Gamma Spectroscopy Signatures of Irradiated Solid-Form TRISO Fuel

Prepared for
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SUMMARY

Nondestructive analysis (NDA) techniques are a major tool for material control & accountancy (MC&A) in nuclear facilities, as they are more efficient than destructive techniques, cheaper, and do not alter the material. Previous work has established NDA methods for quantifying the burnup of fuel used in traditional reactors using isotopic surrogates for uranium and plutonium. These signatures are evaluated for their relevance in advanced reactors, particularly pebble bed reactors that will use TRIstructrual ISOtropic (TRISO) fuel. In addition to evaluating the utility of established NDA signatures of burnup, there is an investigation into a new NDA method that can measure the actinide content directly. Microcalorimetry (μCal) can provide incredibly precise gamma spectroscopy measurements, with approximately 3x better energy resolution and sensitivity to lower energies compared to High Purity Germanium (HPGe) detectors. μCal and HPGe offer complementary measurement techniques with distinct strengths for measuring the burnup of TRISO fuel.

HPGe measurements are short in duration, with measurements taking on the order of minutes. μCal is a different class of gamma spectrometers and requires much longer measurements, on the order of hours for a sample with low enough activity to handle outside of a hot cell. HPGe is considered a high energy resolution gamma detector, while μCal is an ultra-high energy resolution gamma detector. The faster HPGe measurements can measure the ^{137}Cs and ^{134}Cs peaks above 600 keV, which can be used for burnup estimation of nuclear fuel. The longer μCal measurements can directly observe fluorescence x-rays from uranium and plutonium, even at the very low levels of plutonium expected for fuel irradiated to 100 GWd/MTU (< 3% Pu to U). Both the Cs peak ratio and the actinide x-ray ratios were evaluated to quantify burnup from NDA measurements.

Key Findings

The precision of burnup quantification increases with burnup, as shown in the figure below (Figure 14 in the report). Assuming a linear relationship between error and true burnup, HPGe can give an estimate of burnup within 5% error once the burnup exceeds 11.44% Fissions per Initial Metal Atom (FIMA). The more precise μCal signature of burnup was generally under 5% error even at low burnups (approximately 7.5% FIMA). Measuring freshly irradiated actinide targets, there were no identified short-lived fission products that would interfere with the fluorescence x-rays measured by μCal making this a viable NDA technique for PBRs.

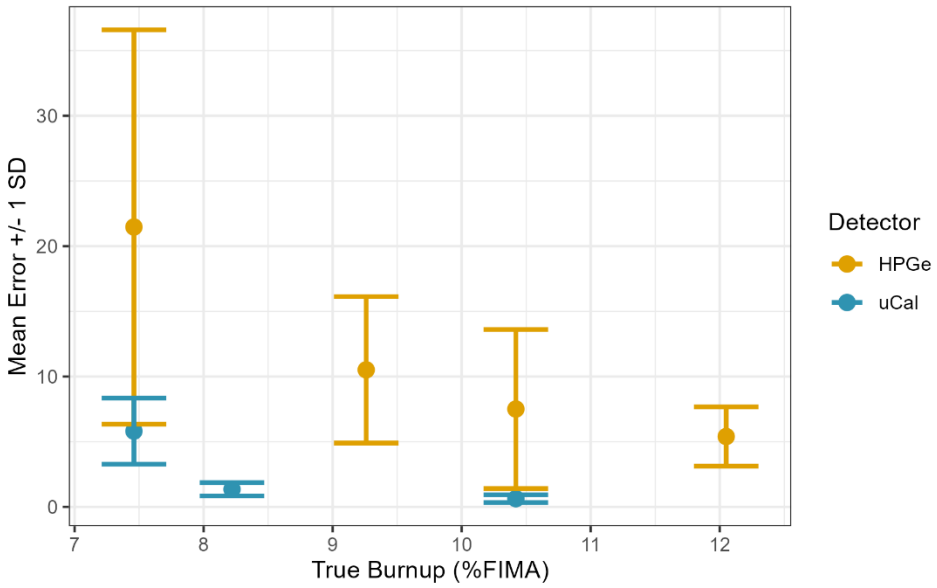

Figure 1. Absolute error as a percentage with a 1 SD error bar based on which NDA technique was used.

Discussion

A hypothetical PBR was discussed in Section 5.1 to illustrate the strengths of both NDA techniques and estimate the increased fuel efficiency when using both systems. This system was developed based on previous modeling efforts to estimate the burnup of a pebble as a function of the passes through the reactor and with conservative estimates for the NDA burnup quantification precisions.

While the cesium ratio measured with HPGe did have approximately an order of magnitude higher error than μCal, HPGe measurements are faster and more routine than μCal. For that reason, pebbles discharged from the reactor could be effectively monitored using HPGe in the first few passes through the reactor if the purpose of measuring the pebbles is to ensure they are safe to recycle. Once the exit burnup of a pebble starts to approach the average and/or maximum burnup, then the improved error of a μCal-based burnup quantification would result in additional fuel utilization.

If relying on HPGe only, TRISO pebbles could be confidently passed through the reactor about four times for a final exit burnup of 67.81 GWd/MTU. If μCal signatures of burnup were available, samples of pebbles could be assayed for the longer measurement time after the fifth pass. It is anticipated this would allow for three additional passes through the reactor, resulting in a final exit burnup of 95.6 GWd/MTU. Using μCal and HPGe together would create the most efficient NDA system and maximize the power generated from TRISO fuel.

1. INTRODUCTION

The goal of this work is to characterize nondestructive, gamma signatures of burnup in TRISO fuel to support the efficient use of fuel in advanced reactors and provide effective guidance for nuclear material control and accountancy (MC&A). This is accomplished through the direct evaluation of traditional and recently developed gamma spectroscopy techniques to identify peak ratios that trend with plutonium content relative to uranium in irradiated fuel.

Gamma spectroscopy is a well-established method for measuring irradiated nuclear fuel [1]. This is typically done by observing a surrogate fission product that has near equal fission yield with the primary fissionable material in fuel (^{235}U for pebble bed reactor fuel), a long half-live, and gamma emissions that have high energy and result in easily resolved peaks. The ratio of ^{134}Cs and ^{137}Cs was identified for measuring irradiated fuel due to the correlation with burnup in traditional fuel assemblies and the fact that it can be easily measured with a High Purity Germanium (HPGe) detector and other gamma methods [1].

Recent advancements in microcalorimetry (μCal) have led to the development of an ultra-high-resolution gamma spectrometry that can be directly integrated into a facility [2]. μCal has exquisite energy resolution up to 300 keV, which allows for the investigation of new isotopic ratios as a measure of burnup. This detector has been evaluated for nondestructive actinide characterization of irradiated TRIstructrual ISOtropic (TRISO) fuel, as well as Eu surrogate isotope ratios.

There are two direct applications of NDA for irradiated TRISO fuel. The first is to determine if a TRISO pebble can be recycled into the reactor. The decision to retire a TRISO pebble is determined by the burnup of the pebble, which is largely driven by the degradation of the physical integrity of the fuel and the chance of structural failure [3], as well as the isotopic composition of the fuel [4]. The second application of NDA for irradiated TRISO fuel is MC&A. Pebble Bed Reactors (PBRs) present challenges distinct from traditional light water reactors due to the fuel form factor and enrichment of the TRISO pebbles [5]. An accurate measurement of the actinide content of discharged TRISO pebbles is key to ensure that nuclear material is accounted for.

NDA has significant benefits over destructive analysis (DA) methods, like mass spectrometry, including not impacting the fuel itself, so the pebble can be recycled or discharged. Additionally, NDA is generally more cost-effective than DA methods. The most common NDA method to quantify isotopics in irradiated TRISO fuel is gamma spectroscopy. There are several gamma spectroscopy techniques of varying energy resolution that can be considered, but generally the higher the resolution, the lower the uncertainty on actinide quantification. Additionally, some techniques have complementary energy ranges for which they are most effective. A comparison of Cadmium Zinc Telluride (CTZ), High Purity Germanium (HPGe), and microcalorimetry (μCal) is shown in Figure 1.

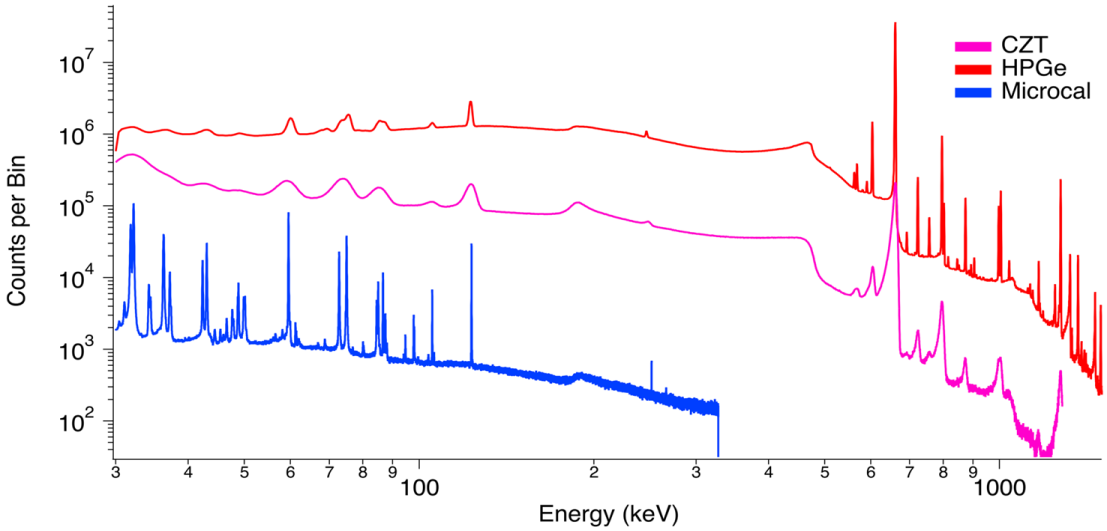

Figure 2. Gamma spectra from high-burnup light water reactor fuel measured by three different types of detectors with different energy resolutions.

HPGe and μCal have been explored in this body of work to understand how these technologies compare for estimates of burnup and actinide quantification. Measurements were taken of samples from irradiated TRISO fuel. The irradiations were part of the Advanced Gas Reactor (AGR) experiments at Idaho National Laboratory (INL). There were two different sample forms measured: TRISO compacts and TRISO particles. The TRISO compacts were measured in the hot cells at Oak Ridge National Laboratory (ORNL) while the particles were measured in a radiological lab at ORNL.

1.1. Advanced Gas Reactor Experiments

The AGR experiments were a series of high-temperature gas-cooled reactor (HTGR) experiments conducted at INL. The primary goal of the AGR program was to develop and demonstrate the technology for a new generation of advanced nuclear reactors that could operate at higher temperatures and provide increased safety and efficiency compared to traditional light-water reactors. The AGR experiments focused on various aspects of HTGR technology, including fuel development, materials testing, and reactor safety. The experiments involved the irradiation of TRISO fuel and materials in the INL's Advanced Test Reactor (ATR) to study their performance and behavior under high-temperature, high-radiation conditions.

Some of the irradiated TRISO fuel was transferred to ORNL for analysis, including intact compacts and individual particles. The compacts are restricted to handling in hot cells due to their activity; they were measured with both HPGe and μCal. The individual particles could be handled outside of hot cells and were measured simultaneously by the two gamma techniques.

The average burnup of each sample was been calculated from a sample of particles. These are approximate burnups, especially at the particle level, but provide a general estimate for the % Fissions per Initial Metal Atom (%FIMA). The samples reported

on here are summarized in Table 1. Throughout the rest of the analysis these will be referred to as the true burnups, despite this approximation.

Table 1. Characteristics of TRISO samples irradiated during the AGR experiments and measured by HPGe and μCal. (*) denotes compacts versus particles.

Series	ID	Calculated Burnup (% FIMA)	Irradiation Dates	Number of Particles	Number of Measurements
AGR2	2-633	7.46	6/22/10-10/16/13	1	3
AGR2	2-623	8.22	6/22/10-10/16/13	2	3
AGR2	2-642	9.26	6/22/10-10/16/13	1	3
AGR2	2-523	10.42	6/22/10-10/16/13	1	4
AGR2	2-542*	12.03	6/22/10-10/16/13	2858	1
AGR2	2-541	12.05	6/22/10-10/16/13	1	4
AGR2	2-211*	12.50	6/22/10-10/16/13	3176	1
AGR5/6/7	5-223*	14.33	2/16/18-7/22/20	3176	1

1.2. NDA Signatures

As can be seen in Figure 1, HPGe and μCal are sensitive to different energy ranges. μCal has exquisite energy resolution, approximately 0.06% full width at half maximum (FWHM) at 123 keV and is sensitive up to approximately 300 keV. HPGe has the highest resolution of the traditional gamma spectrometers in Figure 1. The HPGe used in this work has approximately 0.22% FWHM at 3.5 MeV and can measure photons up to 6 MeV.

The traditional isotopic signature of burnup is the ratio of ^{137}Cs to ^{134}Cs [1] with peaks at 661.66 keV and 604.72 keV, respectively. This isotopic signature is being evaluated for TRISO fuel. The cesium peaks are not directly observable with μCal but are easily observed and resolved in HPGe. Instead, the sensitivity to lower-energy photons of μCal allow for the direct measurement of fluorescence x-rays from uranium and plutonium. The x-rays would provide a direct measurement of the actinide content; however, it is insensitive to isotopics. Both the cesium and the x-ray signatures could be leveraged for burnup calculations as pebbles come out of the reactor while the x-ray signature could also be used for MC&A purposes.

2. IRRADIATED TRISO FUEL MEASUREMENTS

Both the compacts and the particles at ORNL were measured with HPGe and μCal across two years. In both cases, the material had been cooled for over several years, so the utility of these signatures for freshly irradiated fuel needs to be further validated.

2.1. Measurements of TRISO Compacts

Since the compacts, and subsamples of the compacts, had to be handled inside ORNL hot cells at the Irradiated Fuel Examination Laboratory, it was not feasible to measure a sample simultaneously with the two techniques. The μCal measurements were taken with the SOFIA (Spectrometer Optimized for Facility Integrated Applications) μCal gamma system, while the HPGe measurements were taken with a Canberra GL1015 HPGe. Aligning the sample with the collimator/port as well as the μCal detector proved difficult, as the μCal has a window that is flush with the detector, compared to the HPGe where the crystal protrudes from the detector body. Additional handling considerations working in the hot cells contributed to the difficulty in taking measurements and precluded repeat measurements. However, there were successful HPGe and μCal measurements made of three compact-type samples with burnup ranging from 12% to 14%.

The signatures of interest for the two systems, the Cs ratio for HPGe and the x-ray ratio for μCal, are shown in Figure 2. There is a strong linear relationship between the cesium ratio and burnup (R^2*=0.996, t(1) = -16.177, p-value = 0.039*), supporting that the cesium ratio would make a statistically strong and straightforward estimator of burnup for TRISO compacts. The relationship of the x-ray ratio and burnup is not a strong, linear trend (R^2*=0.809, t(1) = 2.057, p-value = 0.288*), however the curvature does follow the expected plutonium content as a function of irradiation [6]. This provides support for use of μCal for MC&A, as it trends with the expected actinide content. The x-ray ratio could also be used for burnup calculations, however the functional form of this relationship required specification and validation with more data. Full analysis and details of this measurement campaign can be found in [7]. To validate the concave shape of the x-ray and burnup relationship, repeated measurements with more samples was required. Due to the extraordinary measurement setup, taking replicate measurements was not feasible in the hot cells. This motivated the move into the Safeguards Extension Lab (SEL) at ORNL where individual particles were measured.

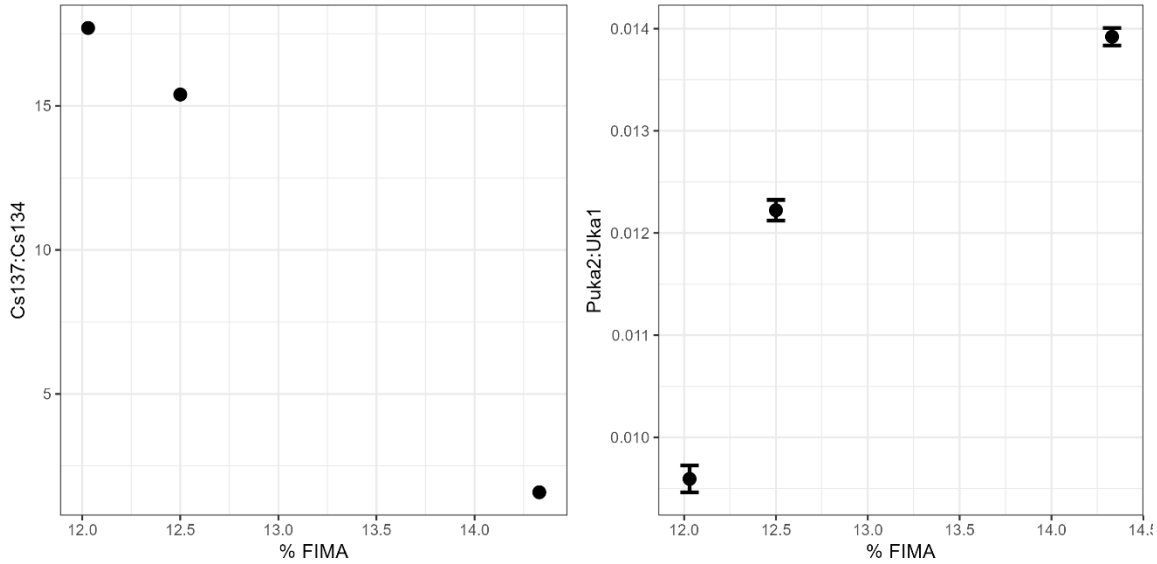

Figure 3. The HPGe isotopic signature of burnup (% FIMA) and the x-ray signature measured by μCal, comparing the Pu $K\alpha_2$ and U $K\alpha_1$ peaks.

2.2. Measurements of TRISO Particles

A major advantage to measuring samples in the SEL lab is the ability to take simultaneous HPGe and μCal measurements. SOFIA has lower detector efficiency than the HPGe, so sample placement was optimized for SOFIA while the HPGe was positioned relative to the sample and SOFIA. In some measurements the outer container holding the particles was affixed to SOFIA to align the sample with the window (Figure 3) and other times the sample was placed on a raised stage (Figure 4).

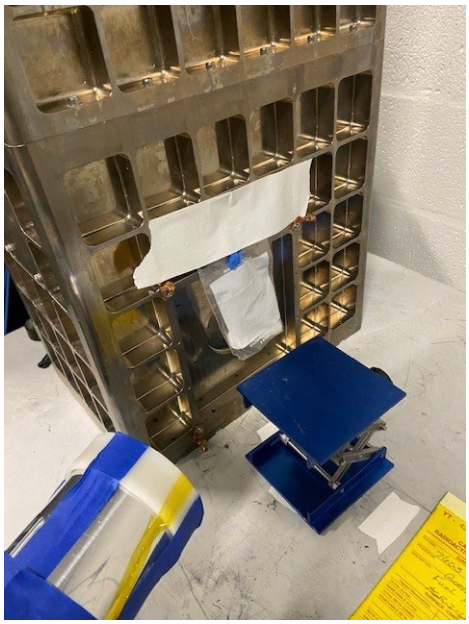

Figure 4. An example of when the sample is placed against the SOFIA detector window and taped in place. The HPGe snout is pictured in the lower left-hand corner.

Figure 5. An example of when the sample is placed on a raised stage to align the sample with the SOFIA detector window. The HPGe snout is pictured to the left.

Samples were measured with SOFIA for an average of 22 hours and resulted in a spectrum of an average of 68 million counts. The average count rate on SOFIA was 900 counts per second (CPS). The HPGe measurements took approximately 10 hours per sample, with an average total count of 136M. The average count rate for the HPGe was 3800 CPS.

Simultaneous measurements produced true measurement pairs of the low-energy, ultra-high-resolution SOFIA detector and the higher-energy, high-resolution HPGe detector. An example of this measurement pair can be seen in Figure 5.

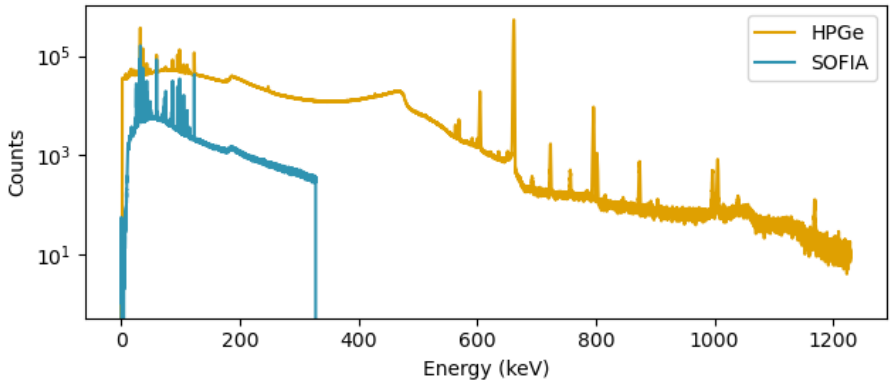

Figure 6. HPGe and SOFIA spectra taken simultaneously from Item AGR2-541 (12.05% FIMA).

The Cs ratio and the fluorescence x-ray signatures in the SOFIA spectra seen in the compact samples were also observed in the particle samples. HPGe spectra of different burnups are shown in Figure 6, where the ^{134}Cs peak is at 604.72 keV and the ^{137}Cs peak is at 661.66 keV. The minor plutonium x-rays can be seen in Figure 7, where the spectra have been normalized to the uranium Kα x-ray at 98.44 keV and offset vertically by a consistent amount to visually compare the spectra.

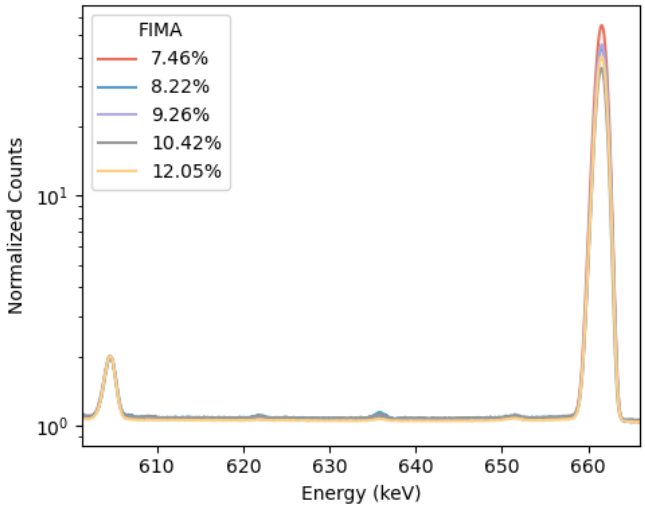

Figure 7. HPGe spectra of five items with different burnups. Spectra have been background-subtracted and normalized to the 604 134Cs peak. The Cs signature is a ratio of the 661 keV peak (^{137}Cs) to the 604 keV peak (^{134}Cs).

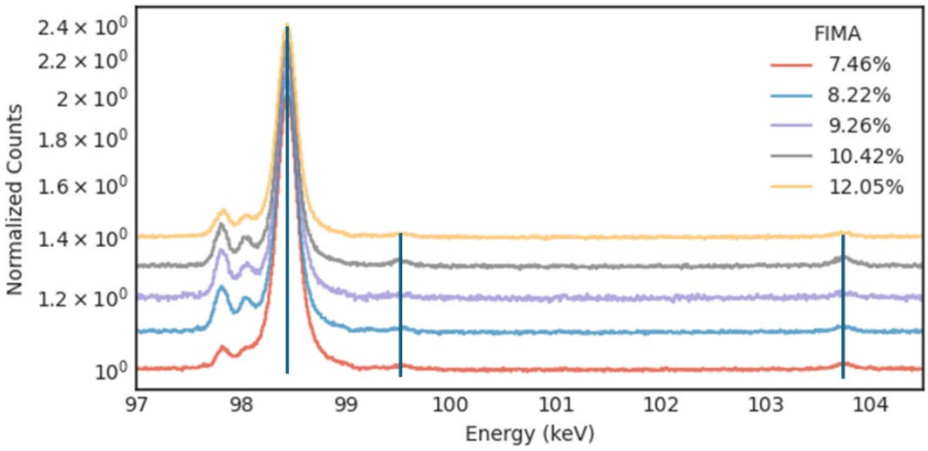

Figure 8. Plutonium x-rays observed in SOFIA spectra. All spectra are normalized to the U Ka at 98.44 keV and offset to show minor differences. The plutonium x-rays are demarked by solid vertical lines at 99.52 keV and 103.73 keV. The Pu $K\alpha_2$ peak is used so there is less of an uncertainty contribution from energy-dependent efficiency corrections.

The peak ratios from the HPGe and μCal spectra are used to estimate how strongly of a signature these are for burnup. A more quantitative approach can be taken with the particle samples due to repeated measurements; however, the burnup estimates are poorer at the particle level compared to the compact level.

3. Burnup Quantification from NDA Signatures

As established from the compact measurements, the cesium signature (HPGe) was expected to follow a strong, linear trend while the actinide x-ray signature (μCal) requires a functional form to be specified. Both methods are explored to calculate burnup.

To calculate the average error between the burnup in Table 1 and the estimated burnup from NDA signatures, a leave-one-out cross-validation method was used. Using the specified trend, regression parameters are estimated using least squares optimization from all measurements except for one, the *left-out* measurement. Once the optimal regression parameters are leaved, the left-out measurement was evaluated using the learned trendline to estimate the burnup of this measurement. This method provides an error between the true and estimated burnup. The average and standard deviation of the absolute error are calculated and reported. It should be noted that only the top-down uncertainty on error is reported; the uncertainty in the regression parameters is highly dependent on the number of measurements used to learn the trendline and, therefore, does not directly apply to this method as it would be used in an operational setting.

3.1. Burnup Quantification with HPGe

The cesium ratios from particle measurements in the SEL are shown in Figure 8. Across this dataset there was a moderately strong, linear relationship (R^2=*0.672, t(18)=-6.075, p < 10^{-4})*, however there was a clear outlier for one of the measurements of the 8.22% FIMA item. Both measurements from this item were excluded from analysis. It should be noted that this is the only particle sample with two particles, rather than one, so the discrepancy may have been due to larger variability in the geometry of the sample with respect to the HPGe. After excluding both 8.22% FIMA measurements, there was a stronger linear relationship (R^2=*0.751, t(13)=-6.524, p-value<10^{-4}*).

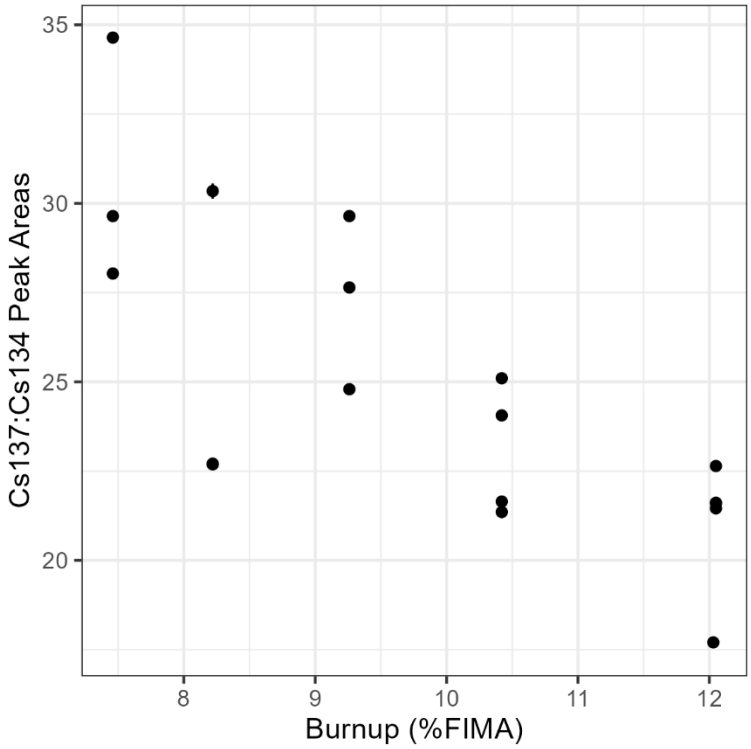

Figure 9. Cesium ratios plotted against burnup for HPGe measurements of irradiated TRISO particles. For further analysis, the 8.22% FIMA item was excluded due to the outlier. Error bars are plotted, however in all cases except for one they are smaller than the marker.

The regression analysis was carried out using a simple linear formula, regressing burnup against the observed ratio:

$$B = \beta_0 + \beta_1 r$$

Where:

- B is the burnup in % FIMA
- r is the cesium ratio (^{137}Cs:^{134}Cs)
- β_0, β_1 are the learned regression coefficients.

The results of the leave-one-out error estimation are shown in Figure 9. Each individual measurement is represented by a gray bar with the black bar showing what the calculated burnup is. The percentage annotations above each estimated burnup is the percent error between the estimation and the calculated burnup. Across the entire dataset there was not a significant bias in the burnup calculations determined by the residuals between the estimated burnup and the true burnup (*t(13) = -0.206, p-value=0.840*).

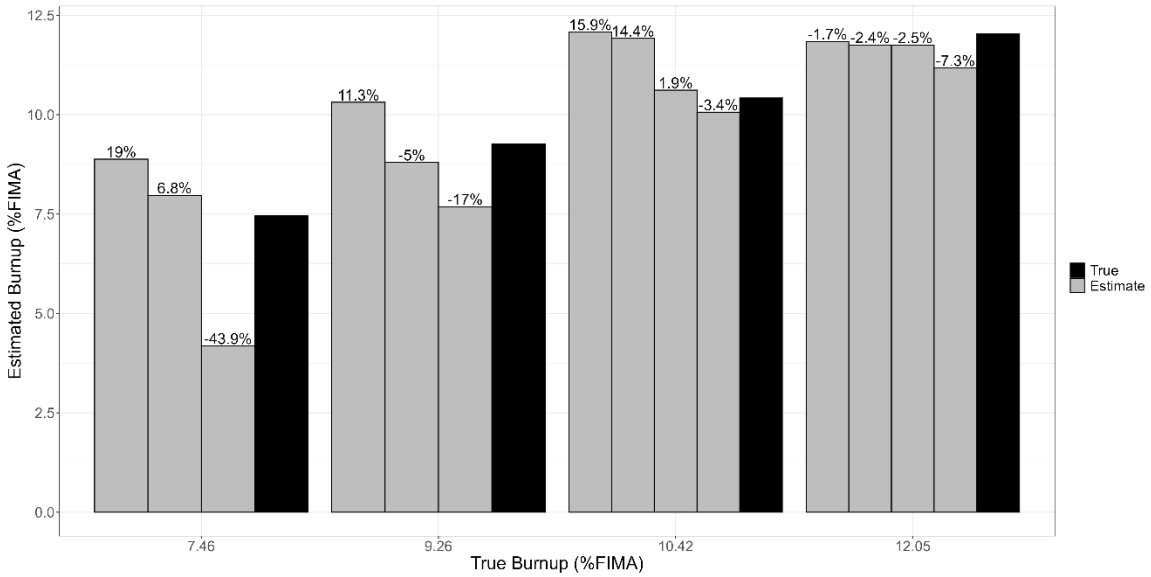

Figure 10. Estimated burnup from the cesium ratio using the leave-one-out scheme to estimate the error. The annotations above each bar indicate the error between the true burnup and the NDA-estimated burnup.

There was a significant trend between the absolute value of the error as a percentage and the calculated burnup (R^2*=0.394, t(12)=-2.796, p-value=0.0162*). This trend is shown in Figure 10. Based on the linear regression, it is expected that the HPGe cesium ratio can give an estimate of burnup within 5% error once burnup exceeds 11.44% FIMA.

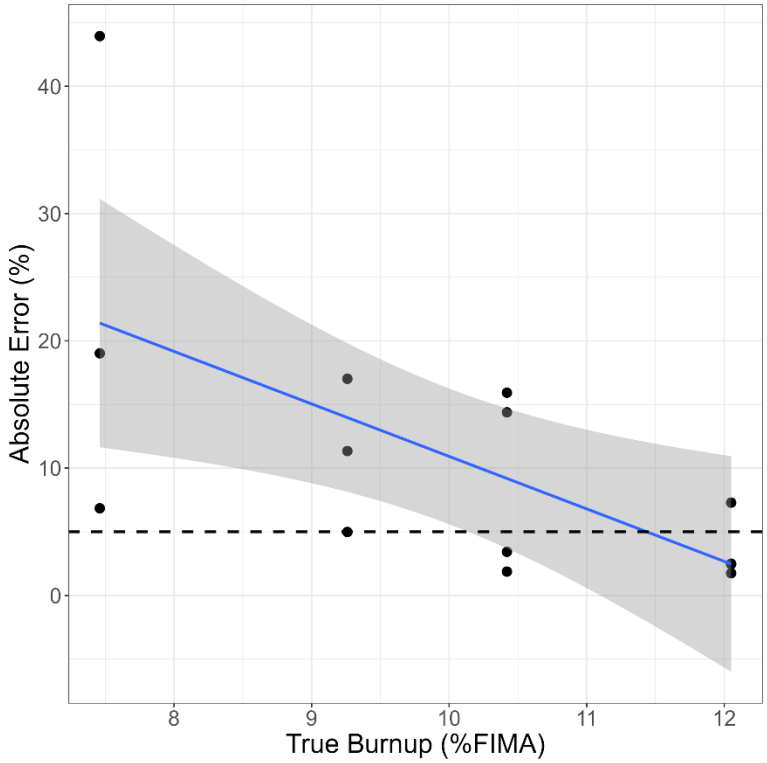

Figure 11. The trend between HPGe-based burnup quantification as a function of the burnup of the item. The blue, solid line shows the fit linear regression, and the dashed line shows an arbitrary 5% target for accuracy.

3.2. Burnup Quantification with μCal

The x-ray fluorescence ratios from particle measurements in the SEL are shown in Figure 11. μCal measurements are more complex and require significant data processing. There were data processing and fitting issues with measurements from one of the 7.46% FIMA measurements and all of the 9.26% FIMA item that are being investigated. From Figure 11 the 12.05% FIMA item also appears to be an outlier or at least does not follow the expected monotonic pattern of plutonium content as a function of burnup. The measurements from the 12.05% FIMA sample are excluded. It should also be noted that a scaling factor of 100 was applied to the x-ray ratio for parameter optimization efficiency.

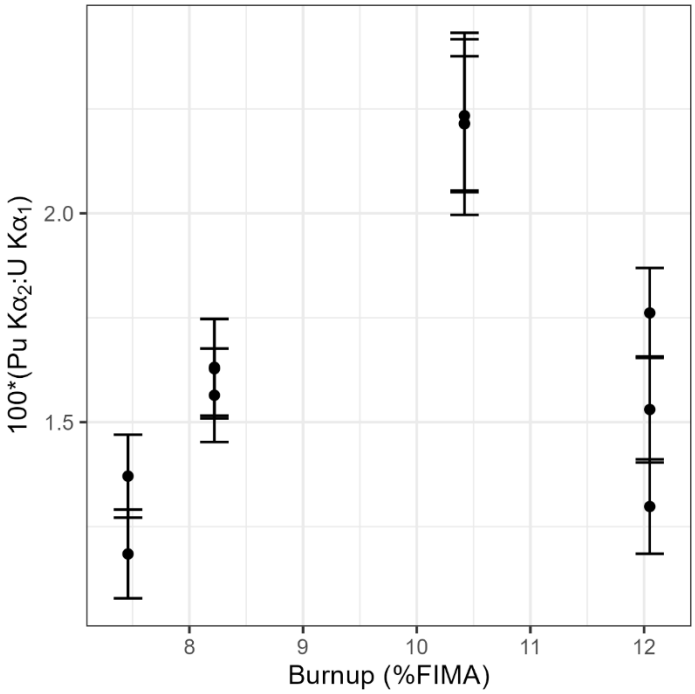

Figure 12. Fluorescence x-ray ratios plotted against burnup for μCal measurements of irradiated TRISO particles. The 12.05% FIMA item was excluded from analysis due to the outlier. Error bars show 1σ.

The regression analysis for the μCal measurements followed a nonlinear trend, communicated here as the ratio regressed against the burnup:

$$r = \frac{p}{q}(1 - e^{-qB}) + C$$

Where:

- B is the burnup in % FIMA
- r is the x-ray ratio expressed as a percentage (100*Pu $K\alpha_2$:U $K\alpha_1$)
- p is a learned term, relating to the production of Pu
- q is a learned term, relating to the decay of Pu
- C is a learned term for the vertical offset

It should be noted that this model contains one more parameter than the linear regression used for HPGe and is estimated on fewer data points. Additionally, the parameters were estimated using the relationship of burnup against the x-ray ratio, but the inverse equation was used to calculate burnup on the left-out measurement.

The results of the leave-one-out error estimation are shown in Figure 12. Across the entire dataset there was not a significant bias in the burnup calculations determined by the residuals between the estimated burnup and the calculated burnup (*t(7) = -0.340, p-value=0.840*).

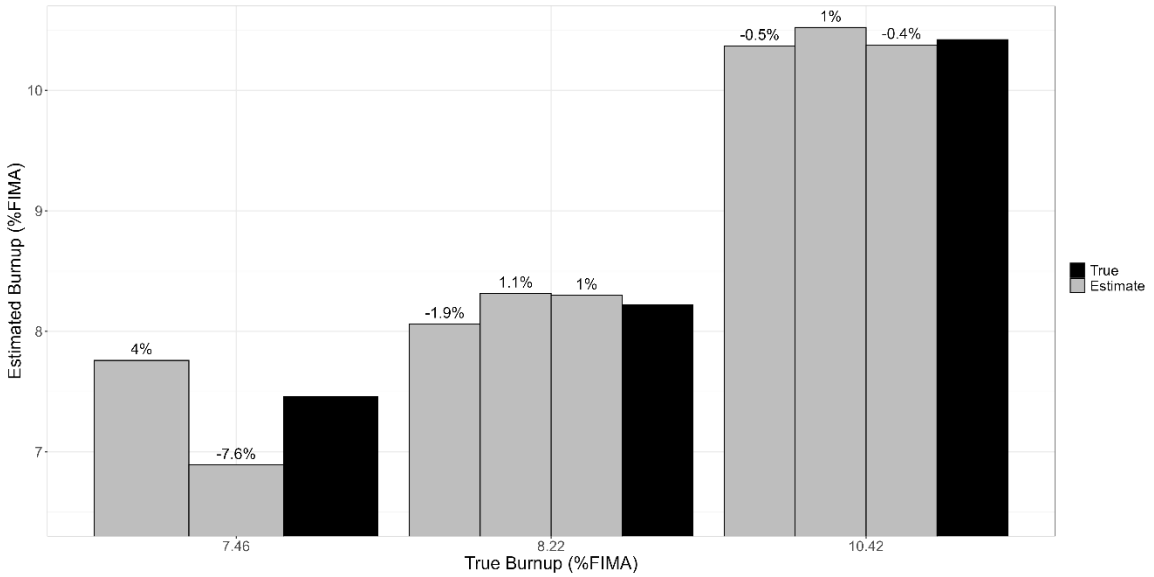

Figure 13. Estimated burnup from the x-ray ratio using the leave-one-out scheme to estimate the error. The annotations above each bar indicate the error between the calculated burnup and the NDA-estimated burnup.

There was not a significant trend between the absolute value of the error as a percentage and the calculated burnup (R^2*=0.471, t(6)=-2.31, p-value=0.0603*). Instead, there was a convex relationship between absolute error as a percentage and the true burnup, as seen in Figure 13. While there was not a linear trend, nearly all of the measurements were within 5% error.

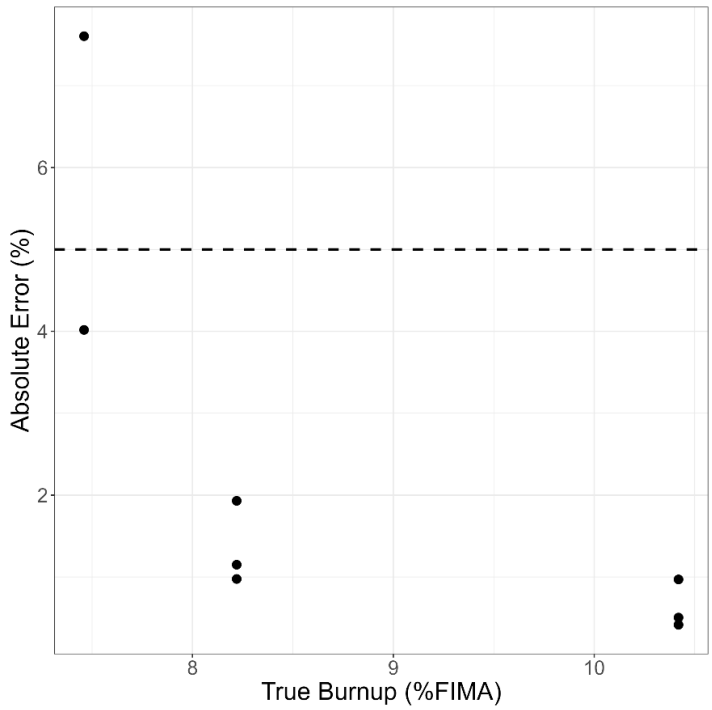

Figure 14. The trend between µCal-based burnup quantification as a function of the burnup of the item. The dashed line shows an arbitrary 5% target for accuracy

3.3. Comparison of HPGe and µCal Burnup Quantification

Using the absolute percent error calculated above, the HPGe and µCal-based burnup quantifications can be directly compared. The average and top-down, statistical variation in the absolute percent error is shown in Figure 14. The statistical variation does not account for the tighter error bars for the HPGe peak ratios compared to the µCal peak ratios. To fully understand the uncertainty budget, a stricter calibration curve should be fit such that the uncertainty in the parameter estimates can be propagated through. Such work is not presented here, as the calibration curves were estimated from a few measurements at only three or four different burnups. However, if this method continues to be validated in preparation for deployment in an operating advanced reactor, there would be ways to improve the parameter estimation rather than using a direct least-squares approach.

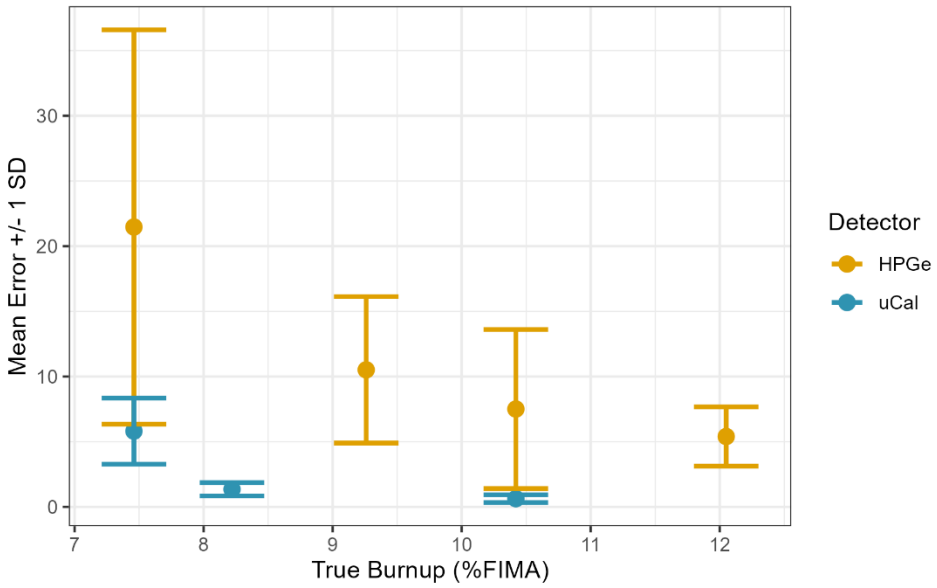

Figure 15. Absolute error as a percentage with a 1 SD error bar based on which NDA technique was used.

In all cases, the error in burnup calculation was lower using µCal compared to HPGe. However, a larger uncertainty in the burnup calculation would likely be acceptable for lower burnup items. This could inspire a triaged approach to NDA measurements on irradiated TRISO fuel in a facility that is discussed further in Section 5.

3.4. Comparison of Compact and Particle Measurements

The relationships explored above were only applied to measurements of irradiated TRISO particles, as that was the form factor that allowed for repeat measurements. However, the applicability of the methods should be validated across both particle and compacts.

For the HPGe measurements, the particle and compact measurements are shown in Figure 15. For all of these measurements, the cesium ratio follows a strong, linear

relationship with burnup ($R^2 = 0.67$, $t(18) = -6.075$, $p\text{-value} < 10^{-5}$). The high energy of these gamma emissions decreases the effect of attenuation, showing a consistent pattern between the compact-form samples and the particle measurements.

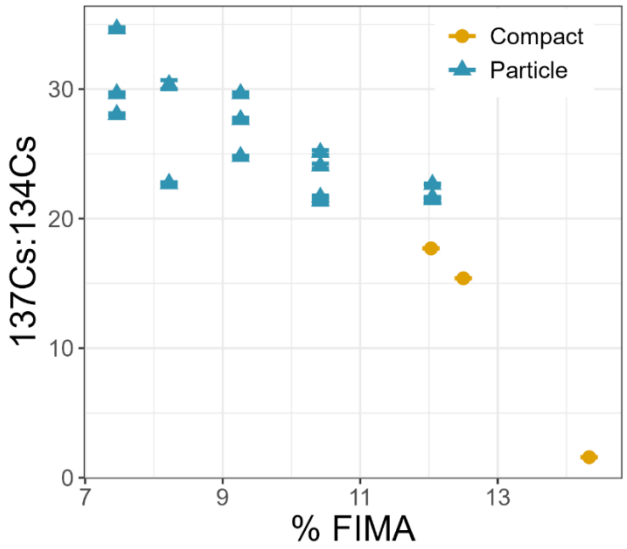

Figure 16. The relationship between the cesium peak ratio, without any decay corrections, and the burnup (% FIMA) is strong and linear.

The relationship between burnup and the x-ray peak ratios does not follow a consistent pattern and is instead impacted by the sample form factor, as shown in Figure 16. However, both types of measurements show the concave pattern, anticipated based on the plutonium content as a function of burnup. The x-rays used for this signature are much lower energy compared to the cesium isotopic ratio, leading to a larger impact of attenuation and the sample's physical form. This explains the similar shape, but on different scales. If a calibration curve method is used, as carried out above, the calibration standards would need to be of the same physical form of operational fuel and/or the translation from particle to compact would need to be better understood and validated.

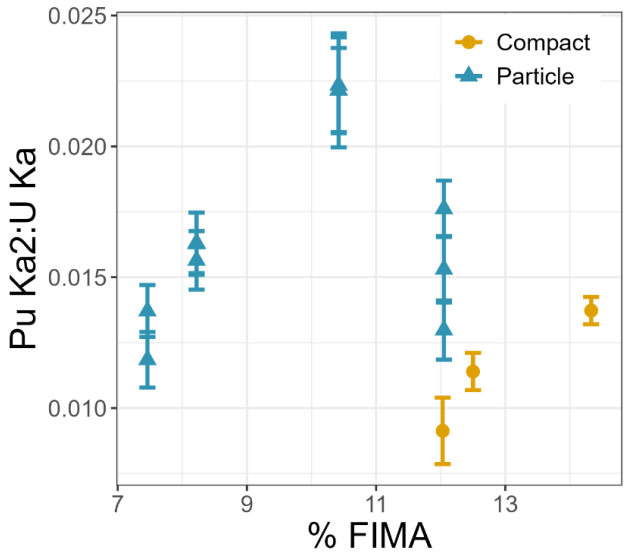

Figure 17. The relationship between the fluorescence x-rays matches the plutonium production as driven by burnup. However, the lower energy peaks are more affected by attenuation and the physical form of the sample.

4. IRRADIATION EXPERIMENTS AT HFIR

After establishing the utility of fluorescence x-rays, we considered whether there would be any interfering emission lines present in freshly irradiated TRISO fuel. To investigate this, we leveraged the capabilities at the Neutron Activation Analysis laboratory at HFIR to irradiate samples of uranium, plutonium, and mixtures of high assay low enriched uranium (HALEU) and plutonium. Samples were prepared and irradiated in the high flux reactor and measured by both HPGe and SOFIA between 30 and 45 seconds post-irradiation.

4.1. SAMPLE INFORMATION

We ran two measurement campaigns at HFIR. During the first campaign we irradiated pure samples of natural uranium and ^{239}Pu. During the second campaign we irradiated more complex mixtures of actinides that closely mimic the uranium and plutonium content of irradiated TRISO fuel. The sample contents are shown in Table 2. These samples were irradiated for either 90 seconds in the first campaign or 60 seconds in the second campaign to achieve similar activity samples. The difference in irradiation times were to account for the different flux of the reactor.

Table 2. Samples irradiated at HFIR, created from depositing liquid solution and allowing it to dry. Samples were encased in a plastic rabbit and irradiated in pneumatic tube 2.

Sample ID	Isotope	^{238}U Weight (ng)	^{235}U Weight (ng)	^{239}Pu Weight (ng)	Enrichment (%)	% Pu Content
DX11	Nat U	21120.5	153.1		0.725	
DX12	Nat U	21185.4	153.6		0.725	
DX13	Nat U	21096.9	153.0		0.725	
DX14	Nat U	21244.3	154.0		0.725	
DX15	Nat U	20955.3	151.9		0.725	
DX16	Nat U	21055.6	152.7		0.725	
D21	Pu-239			102.9		
D22	Pu-240			104.7		
D23	Pu-241			103.3		
D24	Pu-242			103.7		
D25	Pu-243			102.6		
D26	Pu-244			104.8		
EW-15	Mixture	662.4	159.3	4.15	24.0	0.51
EW-16	Mixture	668.9	160.9	4.15	24.0	0.50
EW-17	Mixture	668.2	160.7	8.18	24.0	0.99
EW-18	Mixture	668.0	160.6	8.22	24.0	0.99
EW-19	Mixture	667.8	160.6	12.31	24.0	1.49
EW-20	Mixture	674.2	162.1	12.33	24.0	1.47
EW-21	Mixture	668.2	160.7	16.47	24.0	1.99
EW-22	Mixture	701.0	162.1	16.53	23.1	1.92
EW-23	Mixture	912.5	159.7	21.32	17.5	1.99
EW-24	Mixture	924.3	161.6	21.44	17.5	1.97
EW-26	Mixture	925.8	161.4	21.73	17.5	2.00

4.2. MEASUREMENT CONDITIONS

Samples were placed in a plastic holder directly in front of SOFIA's window. The HPGe had been placed to optimize the field-of-view of the detector, while not creating obstacles for handling high-activity samples. The typical counting time was approximately 45 to 60 minutes long. Initial count rates for SOFIA were on the order of 5-6K CPS while after about 40 minutes the count rate was below 50 CPS. SOFIA spectra have an average of 1.8 million counts per measurement. HPGe measurements were taken simultaneously and had count rates on the order of 13k CPS immediately post-irradiation and 400 CPS after 60 minutes.

4.3. RESULTING SPECTRA

The gamma measurements for the freshly irradiated samples were incredibly dynamic, as we can directly observe short-lived fission products dying away. SOFIA

provides list-mode data, which allowed us to visualize these changes directly in Figure 17. The blue dots indicate individual photons observed at the time shown on the x-axis and with the energy shown on the y-axis. The coadded spectrum is shown along the y-axis. The energy cutoff for the plot is 100 keV to illustrate the low-energy, short-lived fission products, however the full SOFIA energy range is captured in the spectra.

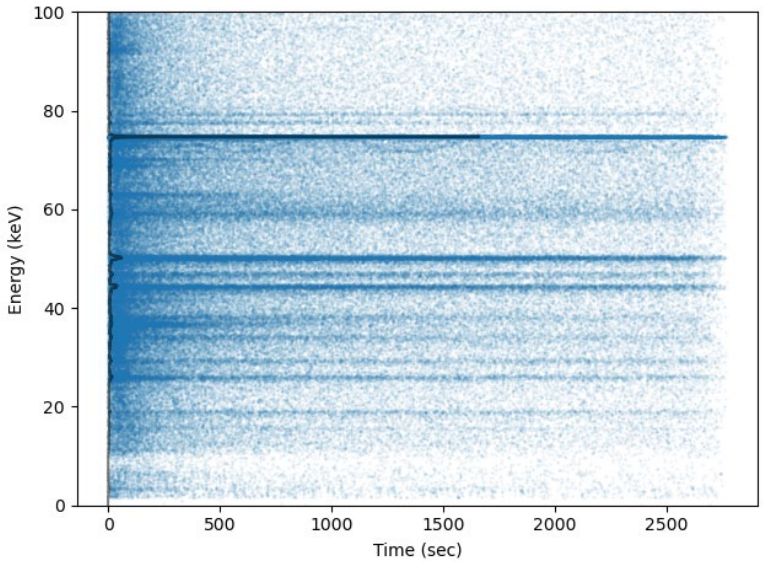

Figure 18. A scatterplot showing the dynamic nature of spectra measured from freshly irradiated samples. This shows the measurement taken from irradiating a natural uranium sample for 90 seconds. The coadded spectrum is shown along the y-axis.

4.4. FISSION-PRODUCT ANALYSIS

SOFIA's list-mode data allow for half-life calculations, by chunking the spectra into arbitrary time bins. Similarly, this can be performed with the HPGe data, as the summed HPGe spectrum is a result of coadding spectra taken for 30 seconds at a time. This provides a unique advantage for energy calibration and peak identification. The prominent peak shown in Figure 17 has been identified as ^{239}U at 74 keV based on the prevalence and a half-life estimate of 1422 ± 18 seconds, determined by the relationship between counts in that peak and measurement time, shown in Figure 18. This peak identification is supported by additional, minor ^{239}U peaks at 44.2 keV and 18.4 keV with similar half-life estimates.

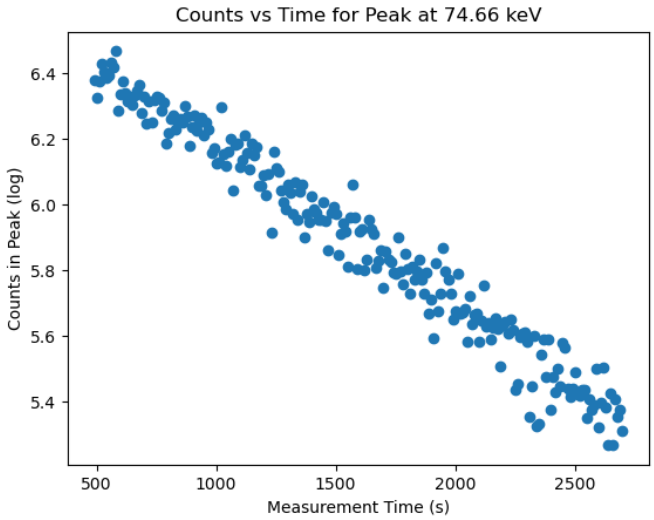

Figure 19. Counts in the 74.66 keV peak by measurement time. The counts are on a log scale. From this strong, linear relationship (R^2= 0.965) we estimate a half-life of 1422.4 seconds ± 18.4 seconds.

There is shared structure in the HPGe and SOFIA spectra, shown in Figure 19, that allows us to confirm the energy calibration of both spectra for the natural uranium samples.

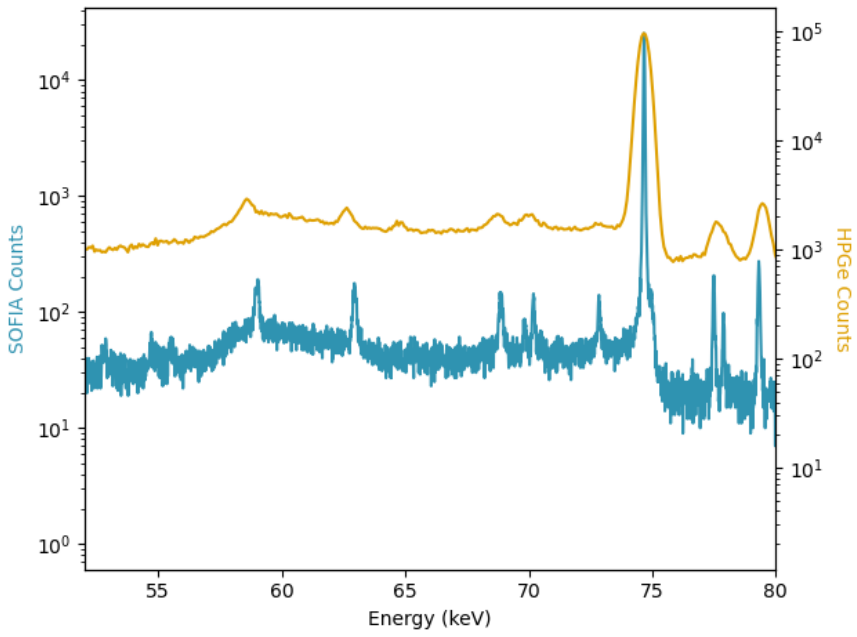

Figure 20. Similar structure in the Compton background between 55 and 75 keV as measured by HPGe (orange) and SOFIA (blue). This provides further confidence in the energy calibration and assists in peak identification.

With energy calibrated spectra, we can investigate if there are any short-lived fission products in the 90-100 keV region that would interfere with fluorescence x-rays as a signature of burnup. The SOFIA uranium spectrum was chunked by time and energy bin in Figure 20. The region of interest around 90-100 keV looks relatively "quiet" after about 100 seconds, indicating that as long as an irradiated TRISO pebble has 100 seconds of cooling time, there would not be a significant interference from ^{238}U fission products. This would not pose any difficulty, as TRISO pebbles are typically cooled for approximately 100 hours before being discharged from the reactor.

The ^{239}Pu and mixture samples resulted in spectra with many more fission products, which require additional attention to process the μCal data. Algorithmic advancements have been and continue to be developed in support of getting the highest resolution and statistics spectra from these measurements. An additional challenge with the current SOFIA data processing pipeline is the requirement to energy calibrate each measurement independently. This is also being addressed by recent technological advances. However, the HPGe energy calibration is much less sensitive and can be presumed dependent between measurements. By leveraging the ^{239}U peak in the sample of the natural uranium, we energy calibrated the spectra from the Pu and mixture samples. The 2D spectra from these measurements are shown in Figure 21 and Figure 22.

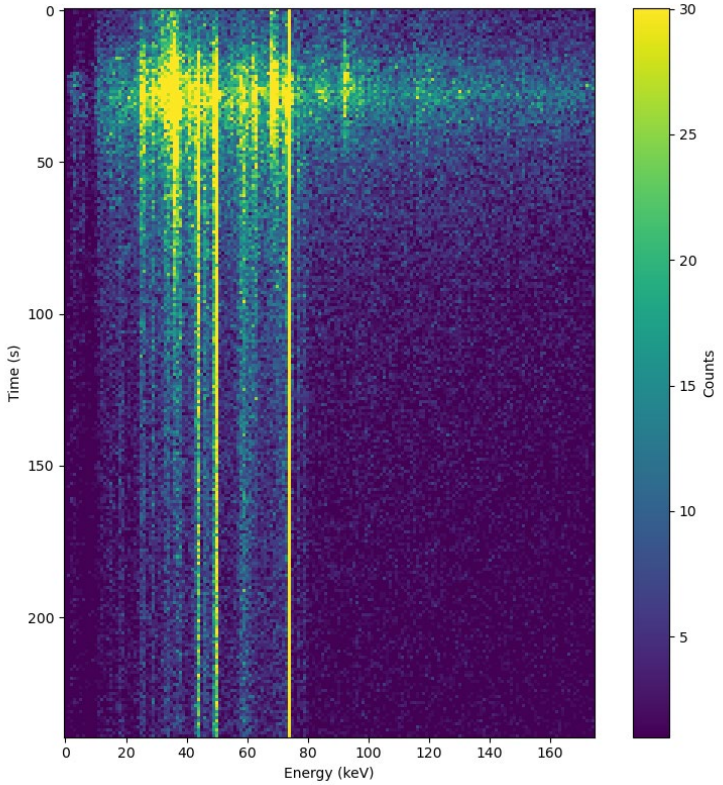

Figure 21. Chunked 2D spectrum of the irradiated natural uranium sample, measured by SOFIA. The color bar maximum was selected to highlight minor peaks; there are time and energy combinations that had more than 30 counts.

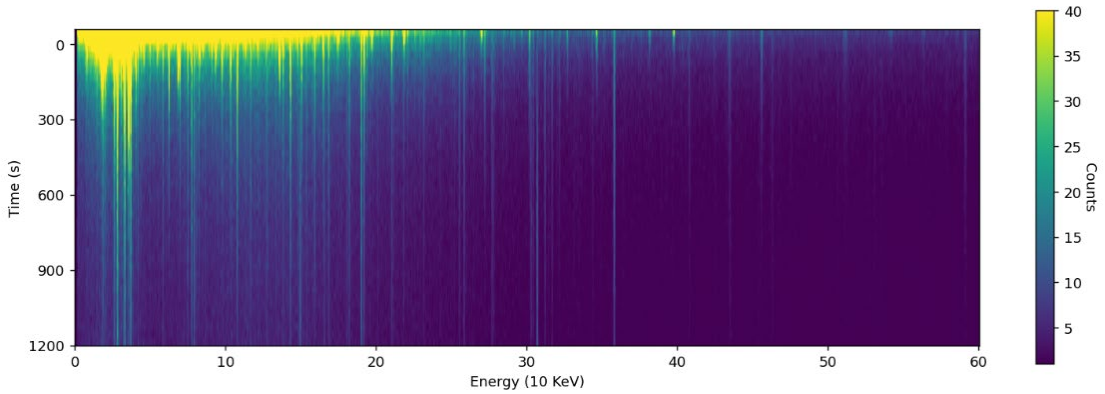

Figure 22. Chunked 2D spectrum of the irradiated 239Pu sample, measured by HPGe.

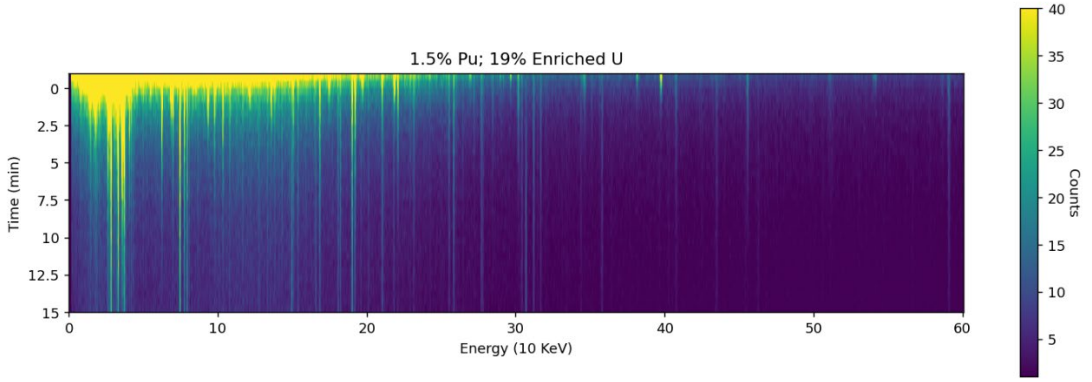

Figure 23. Irradiated mixture of 1.5% Pu by weight in 19% enriched uranium, measured by HPGe.

The samples that contain more ^{239}Pu and ^{235}U have more short-lived fission products in the 90-100 keV region, potentially obscuring the fluorescence x-rays. The resolution of the HPGe, especially in this region, led to inconclusive half-life estimates to calculate the required cooling time to see the x-rays, however that estimate will be more robust with the μCal data, once the processing and energy calibration improvements are implemented. While these fission products might obscure the x-ray signature, once they are well-understood and characterized, they could provide additional nondestructive gamma signatures of burnup and plutonium content.

5. NDA BURNUP QUANTIFICATION IN A PEBBLE BED REACTOR

A general strategy that balances time, cost, and nuclear safeguards is presented below. While the cesium ratio measured with HPGe did have approximately an order of magnitude higher error than μCal, HPGe measurements are faster and more routine than μCal. For that reason, pebbles discharged from the reactor could be effectively monitored using HPGe in the first few passes through the reactor if the purpose of measuring the pebbles is to ensure they are safe to recycle. Once the exit burnup of a

pebble starts to approach the average and/or maximum burnup, then the improved error of a μCal-based burnup quantification would result in additional fuel utilization.

μCal measurements are not complicated but do require longer measurement times compared to HPGe. In general, the HPGe cesium peaks are resolved well-enough after a measurement on the order of minutes, while μCal would require hours-long measurements. Estimated minimum measurement times will be estimated on re-irradiated TRISO particles in FY26.

5.1. Hypothetical Reactor Burnup Quantification System

Using previous work and similar operating HTGR reactors, an estimated burnup quantification and MC&A approach can be conceived using NDA signatures. The maximum burnup for TRISO fuel is generally quoted to be between 80 GWd/MTU and 100 GWd/MTU, accounting for fuel stability and proliferation considerations [8, 9, 10, 11]. This range translates to approximately 8.3 and 10.4 %FIMA. For this exercise, all burnups will be communicated as GWd/MTU.

If a maximum burnup is set to 100 GWd/MTU, from [8], and using previous simulation work to determine the average burnup per pass of a TRISO pebble [12] a burnup quantification system can be designed. Error estimates for HPGe and μCal can be estimated from Section 3.3, assuming a linear trend between error and burnup[1]. Using the average error and the true burnup, the expected burnup calculated from NDA signatures for each pass of a TRISO pebble are displayed in Figure 23. In this scheme, the HPGe burnup signature could be used for the first 4 passes of a pebble through the reactor. After the fifth pass, HPGe could still be used, however the error reported here would lead to a burnup estimate that is potentially higher than the maximum burnup allowed. If relying on HPGe only, this would result in confidently passing the TRISO pebbles through the reactor about four times for a final exit burnup of 67.81 GWd/MTU. If μCal signatures of burnup were available, samples of pebbles could be assayed for the longer measurement time. It is anticipated this would allow for three additional passes through the reactor, resulting in a final exit burnup of 95.6 GWd/MTU. Using μCal and HPGe together would create the most efficient NDA system and maximize the power generated from TRISO fuel.

In addition to using the μCal measurements for pebble recycling, it could also be considered for MC&A. The μCal signature is a direct observation of plutonium fluorescence x-rays and can therefore be used to verify the plutonium content of irradiated TRISO fuel. Further work is needed to create the calibration curve between the x-ray signature and plutonium content. Currently, this is limited by the knowledge of the plutonium in the irradiated TRISO fuel since previous characterization of the AGR particles was conducted on samples of the irradiated fuel. However, it is expected that individual particles will have more heterogeneity. To further prove out

[1] This is likely an over estimation due to the convex shape in Figure 14.

this concept, well-characterized standards will be measured to construct the calibration curve.

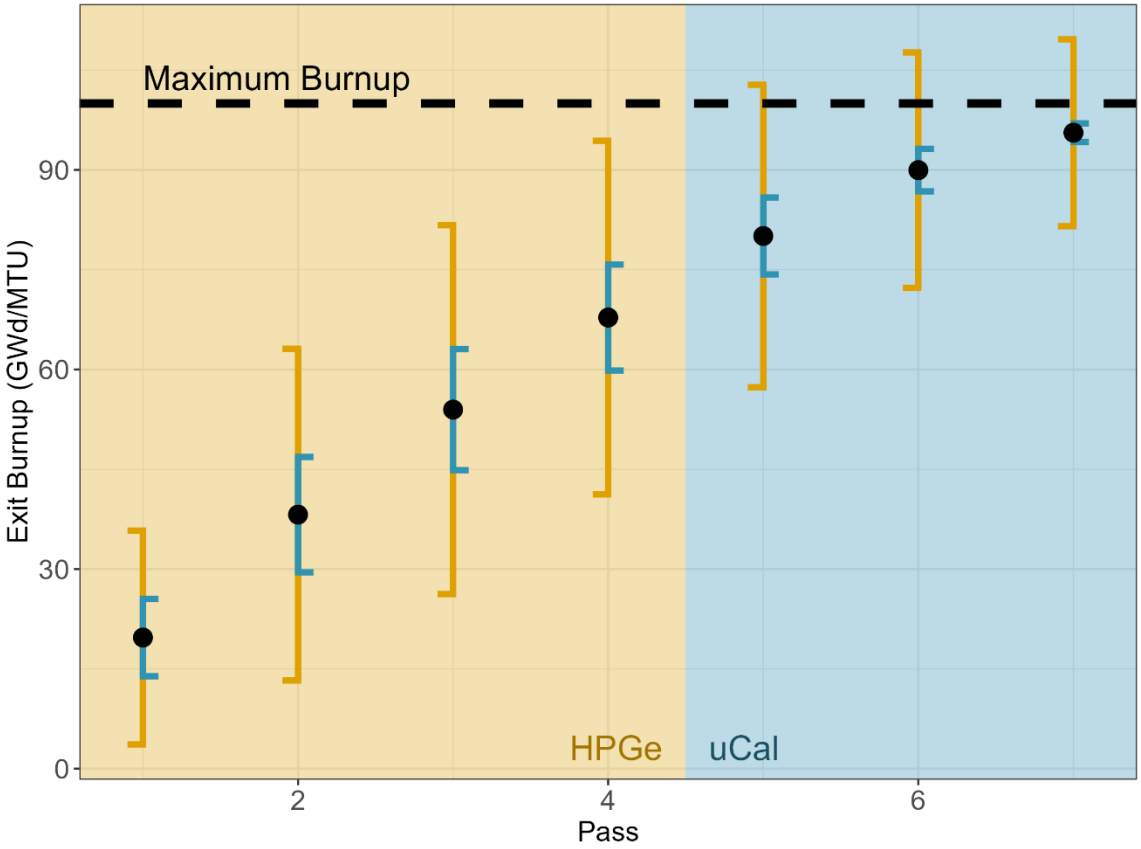

Figure 24. The expected true exit burnup (black marker) with the anticipated error in burnup calculation based on HPGe (orange error bars) and μCal (blue error bars). The maximum burnup of 100 GWd/MTU is denoted by a dashed line. To use fuel most efficiently, HPGe should be used after the first four passes (orange shaded region) and μCal should be used after the fifth pass (blue shaded region).

6. SUMMARY AND FUTURE WORK

We have investigated two nondestructive gamma techniques to measure burnup and plutonium content in irradiated TRISO fuel. HPGe is a proven and reliable technology that offers a signature less perturbed by the fuel form, while μCal measures the plutonium directly. The estimated error in burnup calculation from the cesium peaks measured by HPGe was higher than the error using the x-ray peaks measured by μCal. However, this top-down variance in error does not account for the uncertainty from counting statistics. A bottom-up error propagation was not carried out, as it would be largely driven by the number of calibration measurements and would therefore provide an overestimation of the method.

The fluorescence x-ray ratio measured by μCal may potentially be obscured by short-lived fission products in a short-cooled sample. To investigate this possibility,

measurements were taken of freshly irradiated actinide targets. Natural uranium, Pu-239, and mixtures of 15-19% enriched uranium with 0.5-2% plutonium were irradiated at HFIR. From preliminary analysis, there were more short-lived fission products in the Pu and mixed samples, however based on the count rate of the μCal detector after approximately an hour, it is unlikely that such interfering fission products would be present after approximately 100 hours of cooling time before a pebble is discharged from the reactor, which would be the earliest time feasible for an NDA measurement.

Using slightly inflated error estimates for HPGe and μCal, a burnup calculation scheme was devised for an illustrative PBR in Section 5.1. HPGe requires shorter measurements than μCal and is thus more efficient. This technology is suggested for earlier passes of pebbles to confirm the burnup adheres to an expected pattern. Once the exit burnup of a pebble starts to approach the maximum burnup allowed, the longer μCal measurements would provide greater accuracy and allow for pebbles to be recycled more times, increasing the efficiency of the TRISO fuel.

In addition to quantifying burnup, μCal could be used for MC&A by taking a direct measurement of the plutonium content in irradiated TRISO fuel. This work will be further investigated with well-characterized samples to construct a calibration curve for the fluorescence x-ray signatures. In addition to this measurement campaign, future measurement campaigns are planned to re-irradiate the TRISO particles reported here at HFIR. This will provide a measurement scheme more consistent with measuring TRISO fuel as it is discharged from the reactor core. The work will be carried out to create a comprehensive characterization of NDA signatures of burnup and plutonium content for PBRs.

7. ACKNOWLEDGEMENTS

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