



Gamma Spectroscopy Performance for Irradiated Solid-Form TRISO Fuel

**Prepared for
US Department of Energy**

Mark Croce, Emily Stark, Christine Mathew, Daniel McNeel, Katherine Schreiber, Rico Schoenemann, Matthew Carpenter, Sophie Weidenbenner¹

Callie Goetz, Jason Harp, Jianwei Hu, Robert McElroy, Craig Gray²

¹Los Alamos National Laboratory

²Oak Ridge National Laboratory

**September 2024
LA-UR-24-30542**

DISCLAIMER

This information was prepared as an account of work sponsored by an agency of the U.S. Government. Neither the U.S. Government nor any agency thereof, nor any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness, of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. References herein to any specific commercial product, process, or service by trade name, trade mark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the U.S. Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the U.S. Government or any agency thereof.



SUMMARY

The goal of this measurement campaign is to provide a direct evaluation of nondestructive composition analysis performance between traditional and advanced gamma spectroscopy technologies for burnup characterization of irradiated TRISO materials. This work is part of a strategy to enable cost-effective material accounting and safeguards for pebble bed reactors by making use of nondestructive analysis technologies where possible to reduce reliance on sampling and destructive laboratory analysis. Irradiated TRISO compacts, subsamples of compacts, and individual fuel particles were measured using high-purity germanium and microcalorimeter detectors at the Oak Ridge National Laboratory Irradiated Fuel Examination Laboratory and Energy Systems Test Complex.

The primary conclusions of this work are:

- Microcalorimetry and HPGe both have potentially useful signatures that are related to burnup. The highest quality microcalorimetry signature is the ratio of the Pu $K\alpha_2$ X-ray to the U $K\alpha_1$ X-ray. The Cs isotope ratio from HPGe spectra is also relevant, however it is not a direct measure of the Pu content of the irradiated fuel and requires additional information to reliably infer burnup especially between different reactor types.
- When considering peak ratios from a spectrum collected by a multipixel array, peak ratios with less separation in energy space are more reliable when using small peaks. One factor contributing to this is the uncertainty associated with the relative efficiency curve uncertainty.
- A microcalorimeter spectrum collected for the purpose of determining the U to Pu content in irradiated fuel with a similar background as the hot cell measurements would need approximately $6e7$ counts. This value can be used when considering the development of a microcalorimeter gamma detector to get a sufficient measurement in a given time.

1. INTRODUCTION

The goal of this work is to provide a direct evaluation of nondestructive analysis performance between traditional and advanced gamma spectroscopy techniques for irradiated TRISO materials with particular emphasis on peak ratios that may be used to determine fuel burnup and fissile material content. Characterization of fuel burnup is necessary for a reactor operator to determine whether an individual pebble has reached its target burnup, or if it may be re-introduced into the reactor core. Characterization of fissile material content is necessary to meet nuclear material accounting and safeguards requirements.

2. SIGNATURES OF SOLID VS. LIQUID FUELS

Gamma spectra of solid and dissolved irradiated fuels show substantial similarities in peaks observed, especially from fission product gamma rays. However, there are several notable differences relevant to burnup characterization. While dissolved fuels (as in molten salts or aqueous dissolved samples) are homogeneous in composition, this cannot necessarily be assumed for solid fuel forms. Traditional light water reactor fuel pins are known to vary significantly in burnup across their diameter. Furthermore, the fuel pin diameter is large compared to the gamma-ray attenuation length (for example, the mean free path for 186 keV gamma rays is 0.07 cm in sintered UO_2 [1]) which means that a gamma-ray measurement is only sensitive to the outer surface of the fuel [2]. TRISO pebble fuels are a much more favorable configuration to determine bulk average composition with passive gamma spectroscopy. The fissile material is distributed in small particles in a carbon matrix with a kernel diameter of only several hundred microns [3]. Although gamma-ray attenuation within the fissile material kernel and surrounding matrix is still relevant, this means that gamma spectroscopy is more feasible to determine the average properties of TRISO fuels relative to traditional fuel pins.

A second key difference between solid and liquid fuels is the intensity of self-induced X-ray fluorescence peaks as shown in Figure 1. X-ray fluorescence of uranium and plutonium in irradiated fuels is understood to result primarily from beta-decaying fission products. Fission products and actinides are concentrated within the kernel of TRISO particles, therefore dramatically increasing the production of actinide fluorescence X-rays compared to liquid fuels. This means that self-induced fluorescence of uranium and plutonium K X-rays is an effective signature to indicate fissile material composition and burnup in irradiated TRISO fuels.

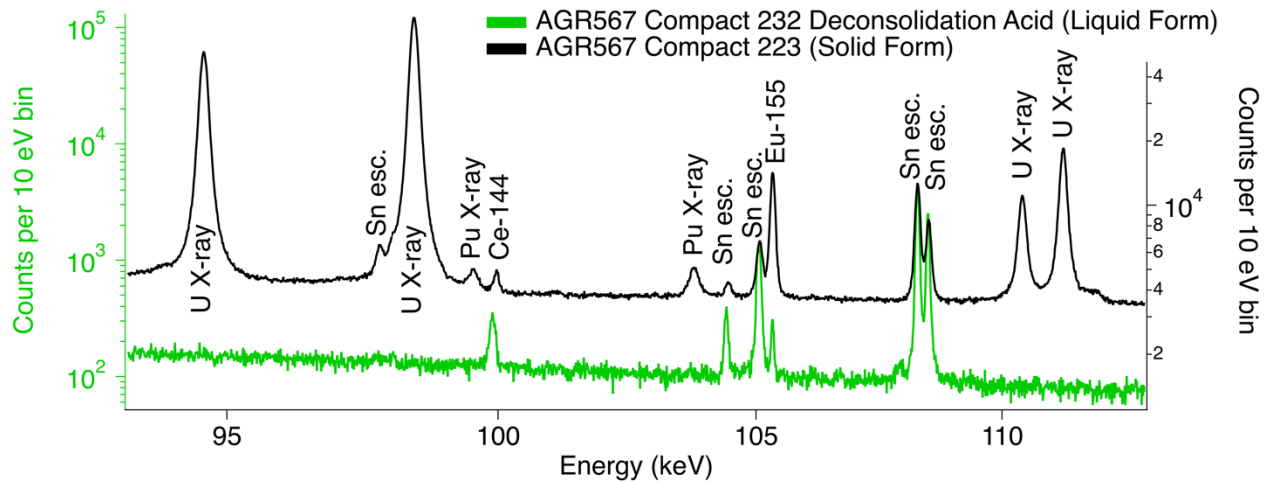


Figure 1: U and Pu fluorescence X-rays are clearly observed in microcalorimeter spectra of solid-form TRISO fuels, in contrast to dissolved fuels (from [4] *NDA of TRISO Fuels FY23 Report*). This signature allows the Pu/U element ratio to be directly and nondestructively determined – an important signature for safeguards.

3. MEASUREMENTS

Solid form TRISO fuels were measured in two locations at Oak Ridge National Laboratory. Table 1 summarizes the measured items. Intact compacts and subsamples of compacts consisting of many particles were measured through a port on the East Cell of the ORNL Irradiated Fuel Examination Laboratory. Items were placed using remote manipulators at the end of a lead collimator that extended into the hot cell. Detectors were located on a cart (Figure 2) at the other end of the collimator. This configuration allowed the detectors to be easily repositioned to align with the collimator and to adjust the count rate when needed. Background was measured with each detector type. Intact compacts were packaged in “dog bone” containers which consist of a section of stainless steel tube with compression fitting end caps (Figure 3). Subsamples of compacts were packaged in aluminum pellet cans.

Substantial background was observed in spectra acquired at the Irradiated Fuel Examination Laboratory due to scattering of gamma rays from the inventory of irradiated fuels within the hot cell. This motivated a second series of measurements at the Energy Systems Test Complex where single irradiated TRISO particles can be handled outside of a hot cell and measured in a relatively low-background environment (Figure 4).

Series	ID	Estimated Particles	Calculated Burnup GWd/MTU	Calculated Burnup %FIMA	Type	Irradiation Dates
AGR2	643A	235	69.7	7.26	UCO	6/22/10-10/16/13
AGR2	642-03	200	93	9.69	UCO	6/22/10-10/16/13
AGR2	621A	235	97.5	10.16	UCO	6/22/10-10/16/13
AGR2	622A	235	97.8	10.19	UCO	6/22/10-10/16/13
AGR2	331-03	150	99	10.31	UO2	6/22/10-10/16/13
AGR2	2-311A	150	101.8	10.6	UO2	6/22/10-10/16/13
AGR2	2-312A	150	102.3	10.66	UO2	6/22/10-10/16/13
AGR2	2-332-03	150	103.5	10.78	UO2	6/22/10-10/16/13
AGR2	2-542	2858	115.5	12.03	UCO	6/22/10-10/16/13
AGR2	2-521A	235	117.9	12.28	UCO	6/22/10-10/16/13
AGR2	212A	235	121.2	12.62	UCO	6/22/10-10/16/13
AGR2	2-222-03	200	121.8	12.69	UCO	6/22/10-10/16/13
AGR2	2-211	3176	120	12.5	UCO	6/22/10-10/16/13
AGR5/6/7	5-222A	235	134.6	14.02	UCO	2/16/18-7/22/20
AGR5/6/7	5-224A	235	137.6	14.33	UCO	2/16/18-7/22/20
AGR5/6/7	5-159A	235	89.2	9.29	UCO	2/16/18-7/22/20
AGR5/6/7	5-413A	235	135	14.06	UCO	2/16/18-7/22/20
AGR5/6/7	5-223	3176	137.6	14.33	UCO	2/16/18-7/22/20
KP	223/50651	20	not provided	12.43		discharged 2/2022
AGR5/6/7	221-RS24	1	not provided	14.03	UCO	2/16/18-7/22/20

Table 2: Summary of measured TRISO fuel materials. All items were measured at the ORNL Irradiated Fuel Examination Laboratory hot cells except for item ID 221-RS24, which was measured at the ORNL Energy Systems Test Complex laboratory for lower background. All items were measured with a Canberra GL1015 HPGe detector and items highlighted in blue were measured with the SOFIA microcalorimeter spectrometer.

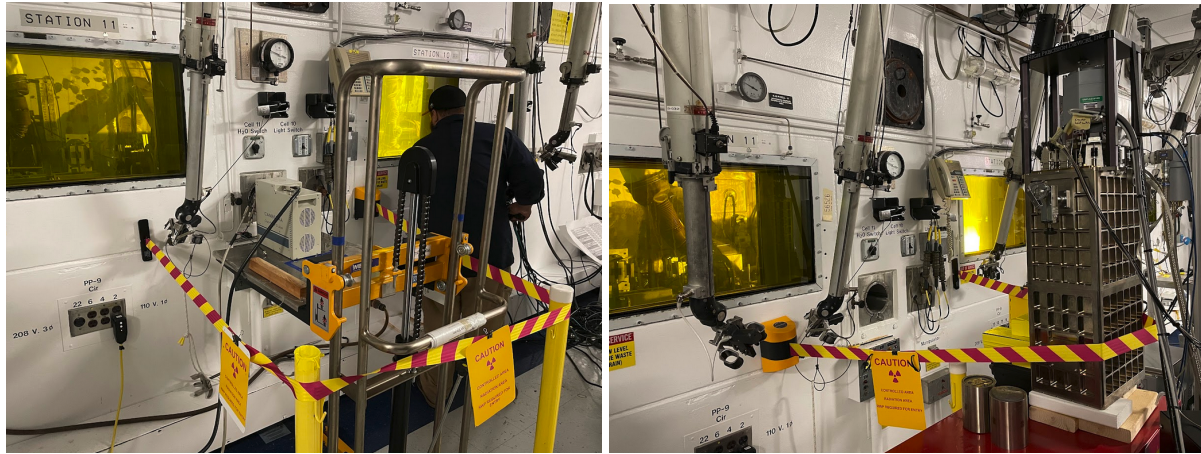


Figure 2: Canberra GL1015 HPGe (left) and SOFIA microcalorimeter (right) detectors were positioned on a cart in front of the ORNL Irradiated Fuel Examination Laboratory East Cell port. The distance between the detector and port was varied to adjust count rates. Temporary radiation areas were established around the detector due to the gamma dose rate.

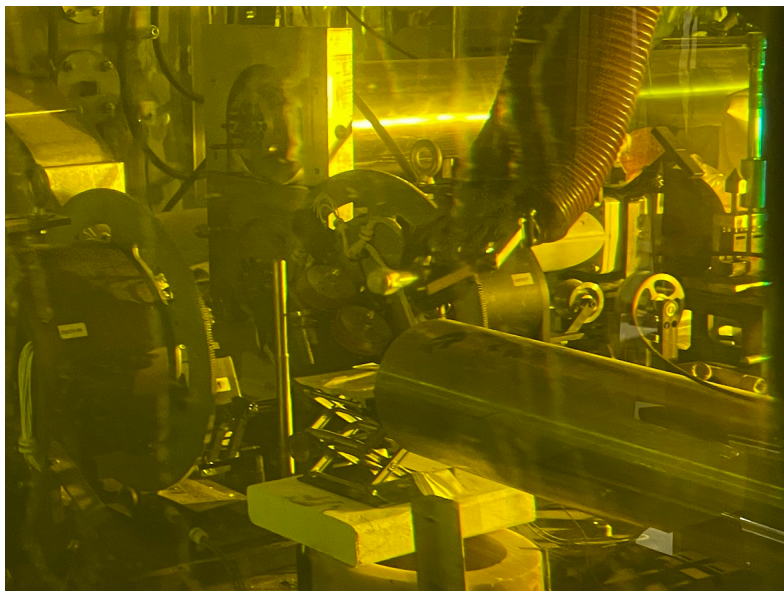


Figure 3: Compacts and subsamples were placed on a stand at the end of the East Cell port collimator. Here, an intact compact packaged in a “dog bone” container consisting of compression fittings on a stainless steel tube, is held in the manipulator.



Figure 4: Complementary measurements were done on a single TRISO particle in a relatively low-background laboratory at the ORNL Energy Systems Test Complex. The particle was packaged in a vial and placed on the table in front of the microcalorimeter and (not pictured) HPGe detectors.

4. ANALYSIS AND RESULTS

The goal of this analysis was to find the signature that most closely and reliably corresponded with burnup and the Pu content of the irradiated fuel. The energy range for the microcalorimeter data spans approximately 20 keV to 300 keV, while the energy range for the HPGe data spans 30 keV to 1200 keV. Microcalorimetry demonstrated approximately 6.6x better resolution than HPGe using the Ce-144 peak at 133.5 keV. This resolution allows for the direct observation of the small Pu $K\alpha$ X-ray fluorescence peaks, as seen in Figure 5.

Four signatures were identified for investigation: three from uCal and one from HPGe. They are described in Table 2. In particular, the question of which Pu x-ray to consider brought up questions of uncertainty quantification when dealing with multipixel array microcalorimeters when correcting for efficiency. The SOFIA detector consists of hundreds of individual detectors that are coadded to create a final spectrum. When a ratio of peak areas is transformed into a mass ratio, an efficiency correction is applied. Previous irradiated fuel analysis did not consider the relative change in the uncertainty in efficiency when considering peaks that are not within ~ 1 keV of each other. This understanding was needed to determine which Pu X-ray would provide the more reliable signature of burnup. The Pu $K\alpha 1$ X-ray is larger than the Pu $K\alpha 2$ X-ray, but it is farther in energy space, and therefore the efficiency difference is larger and potentially more variable. To determine the impact of a larger energy range, a study was conducted using the multiple individual

detectors in SOFIA and a previous measurement campaign of Pu reference materials which provided many more peaks for analysis.

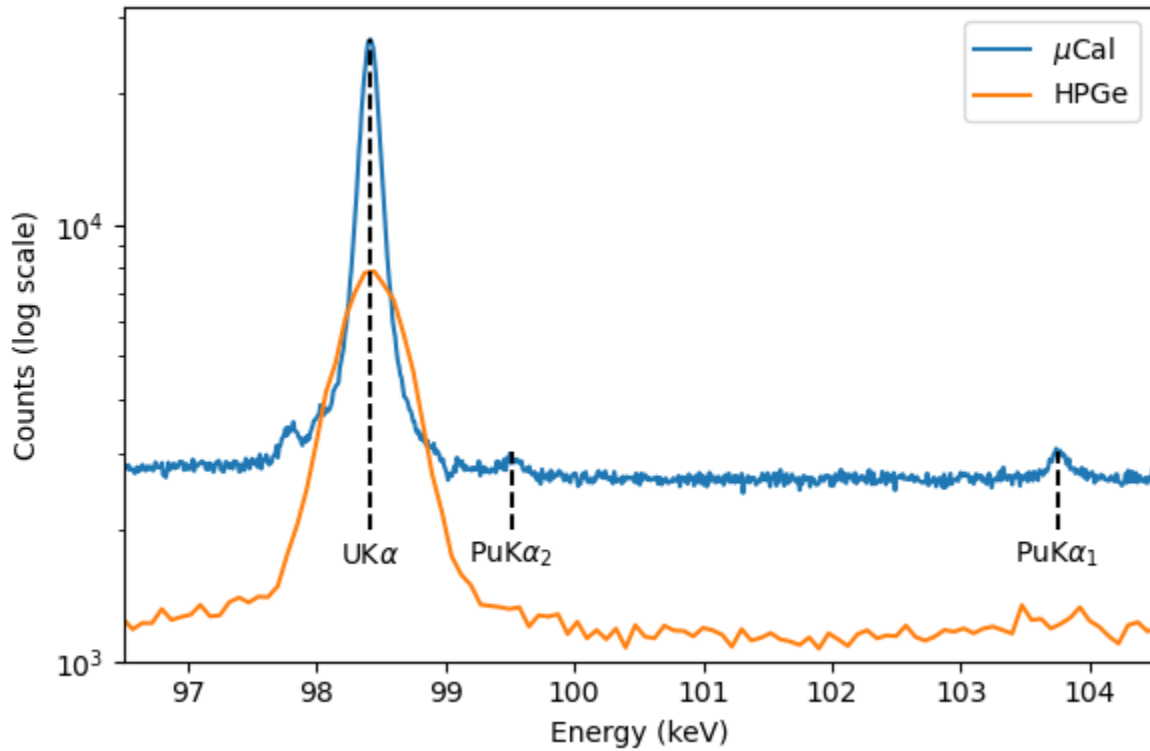


Figure 5. A comparison of the SOFIA microcalorimeter and Canberra GL1015 HPGe resolution. The weak Pu X-rays can be seen in microcalorimeter but not in HPGe data.

Ratio	Numerator	Num. Energy (keV)	Denominator	Den. Energy (keV)
Pu Ka2:U Ka1 (microcal)	Pu Ka2	99.53	U Ka1	98.44
Pu Ka1:U Ka1 (microcal)	Pu Ka1	103.75	U Ka1	98.44
Eu155:Eu154 (microcal)	Eu155	86.55	Eu154	123.07
Cs134:Cs137 (HPGe)	Cs134	604.72	Cs137	661.66

Table 2. Observed signatures considered for burnup characterization

4.1. Uncertainty due to Efficiency Corrections Across Energy Ranges

Thanks to recent advancements in microcalorimeter data processing, an energy calibrated spectrum can be exported from each individual detector component, referred to as pixels. Fifteen measurements taken from 5 Pu reference materials (CBNM series) were processed and a thorough downselection and energy calibration was performed to compare the efficiency curves from pixel to pixel. The SAPPY isotopic analysis software was used to analyze the spectra and estimate the efficiency curves using the well-known isotopic compositions. The full efficiency curves from each included pixel per measurement can be seen in Figure 6.

To capture the effect of using peaks farther away in energy space, each efficiency curve was normalized to the denominator of the ratios in Figures 7 and 8. When considering the choice of X-rays, the efficiency curves in this region are much more spread out at the Pu $K\alpha_1$ energy than the Pu $K\alpha_2$. Approximately 5.5x larger spread of the efficiency curve at the higher energy than the lower energy was observed. For comparison, the impact of spanning over 30 keV, as in the case of the Eu ratio, shows a spread in the efficiency curve 88x larger than the 1 keV span comparing the U $K\alpha$ and Pu $K\alpha_2$ peaks.

The interpretation and utilization of this finding is not straightforward in how this uncertainty would propagate. Since these pixels are eventually coadded, rather than averaged, to create the final spectrum and these are relative efficiency curves, the propagation is difficult. What can be interpreted is that ratios of peaks that have smaller energy differences are less impacted by variable efficiencies, and therefore are likely more reliable signatures.

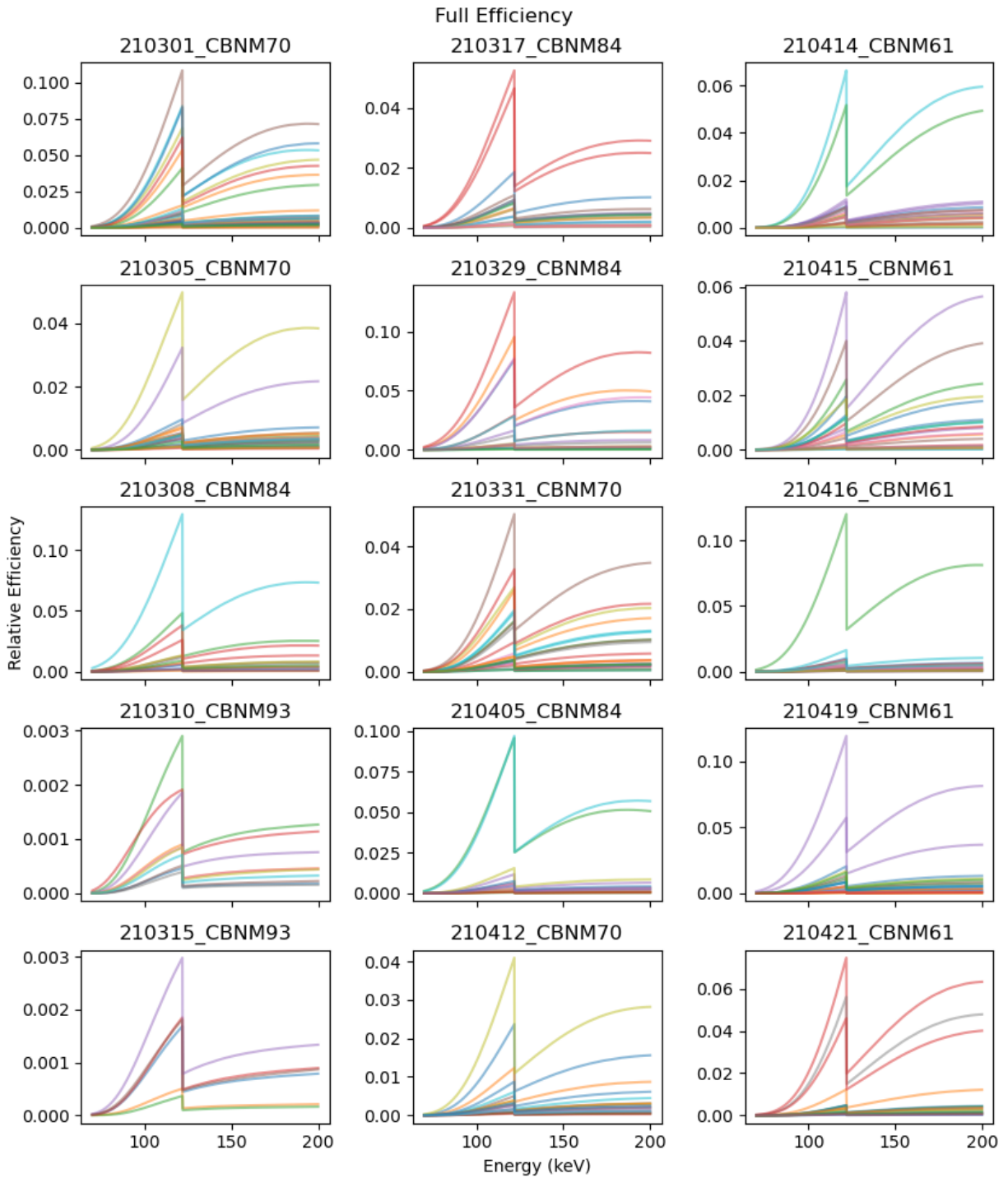


Figure 6. Spread observed in the full efficiency curves from the selected pixels measuring Pu reference standards (CBNM series). Pu spectra provide numerous peaks from which to calculate the curves.

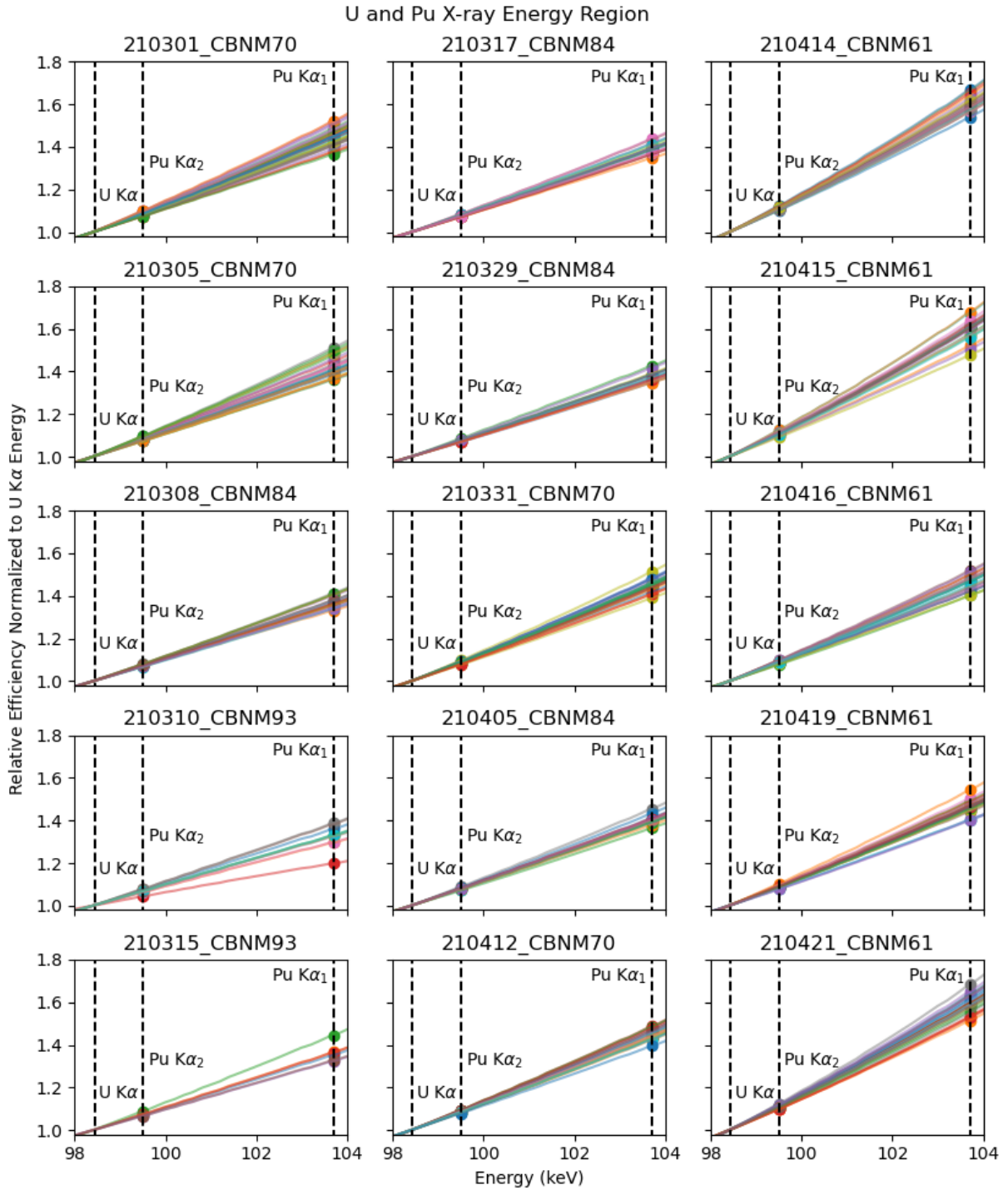


Figure 7. Normalized efficiency curves at the U $K\alpha_1$ X-ray at 98.44 keV. The energies of the Pu $K\alpha_2$ and $K\alpha_1$ (99.53 and 103.73 keV) are shown by the two rightmost dashed lines and the relative efficiencies normalized to the U $K\alpha_1$ are shown by closed markers.

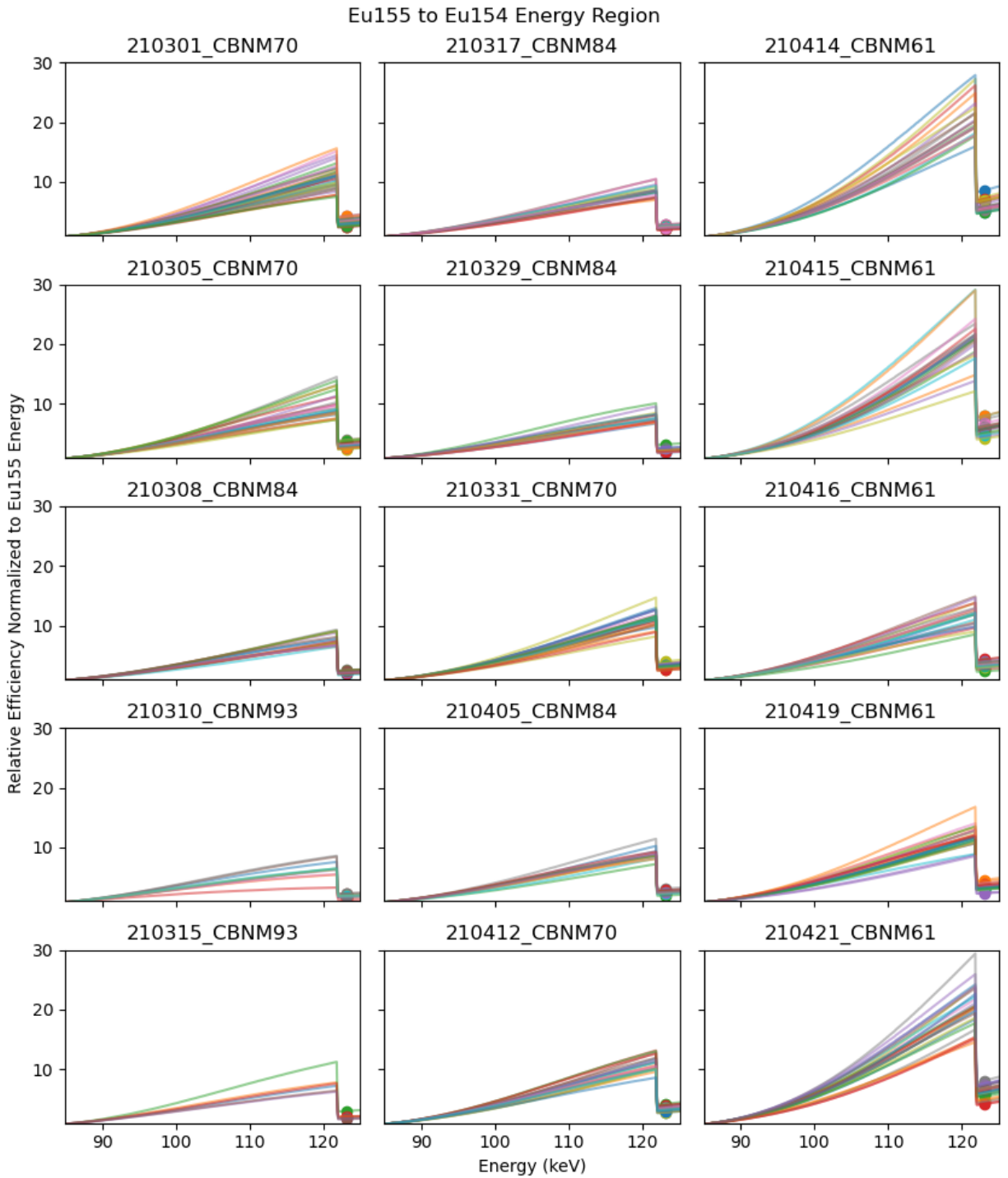


Figure 8. Normalized efficiency curves at the ^{155}Eu gamma ray at 86.55 keV. The relative efficiency at the energy of the ^{154}Eu gamma ray (123.07 keV) is shown by the closed markers.

4.2. Evaluating the four peak area ratios

Results of the four ratios in Table 2 from three irradiated TRISO fuel items are shown in Figure 9. Results from AGR2 items 2-211 and 2-542 and the AGR5 item 5-223 are plotted. Additionally, the KP223-50651 item was considered, however this item produced clear outliers from the other three for both the microcalorimetry ratios and the HPGe ratio. This is potentially due to different material properties, as the cooling time was not that dissimilar from the cooling time for the 5-223 item and the burnup was similar to both the 2-211 and 2-542 items.

The most linear relationship is the HPGe signature from the Cs isotopes, however this is not a direct measurement of the Pu content of the fuel. Use of the Cs ratio to determine burnup is likely to require calibration to a particular reactor type. When comparing the microcalorimetry signatures, the ratio using the Pu K α 2 falls in a more consistent pattern compared to the ratio using the Pu K α 1 peak. This supports the finding above that using signatures with peaks closer in energy space are more reliable. The nonlinear relationship between the Pu K α 2 and the U K α peak is likely due to the fact that ^{239}Pu fission becomes increasingly significant at high burnup and so the net Pu production rate decreases. The Eu isotope peak area ratio has the same nonmonotonicity.

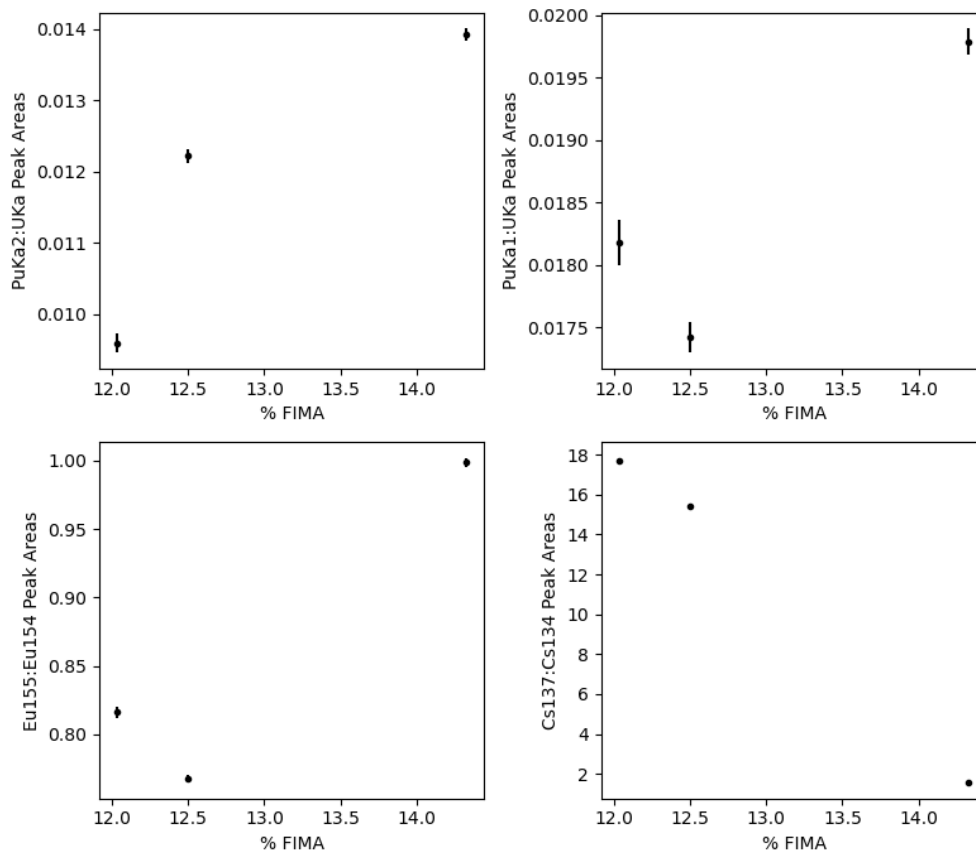


Figure 9. Peak area ratios of the four signatures described in Table 2. The two items with around 12% FIMA were irradiated during the same period (AGR2 items). Error bars indicate 1 standard deviation considering only the square root of the peak areas.

4.3. Simulating Shorter Microcalorimeter Spectra

The counting time between the existing SOFIA microcalorimeter spectrometer and HPGe is drastically different. Future microcalorimeter instruments will benefit from ongoing work to increase the speed of individual pixels and the number of pixels in the detector array. In an attempt to provide an early estimate of the number of counts needed to quantify the peak ratios well, the two AGR2 items are considered. This choice allows evaluation of the smallest difference in % FIMA with the same cooling time. From the spectra, Monte-Carlo methods were applied to simulate spectra with shorter measurement times, controlling the total number of counts in the spectrum. SAPPY was used to extract peak areas and plot the ratios and uncertainties in Figure 10.

From the previous finding the Pu $K\alpha_2$:U $K\alpha$ peak ratio has the closest relationship to burnup, therefore that is the most important signature to consider. Figure 10 shows that there are diminishing returns in the uncertainty in the ratio, however it does continue to decrease. After $6e7$ counts, there is reasonable separation between the 12.5% FIMA and 12.03% FIMA samples. Interestingly, the separation using the Pu $K\alpha_1$ and U $K\alpha$ peaks is not as pronounced. The Eu ratio is the most separable, however this ratio is observed to be only weakly correlated with burnup. The current SOFIA instrument is capable of operating at approximately 2000 counts per second and would require 8.3 hours to collect $6e7$ counts. A future instrument using a larger array and faster pixels (under development through other work), assuming 1000 pixels at 100 counts per second per pixel, would require 600 s of measurement time.

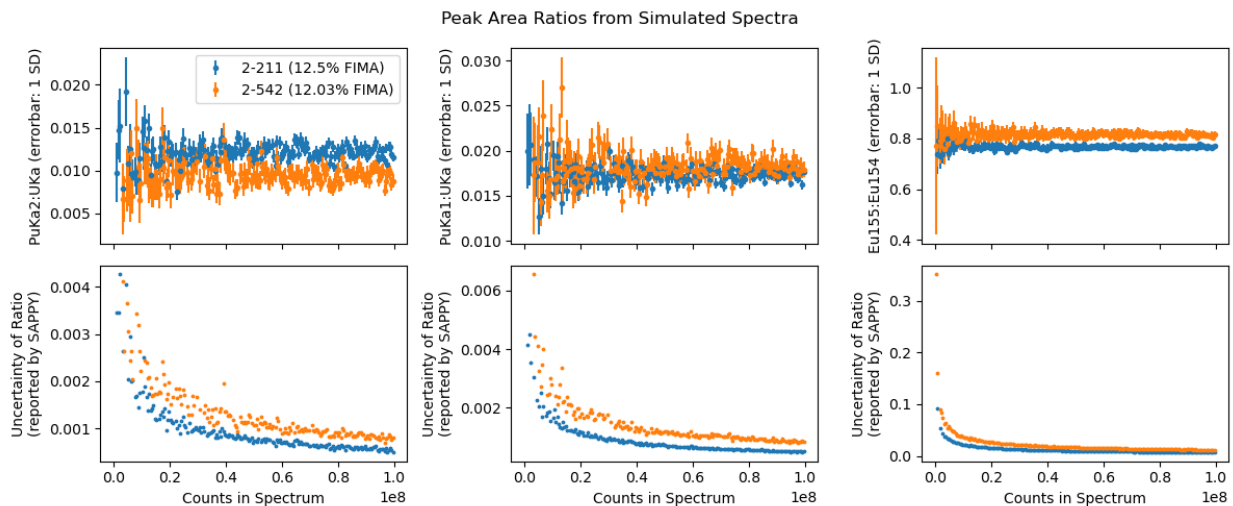


Figure 10. Results from simulated shorter measurement times on a microcalorimeter system. The top row of plots shows the peak area ratios and the bottom row of plots show the impact of uncertainty. It should be noted that the uncertainty considered here only considers the square root of the peak area and is not a final uncertainty.

5. CONCLUSIONS AND NEXT STEPS

The primary conclusions of this work are:

- Microcalorimetry and HPGe both have potentially useful signatures that are related to burnup. The highest quality microcalorimetry signature is the ratio of the Pu K α 2 X-ray to the U K α 1 X-ray. The Cs isotope ratio from HPGe spectra is also relevant, however it is not a direct measure of the Pu content of the irradiated fuel and requires additional information to reliably infer burnup especially between different reactor types.
- When considering peak ratios from a spectrum collected by a multipixel array, peak ratios with less separation in energy space are more reliable when using small peaks. One factor contributing to this is the uncertainty associated with the relative efficiency curve uncertainty.
- A microcalorimeter spectrum collected for the purpose of determining the U to Pu content in irradiated fuel with a similar background as the hot cell measurements would need approximately 6×10^7 counts. This value can be used when considering the development of a microcalorimeter gamma detector to get a sufficient measurement in a given time.

Two peak ratios stood out as being useful for characterizing irradiated fuel in available data. The primary question to answer in future work is whether this remains true for fuel that was very recently discharged from the reactor (or is being cycled through in the case of an online burnup measurement system), and for a wide range of fuel burnup. Measurements of TRISO fuel shortly after irradiation are planned for FY25. If the hypothesis that self-induced X-ray fluorescence is stronger for fuels with more intense short-lived fission products is validated, then this work could lead to a practical and robust method for nondestructively determining fissile material content of irradiated fuels.

6. REFERENCES

1. Reilly et al., *Passive Nondestructive Assay of Nuclear Materials*, NUREG/CR-5550, US Government Printing Office, 1991.
2. Hoover et al., *Measurement of Plutonium in Spent Nuclear Fuel by Self-Induced X-ray Fluorescence*, Proceedings of the Institute of Nuclear Materials Management Annual Meeting, 2009.
3. Petti D, P Martin, M Phélip, and R Ballinger, *Development of Improved Models and Designs for Coated-particle Gas Reactor Fuels*. INEEL/EXT-05-02615. Idaho National Engineering and Environmental Laboratory, Idaho Falls, Idaho, 2015.
4. Croce et al., *NDA of TRISO Fuels FY23 Report*, LA-UR-23-32266, 2023.