstrates that quantification of ^{242}Pu content is possible in some samples [4].

These peak areas are then passed to a separate fitting routine which performs a simultaneous fit for both the decay intensities of each isotope as well as the shape of the total source-detector efficiency curve (see Section 3). The model to be optimized is:

$$A_j = \Sigma_k I_k BR_{j,k} \eta(E_j, \vec{p}). \tag{1}$$

Here the index j covers the peaks while the index k cover the isotopes; the sum on the right-hand side of the equation reflects the fact that some peaks can be produced by the decay of two different isotopes (e.g the gamma-ray peak at 98.97 keV can be produced directly through the decay of ^{241}Am, or indirectly by the decay of ^{241}Pu to the short-lived isotope ^{237}U). A_j are the peak areas. I_k are the

"decay intensities" for each isotope, defined as the number of decays per unit time for that isotope. $BR_{j,k}$ is the gamma-ray emission probability for isotope k to decay into peak j. E_j is the energy for peak j, and $\eta(E_j, \vec{p})$ is the total source-to-detector efficiency curve, with $\vec{p}$ representing the parameters of the efficiency curve model. The fit that we perform here accounts for uncertainties in the gamma-ray emission probabilities, as well as the uncertainties in the peak areas, which reflect counting statistics and peak overlaps.

After performing this fit, the isotope intensities are turned into isotopic mass ratios using the half-lives and atomic masses of each isotope. The uncertainties of the half-lives are also accounted for here, but in practice these uncertainties are much smaller than the uncertainties in emission probabilities, so that the total uncertainties are dominated by a combination of counting statistics and the emission probabilities.

We currently use a semi-physical model for the efficiency curve:

$$\eta(E, x_{Pu}, x_{Cd}, p_0, p_1) = \frac{1 - e^{-\mu_{PuO_2}(E)x_{Pu}}}{-\mu_{PuO_2}(E)x_{Pu}} \; e^{-\mu_{Cd}(E)x_{Cd}} \; E^{p_0} p_1^{1/E}. \tag{2}$$

Here the $\mu(E)$ functions are the total x-ray absorption coefficients, excluding coherent scattering, taken from the XCOM database [9]. The first term covers the self-absorption of photons within a PuO_2 source, the second any attenuation between the source and the detectors, and the last two terms are a detector efficiency term borrowed from FRAM.

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

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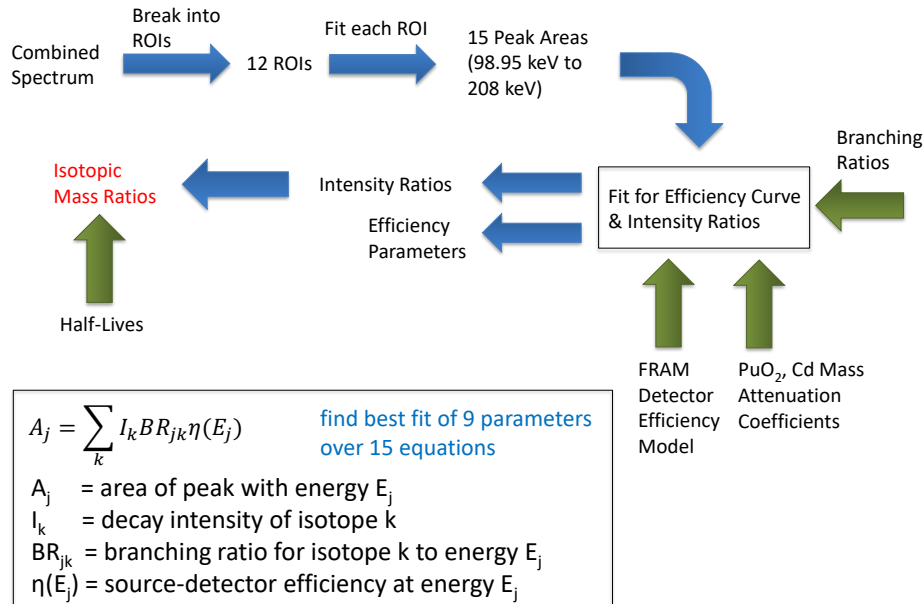$\eta(E_j)$ = source-detector efficiency at energy E_j

Fig. 1: Diagram showing the procedure that we use for turning a microcalorimeter or HPGe spectrum into a best-fit estimate of isotopic ratios. Starting with a combined spectrum we break into ROIs, and fit the spectrum in each ROI, extracting 15 peak areas from 98.5 keV – 208 keV. We then perform a fit to extract efficiency curve parameters and isotopic decay intensities from the 15 peak areas. Finally we use isotopic half-lives and masses to infer isotopic mass ratios.

3   Fitting Procedures

Figure 2 shows the result of a SAPPY fit to a complicated ROI for microcalorimeters, and Figure 3 shows a fit for a similar ROI for HPGe. We highlight several items regarding these ROI fits:

- Each ROI is fitted completely independently.

- SAPPY performs a proper maximum-likelihood fit, accounting for the Poisson statistics of the counts in each bin [10].

- For microcalorimeter spectra, SAPPY uses a "double-gaussian" distribution for the gamma-ray peak shape, as described in [7]. For x-ray peaks it uses a Voigt function (the convolution of a Gaussian and Lorentzian). The Gaussian widths are floated but common to all peaks within each ROI; the Lorentzian peaks are unique to each peak, and can be either floated or fixed.

- For HPGe spectra, SAPPY uses a Gaussian peak-shape with a low-energy Bortels tail [11].

- The background model is a smooth change in background level across the ROI where the change in level is proportional to the peak model. This is equivalent to assuming that a fixed fraction of events in each peak are scattered into a low-energy tail that is constant in energy.
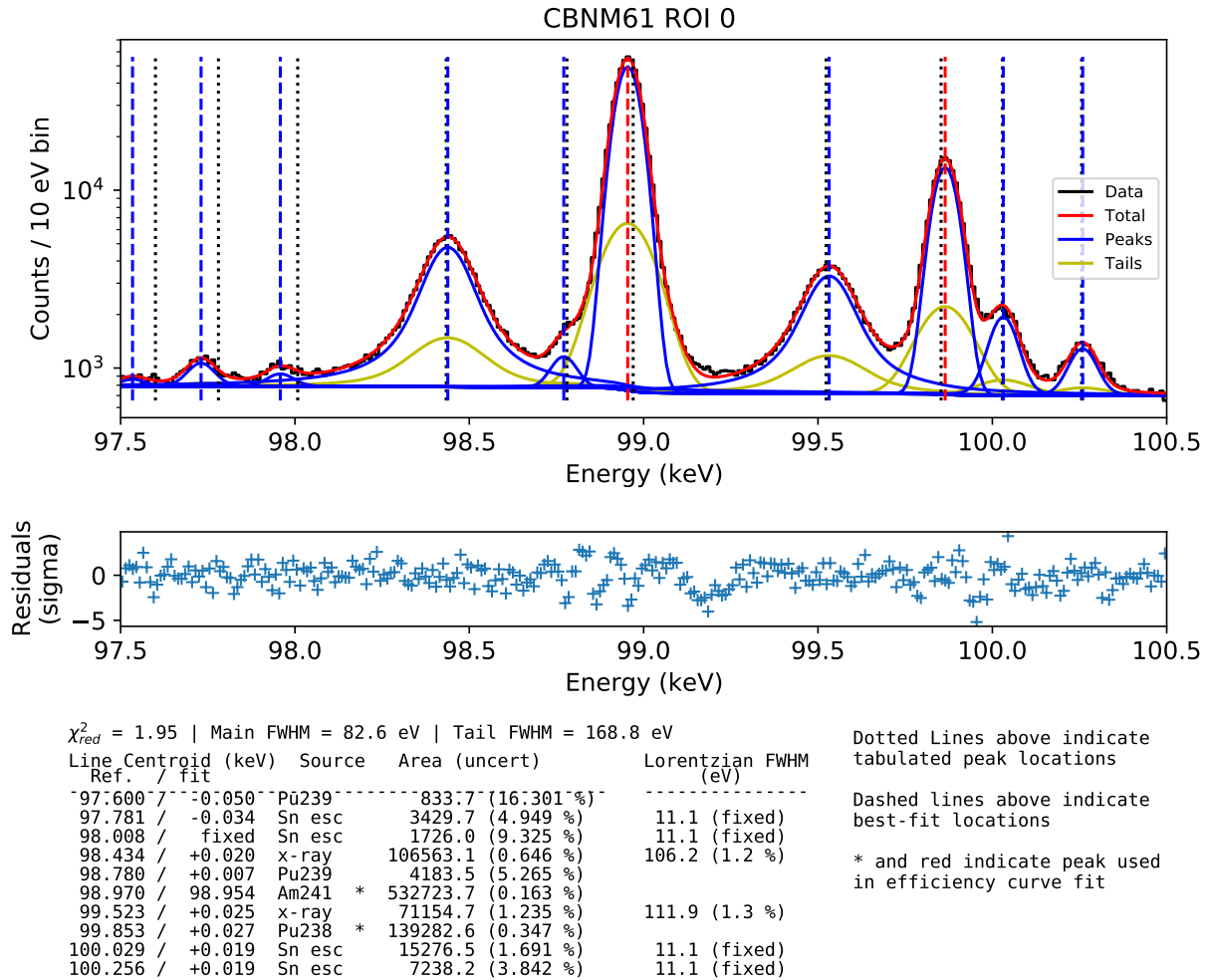
Fig. 2: Output from SAPPY when fitting the lowest-energy ROI for microcalorimeter data. The sample is CBNM61, which is ~ 62.5 % ^{239}Pu by mass. The ROI extends from ~ 97.5 keV – 100.5 keV. Ten peaks are included in the fit, including 2 x-ray peaks, 4 Sn escape peaks, 2 "nuisance" peaks, and two peaks whose areas are used in isotope ratio extraction: The 98.97 keV peak of ^{241}Am and the 99.853 keV peak of ^{238}Pu (marked with * in the tables below).

- The background levels at the edges of the ROI are floated, allowing for uncertainties in background level to be incorporated naturally by the fitting routine.

- For microcalorimeter spectra we account for relevant "escape" peaks from the Sn microcalorimeter absorbers.

- The fit outputs an uncertainty for all peak areas within the ROI.

The result of SAPPY's fit to the to the combined efficiency/isotope model described above is shown in Figure 4. This fit is highly nonlinear, and so our fitting routine randomly chooses different "starting values" for the parameters to ensure that we find a good global fit. This iterative process
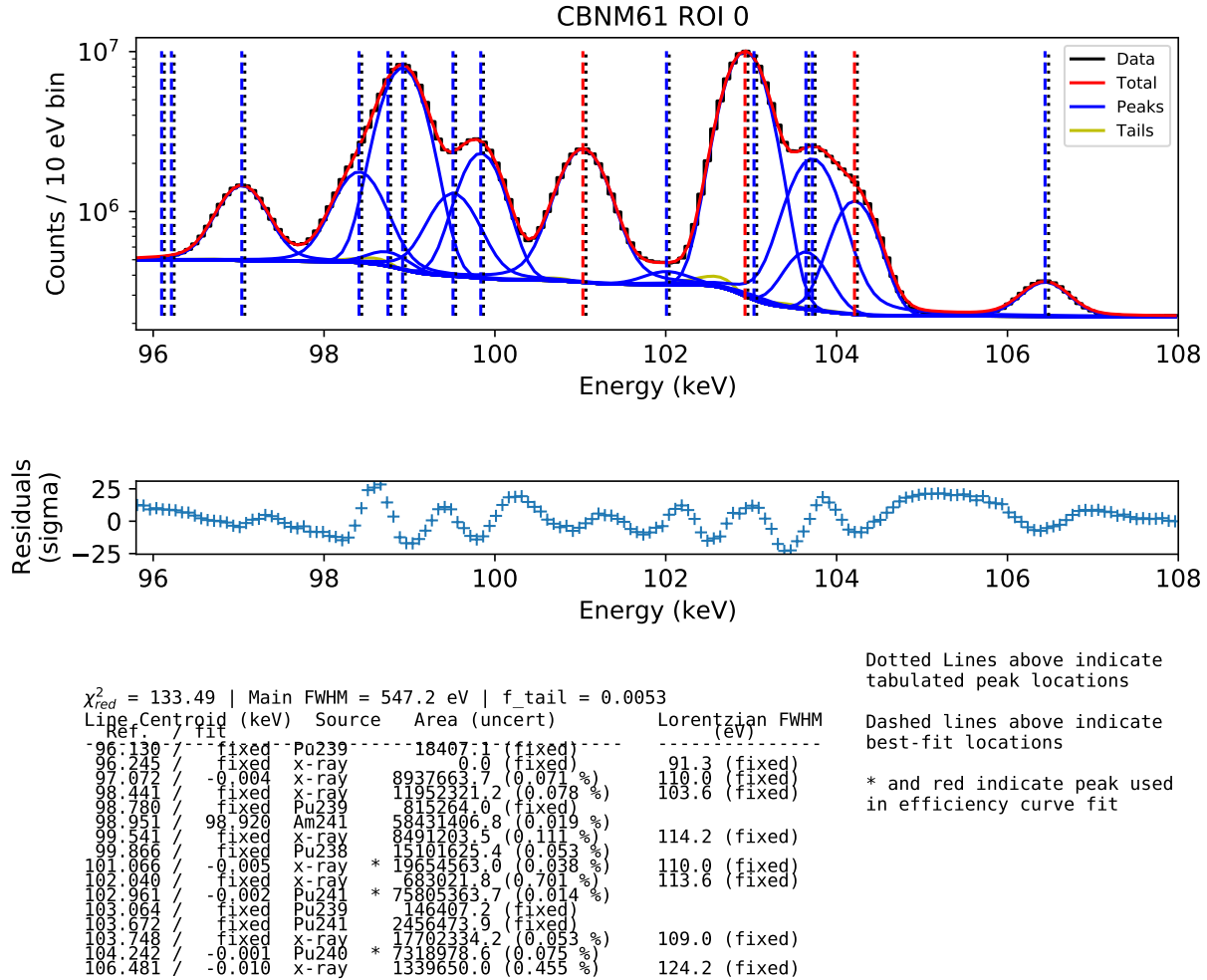
Fig. 3: Output from SAPPY when fitting the lowest-energy ROI for HPGe data. As in Figure 2 the sample is CBNM61, but for HPGe the ROI is much wider: 95.5 keV – 108 keV. This wider ROI is chosen to avoid placing the ROI edge near the tail of a peak, which can bias results. Sixteen peaks are included in the fit, including 8 x-ray peaks, 5 "nuisance" peaks whose area are not used in the efficienct/isotope fit, and three peaks whose areas are used in isotope ratio extraction (marked with * in the tables below).

repeats until five sets of starting values find the same equally good solution. The fit performed is a maximum likelihood fit that accounts for the uncertainties in the peak areas (which are extracted from the ROI fits), as well as the uncertainties in the tabulated values for the gamma-ray emission probabilities. We use ENSDF as our source for these probabilities and their uncertainties [12].

We have used Monte-Carlo simulations of our system to verify that the combination of our fitting routines and our efficiency curve model introduce less than 0.1 % relative uncertainty and bias in the extracted isotopic ratios. This is much smaller than the uncertainties of the gamma-ray emission probabilities.
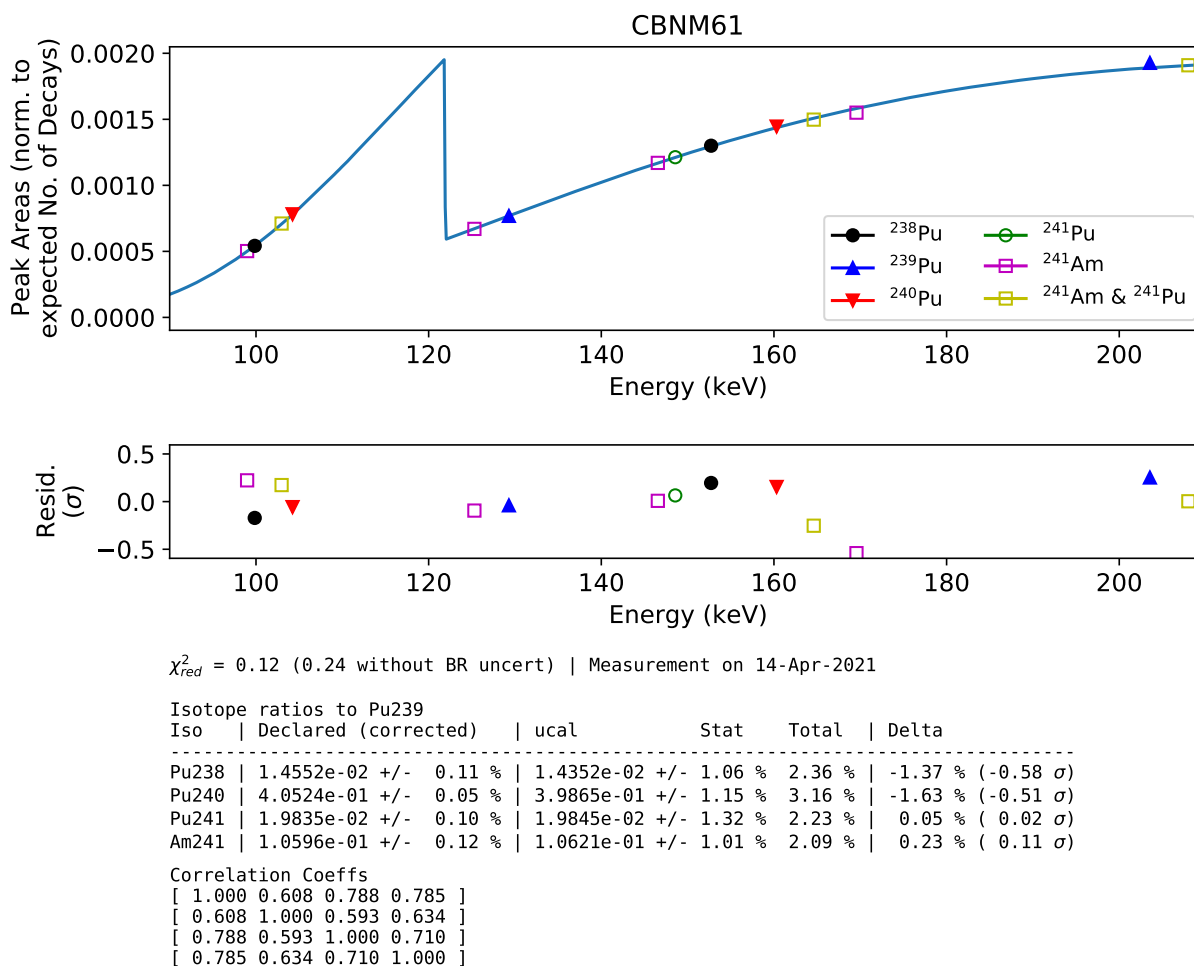
Fig. 4: Output from SAPPY from performing the efficiency/isotope fit. The upper plot shows the shape of the best-fit efficiency curve in solid blue; markers show the measured peak areas normalized to the expected number of measured decays based on the best-fit efficiency curve and best-fit isotope quantities. The peak shape and color indicate the isotope. The lower plot shows the residuals. The upper table gives the declared and measured isotopic mass ratio to ^{239}Pu along with their uncertainties, and the difference ("Delta") between the measured and declared values. The declared values have been decay-corrected to the measurement date. The bottom table gives the correlation coefficients between the four isotope ratios.

4 Future Directions

In the future we plan to extend SAPPY in several directions. First, SAPPY currently only handles the 5 isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am. A top priority is extending it so that the supported isotopes can be easily configured by a user. This will allow us to use SAPPY to extract information such as U enrichment levels.

Second, SAPPY's user interface is currently text-based. We are actively working to build a GUI interface for SAPPY so that it is easier for people outside our team to use.

Finally, it is possible to run the efficiency/isotope fitting routine in "reverse", taking declared isotopic content of a sample as input, and then extracting gamma-ray emission probabilities. Microcalorimeter spectrometers have already been demonstrated to be capable of reducing uncertainties in these emission probabilities [13], and making this "reverse" capability easier to access from SAPPY will allow us to extend this analysis to below the Pu K-edge.

5   Conclusions

We have described a new software package, SAPPY, for extracting isotopic information from both microcalorimeter and HPGe gamma-ray spectra. A common set of reference data (half-lives and gamma-ray emission probabilities) are used for both microcalorimeters and HPGe, which allows direct comparisons of the accuracy of these technologies for performing isotopic analysis. The software also directly incorporates uncertainties in nuclear data such as gamma-ray emission probabilities in a consistent way.

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