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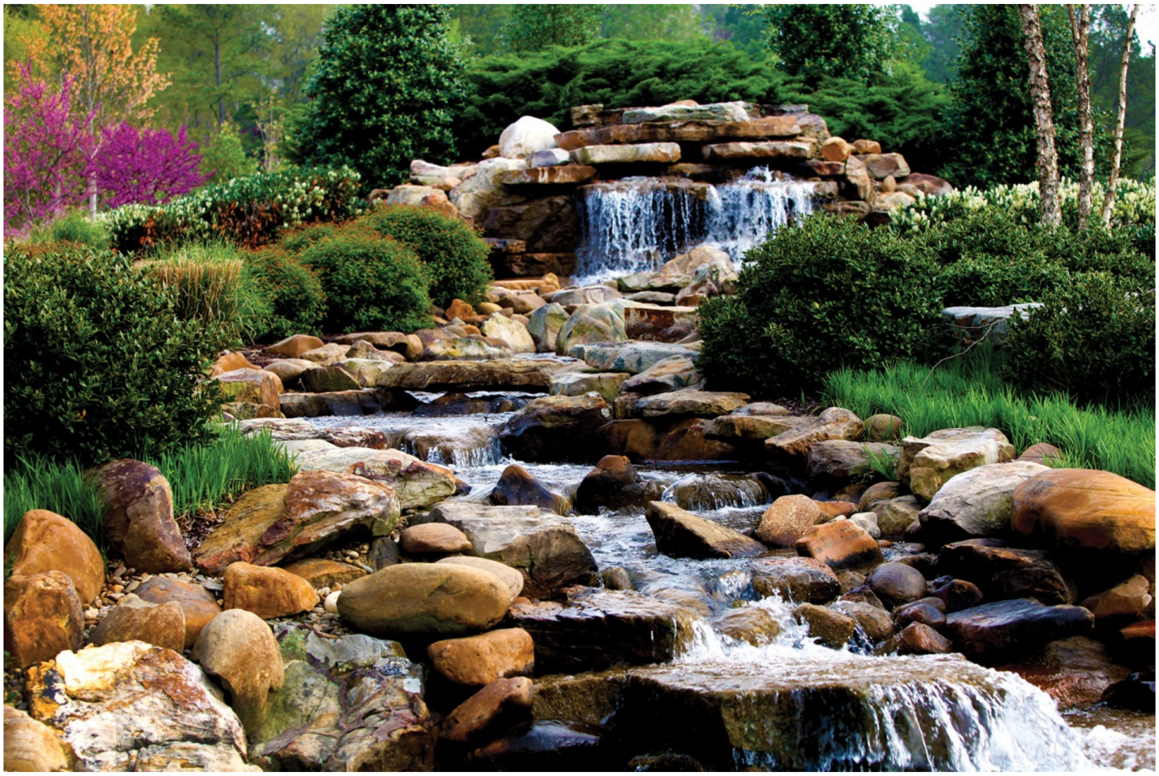
Gamma Spectrometry Code Rodeo for Uranium Enrichment—FY 2022 Report



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March 2023

OAK RIDGE National Laboratory

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Nuclear Nonproliferation Division

GAMMA SPECTROMETRY CODE RODEO FOR URANIUM ENRICHMENT— FY 2022 REPORT

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OAK RIDGE NATIONAL LABORATORY
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EXECUTIVE SUMMARY

In the first two quarters of FY22, data acquisition continued at ORNL and LLNL using uranium sources of known enrichments. This was an FY21 task which could not be completed in FY21 because of problems encountered with the ORNL M400 CZT in Q4 of FY21, and the subsequent repairs. The detector was received back from H3D in the first of September 2021 , and the measurements resumed . Measurements using the repaired detector were completed in Q1 of FY22. The spectra were distributed by ORNL to the analyzing labs. Analysis results were received in Q2 of FY2022. The results from the various codes were intercompared and an ANOVA analysis was performed. Random and systematic uncertainties were established for each code. The ANOVA results and discussions were included in a revised version of FY21Annual Report issued in March 2022. A paper was presented at the INMM 2022Annual Conference, with the analysis results from the various isotopic codes, and the ANOVA table with random and systematic uncertainties. The Project Work Plan (PWP) for FY22 included a task to perform field testing of the M400 CZT and the analysis codes using UF_6 cylinder measurements at the Framatome Fuel Fabrication Facility in Richland, WA. PNNL was the lead for the field testing task. PNNL drafted a Field Test Plan, and refined it based on comments received from the team. PNNL coordinated with Framatome facility, the logistics of carrying out the field testing . A collimator and shield made out of TFlex (tungsten impregnated polymer) was designed and professionally manufactured. The collimators were used in the field test measurements. The measurements at Framatome were completed on April 21, 2022. A total of 34 Type 30B cylinders were measured using three M400 detectors (PNNL, LLNL, and ORNL detectors). Measurements using M400 were taken at two locations on the side of each cylinder and from the end-on bottom location. Additionally, HPGe measurements were taken at the end-on location to establish ground truth. Cylinder wall thickness measurements were also made at all three locations. To gain a better understanding of the effect of background from surrounding cylinders, the same five cylinders measured individually in low background locations were measured again in the cylinder storage yards. Due to inclement weather, manufacturer delays, shipping delays, and equipment failure, the measurement campaign spanned twice as long compared to the original timeline. Gamma-ray spectra from M400 and HPGe detectors, along with the cylinder data and photographs were organized and shared with the collaborators for further analysis. Spectra were analyzed by the participating laboratories. FY22 PWP also consists of tasks related to plutonium source measurements, adapting the codes to analyze plutonium spectra, and inter-comparison of the results from various codes. Plutonium spectra are being acquired at LANL, ORNL, and LLNL. LANL, SNLand LLNL are in the process of modifying FRAM, GADRAS, and CZTU, respectively. The plutonium related tasks will be completed in Q2 of FY23.

ABSTRACT

A new generation of cadmium zinc telluride (CZT) detectors with a higher energy resolution has become available and is being evaluated by the International Atomic Energy Agency (IAEA) for safeguards verifications in the field. Under the "Gamma Rodeo" project, the isotopic codes are being modified and their performance was benchmarked using uranium spectra acquired using the higher resolution CZT model M400. In Q1 of FY22, data acquisition using uranium sources of known enrichments was continued. This task was part of FY21 PWP, but because of detector problems and repairs, the task spilled over into FY22. The measurements were completed, and the analysis results were received in Q2 of FY22. The results from the various codes were intercompared and an ANOVA analysis was performed. Random and systematic uncertainties were established for each code. The ANOVA results and discussions were included in a revised version of FY21Annual Report issued in March 2022. A paper was presented at the INMM 2022Annual Conference, with the analysis results from the various isotopic codes PNNL led the efforts to conduct the field testing at the Framatome Fuel Fabrication Facility in Richland, WA. PNNL drafted a field test plan, negotiated the contract with Framatome, and executed the logistics of the field testing task. Field testing was conducted at the Framatome facility for two weeks in April 2022 using thirty-four 30B UF_6 cylinders. Three M400 CZT and an electrically cooled High Purity Germanium (HPGe) detector were used in the measurements. Spectra were distributed to the participating laboratories. Analysis results from field test spectra are available from GADRAS and FRAM codes. Measurements are in progress at ORNL, LANL, and LLNL using plutonium sources. Analysis capability of FRAM, GADRAS, and the LLNL code are being extended to be able to analyze M400 plutonium spectra. Problems with the ORNL M400 CZT caused delays and impacted the progress of FY22 tasks.

1. INTRODUCTION

The gamma rodeo project takes advantage of the new developments in the CZT detector technology and aims to adapt software packages used by the IAEA for verification of ^{235}U enrichment declared by facilities. The rodeo exercise will focus on analyses using the following codes: (i) GEM[1], [2], (ii) FRAM[3], (iii) CZTU[4], (iv) GADRAS[5]. The GEM code uses the infinite thickness approach to determine uranium enrichment. FRAM and CZTU use a method of peak ratios, while GADRAS models the whole spectrum based on response calibration functions.

The M400 is a CZT-based room temperature spectrometer produced by H3D, Inc. The M400 demonstrates superior energy resolution, effective isotope identification capabilities, and convenient usability features when tested in a controlled laboratory environment. These characteristics define the M400 as a potential platform for IAEA field detection applications, that could become suitable for nuclear material characterization (e.g., enrichment verification) and nuclear safeguards missions.

In the first two quarters of FY22, data acquisition continued at ORNL and LLNL using uranium sources of known enrichments. This was an FY21 task which could not be completed in FY21 because of problems encountered with the ORNL M400 CZT in Q4 of FY21, and the subsequent. The detector was received back from H3D in the first of September 2021, and the measurements resumed. Measurements using the repaired detector were completed in Q1 of FY22. The spectra were distributed by ORNL to the analyzing labs. Analysis results were received in Q2 of FY2022. The results from the various codes were intercompared and an ANOVA analysis was performed. Random and systematic uncertainties were established for each code. The ANOVA results and discussions were included in a revised version of FY21Annual Report issued in March 2022. A paper was presented at the INMM 2022Annual Conference, with the analysis results from the various isotopic codes, and the ANOVA table with random and systematic uncertainties.

The FY22 PWP consists of the following tasks.

Task 1: Custom shield and collimator for M400 CZT

Task 2: Field test of M400 CZT for uranium analysis

Task 3: Synthetic Spectra Generation (ON HOLD)

Task 4: Plutonium source measurements using M400 CZT

Task 5: Code development and analysis of M400 plutonium source spectra

The following sections elaborate on the work that was carried out under each of the above tasks.

2. TASK 1: Custom shield and collimator for M400 CZT

A custom rectangular collimator and shielding was designed at ORNL and professionally manufactured by Eichrom Technologies, Inc. The collimator was made out of T-Flex®(tungsten impregnated polymer) a non-toxic alternative to lead. The density of the TFlex material is 6.93 $g.cm^{-3}$. The thickness of the TFlex layer in the shielding around the detector, as well as the TFlex layer that forms the collimator aperture is 1.27 cm (0.5 inches). An inner lining of copper of thickness 0.3175 cm (1/8 inches) was installed to attenuate the tungsten X-rays from TFlex. The shield consists of 1.27 cm thick TFlex back shield. The shield/collimator unit can be mounted on a tripod for field use. The TFlex shielding and collimator assembly has been procured by each of the laboratories participating in the "Gamma Rodeo" project. . Figures 1 and 2 show the shield and collimator for the M400 CZT.

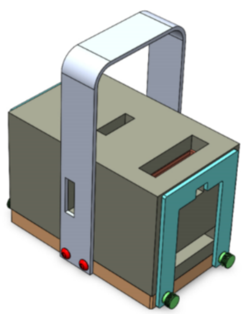

Figure 1. Custom shield/collimator for M400 CZT

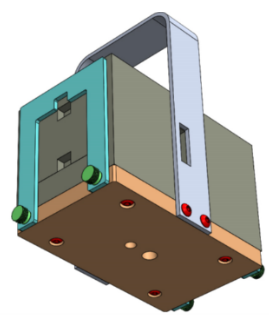

Figure2: Bottom view showing the tripod mount

3. TASK 2: FIELD TEST OF M400 CZT FOR URANIUM ANALYSIS

To further validate the performance of the high energy resolution CZT detector and the isotopic analysis codes, a field measurement campaign consisting of UF_6 cylinder measurements at a fuel fabrication facility (Framatome, Richland WA) was carried out. Type 30B cylinders containing UF_6 were measured by personnel from PNNL, ORNL, and LLNL, using their respective collimated M400 detectors. The spectra were provided to participating labs (PNNL, LANL, LLNL, SNL, and ORNL). For the field measurements of UF_6 cylinders, the analyzing labs will provide the isotopic analysis results to ORNL, which is the control lab. ORNL will inter-compare results from various codes and perform a statistical analysis to establish systematic and random uncertainties. Analysis results from the field test spectra are available from FRAM (LANL) and GADRAS (SNL) codes. Results from GEM (PNNL) and CZTU (LLNL)analyses are expected in Q1 of FY23. FY22 tasks, once completed, will provide an evaluation of the various analysis codes with possible recommendations pertaining to safeguards verification measurements and analysis.

The Gamma Rodeo team spent the early part of FY22 Q2 in preparing for the field campaign at Framatome Richland, WA. The preparations included completion and approval of the field test plan, contracting with Framatome, reviewing cylinder inventory, training, and field logistics planning. The measurements at Framatome were completed on April 21, 2022. A total of 34 Type 30B cylinders were measured using three M400 detectors. Table A.1in Appendix A gives the specifications of the 34 cylinders used in the field test. The M400 CZT detectors were mounted on tripods and positioned at the midplane of the UF_6 cylinders. The distance from the outer wall of the UF_6 cylinder to the front face of the collimator was 3.81 cm (or 1.5 inch). For measurements using LLNL detector, a copper filter of thickness 0.3175 cm (1/8th of an inch) was installed. The fitting function used by the CZTU code is sensitive to the tungsten X-rays at 59 keV and 68 keV, and therefore it was necessary to practically eliminate the tungsten X-rays using copper filter. Figure 3 below shows the measurements locations along the cylinder. Measurements using M400 were taken at locations L1, L2, and L3. Additionally, HPGe measurements were taken at location L3 to establish ground truth. Repeat measurements of 15 minute counts (real time) were performed. Background measurements were performed using an empty 30B cylinder, and also without a cylinder. Figures 4,5, and 6 are example photographs from the field test measurement campaign.

Cylinder wall thickness measurements were also made at all three locations. To gain a better understanding of the effect of background from surrounding cylinders, the same five cylinders measured individually in low background locations were measured again in the cylinder storage yards (see Figure 7). Due to inclement weather, manufacturer delays, shipping delays, and equipment failure, the measurement campaign spanned twice as long compared to the original timeline. Gamma-ray spectra from M400 and HPGe detectors, along with the cylinder data and photographs were organized and shared with the collaborators for further analysis.

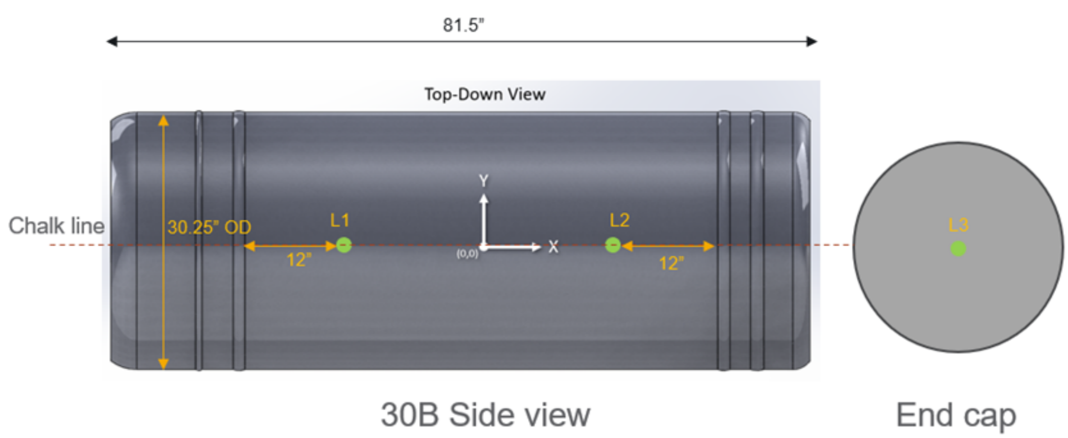

Figure 3. Graphic depicting measurement locations long the 30B UF_6 cylinder

Figure 4. A 30B UF_6 cylinder located in place in front of the measurement trailer.

Figure 5. An unusual mid-April snowstorm in the Pacific Northwest impacted the field tests.

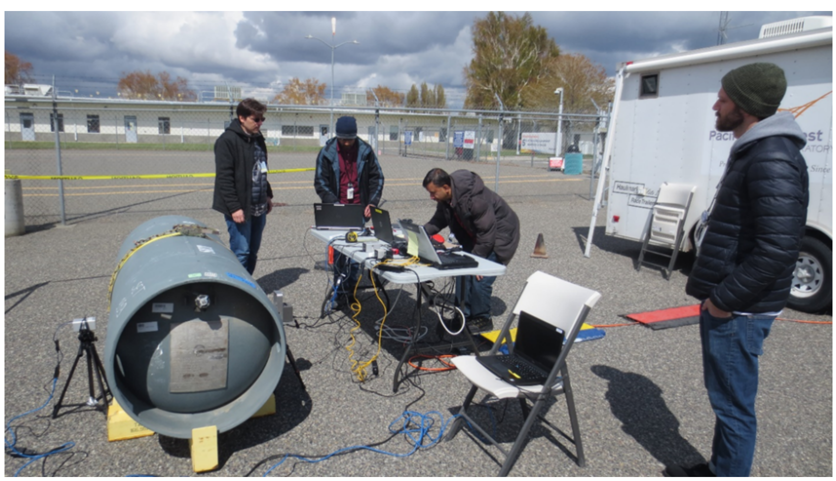

Figure 6. Field test measurements in progress.

Figure 6. Picture shows the electrically cooled HPGe set up in location L3

Figure 7. M400 CZT measurements in the Framatome cylinder yard

Additional photographs can be found by accessing the link below for the drop box folder.

https://www.dropbox.com/sh/5ecyacoo85dksva/AABRFzjpnVJo-dZGoaD_M_hda?dl=0

3.1 ANALYSIS RESULTS OF FIELD TEST SPECTRA USING THE GADRAS CODE

Full Spectra Analysis (FSA) and Differential Attenuation Analysis methods and their combination were used to fit the UF_6 spectra acquired using the three M400 CZT detectors, as well as the electrically cooled HPGe detector. Example screenshots of GADRAS analysis are shown in figures 8, 9, and 10.

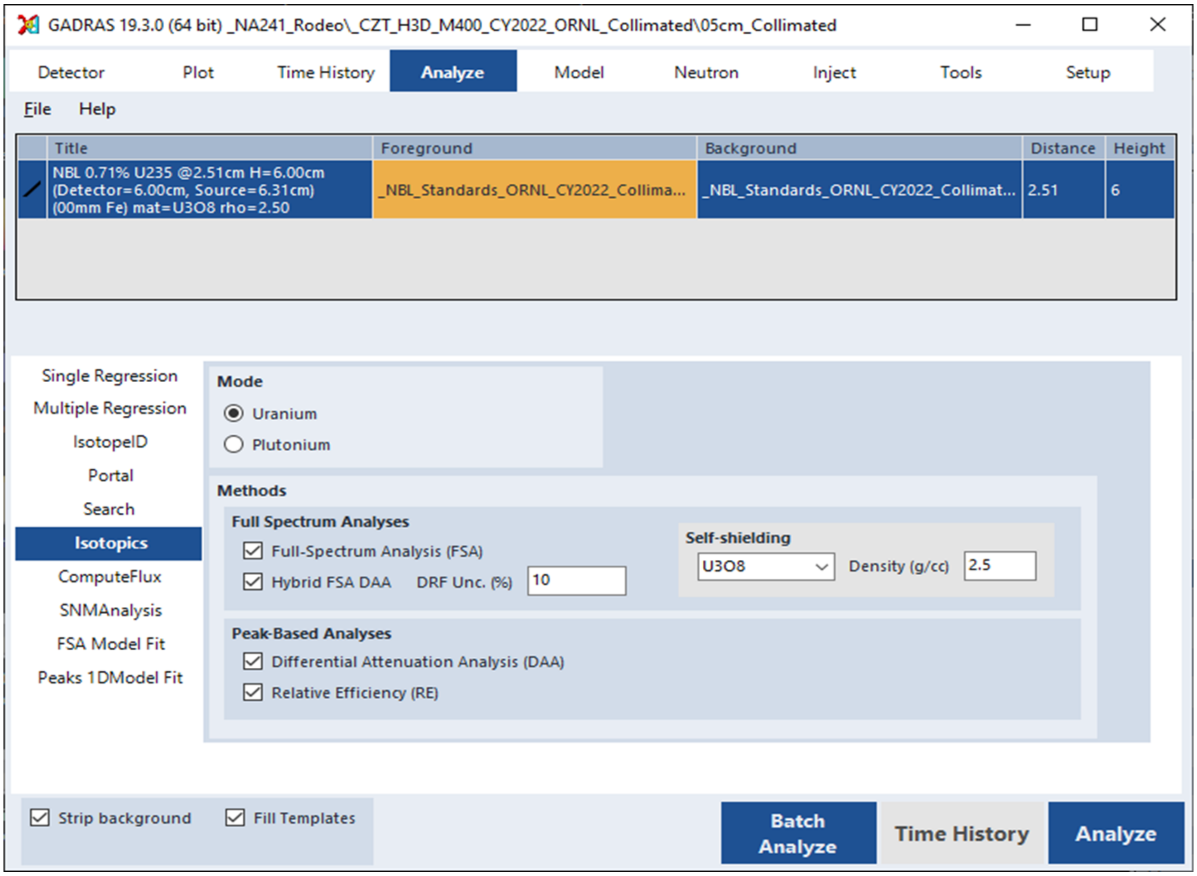

Figure 8. Example analysis screenshot from beta version of GARDAS

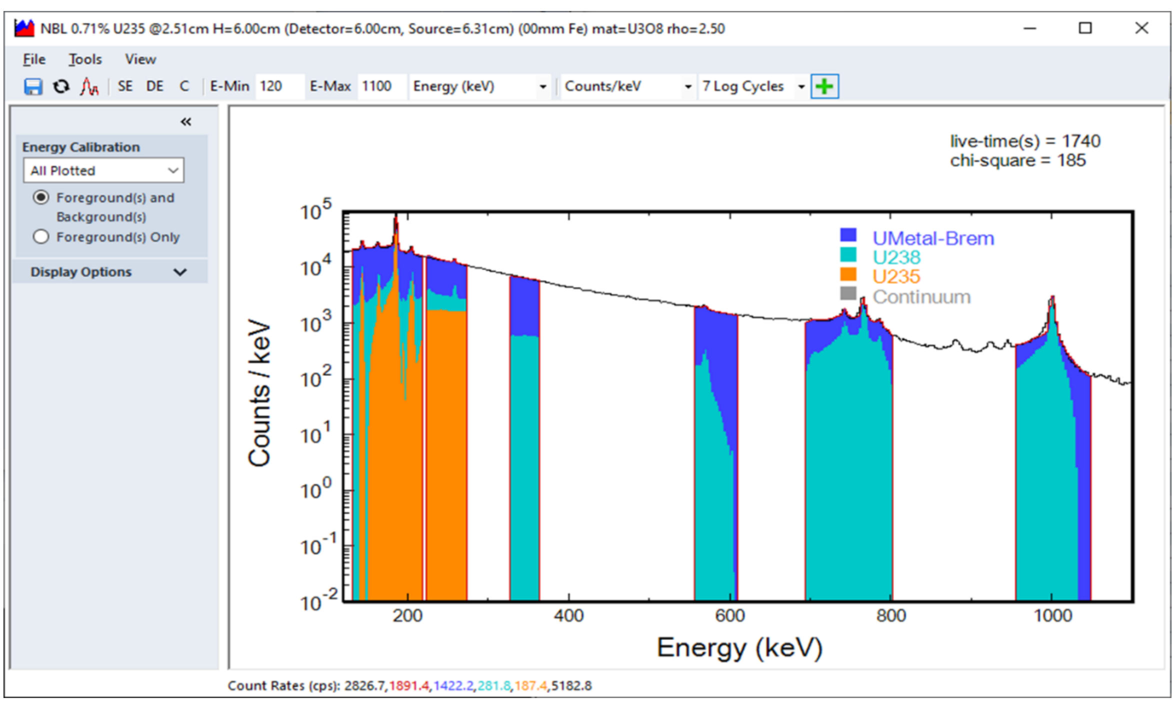

Figure 9. GADRAS fit for a M400 CZT gamma spectrum acquired using a natural uranium source

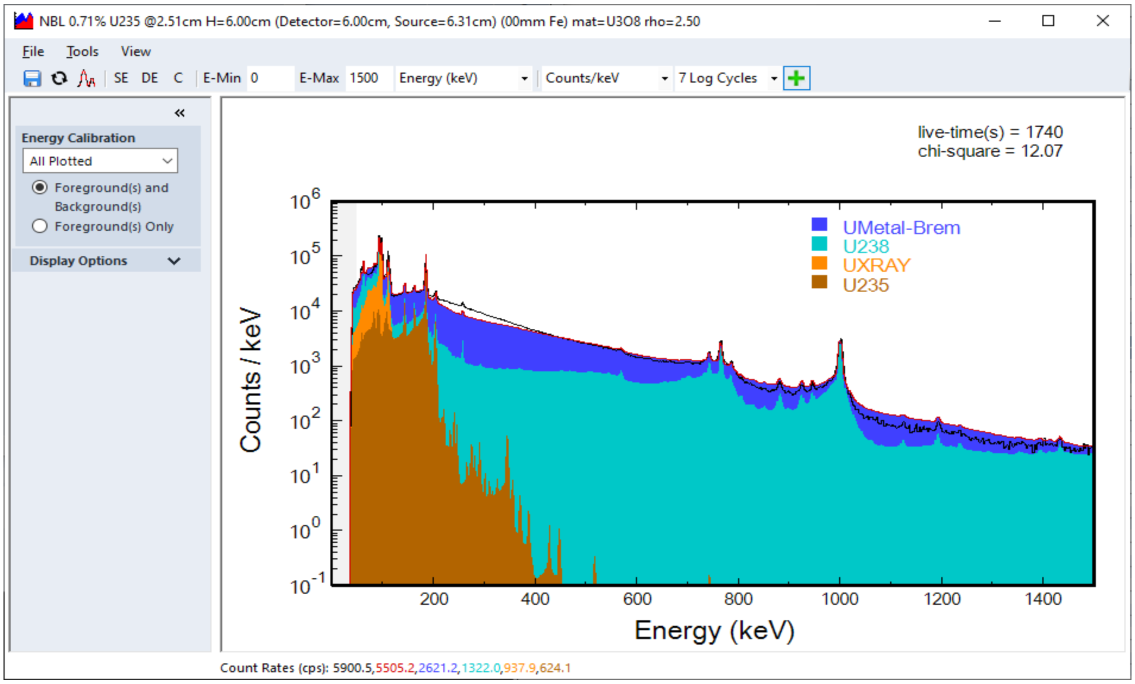

Figure 10. Contributions to spectrum as modeled by GADRAS

Figures 11,12, and 13 give the GARDAS results for spectra acquired using the LLNL, ORNL, PNNL M400 CZT detectors respectively. Results are given for spectra acquired with each detector at UF_6 cylinder locations L1, L2, and L3. Figure 14 gives the GADRAS results for spectra acquired using the electrically cooled HPGe detector at location L3.

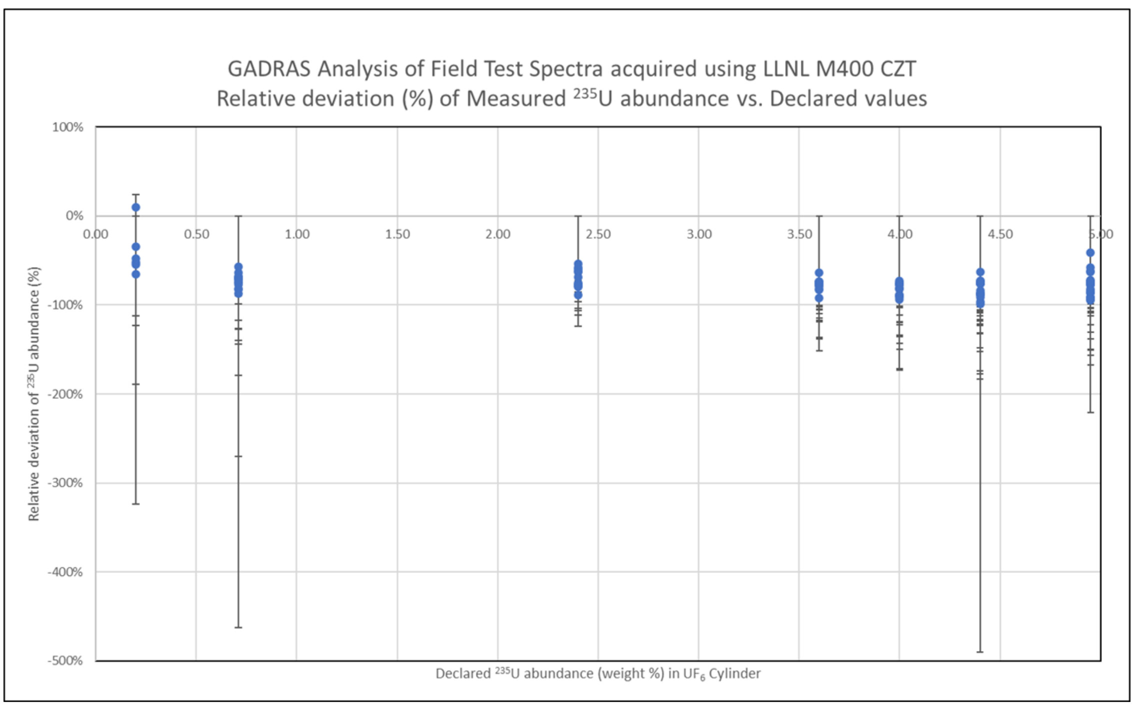

Figure 11. GADRAS results for LLNL M400 CZT UF_6 spectra from field testing

For the measurements performed using the LLNL CZT detector, the average deviation of the measured ^{235}U abundances with respect to the declared value is -75% ± 14%. GADRAS solves for the isotopics by fitting the whole spectrum, including contributions from ^{235}U and ^{238}U daughters ^{234}Th and ^{234m}Pa. In the UF_6 cylinders, the thorium and protactinium daughters of ^{238}U are in-homogeneously mixed in with ^{235}U. Therefore, gamma spectrometry codes that fit the whole spectra or use the "peak ratio" approach to determine ^{235}U abundance, will potentially yield large deviations from the true value as is the case with the results from the Framatome field test spectra. The measured abundances are mostly underestimated compared to the declared values.

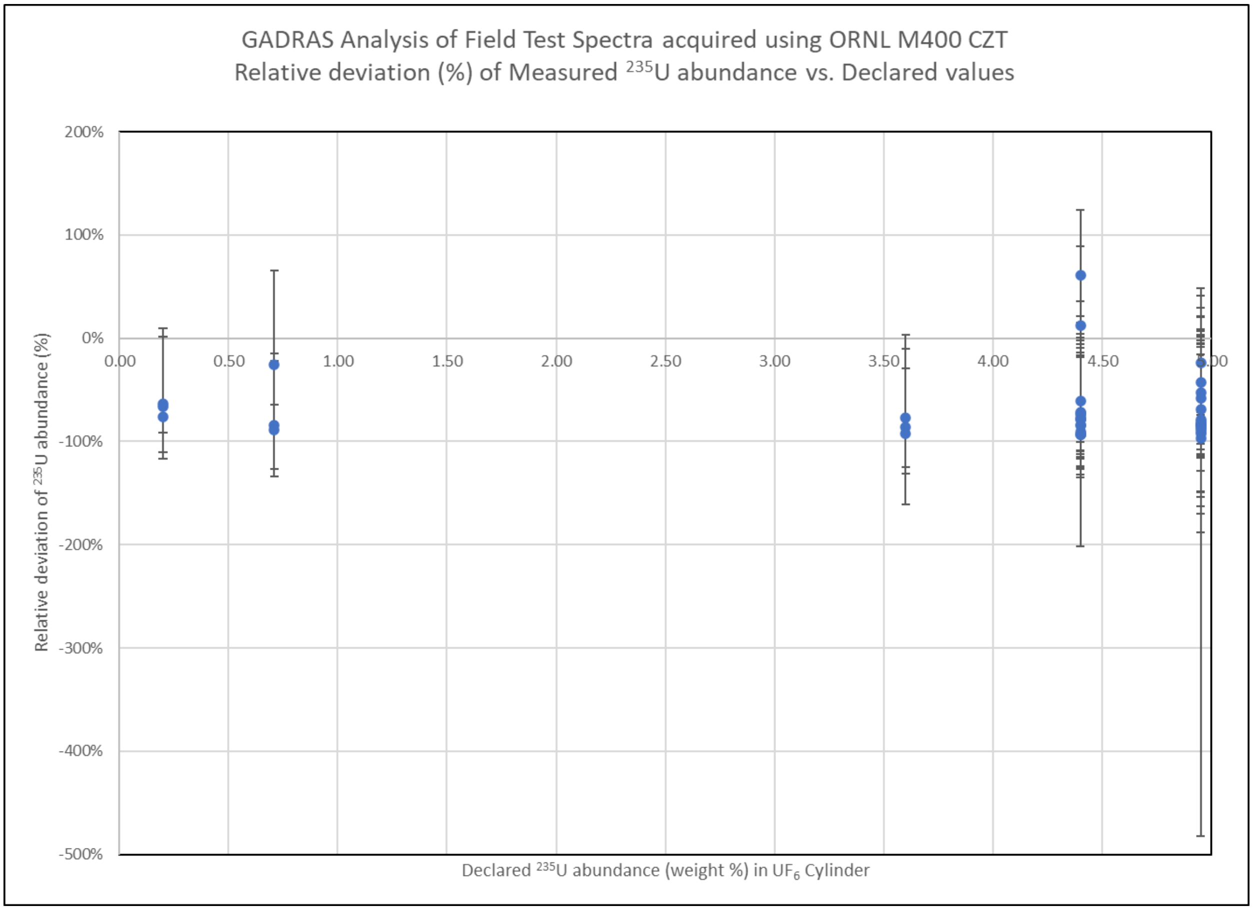

Figure 12. GADRAS results for ORNL M400 CZT UF_6 spectra from field testing

The measurement data available from the ORNL CZT detector is about half the number of data points when compared to LLNL and PNNL detectors. The reason is, approximately halfway through the field testing, ORNL detector suffered a water intrusion at the USB port for power (because of inadvertent exposure to rainwater) and suffered a catastrophic failure. Based on the available data, the average deviation of the measured ^{235}U abundances with respect to the declared value is -71% ± 30%.

The results from the PNNL CZT measurements are given in Figure 13. The average deviation of the measured ^{235}U abundances with respect to the declared value is -68% ± 23%. Thus, on average, the GADRAS analysis results from LLNL, ORNL, and PNNL CZT detectors all underestimate the ^{235}U abundances in the Framatome UF_6 cylinders by -75%, -71%, and -68%, respectively.

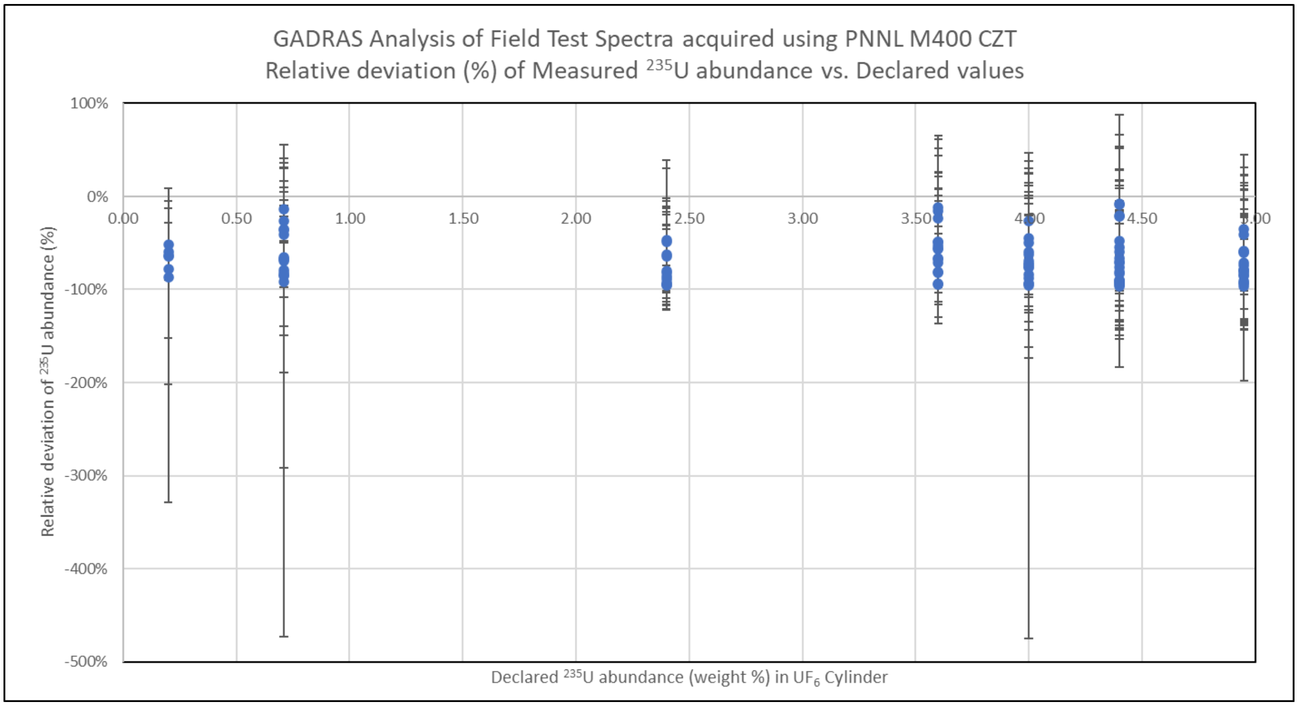

Figure 13. GADRAS results for PNNL M400 CZT UF_6 spectra from field testing

The results from the GADRAS analysis of electrically cooled HPGe spectra are given in Figure 14.

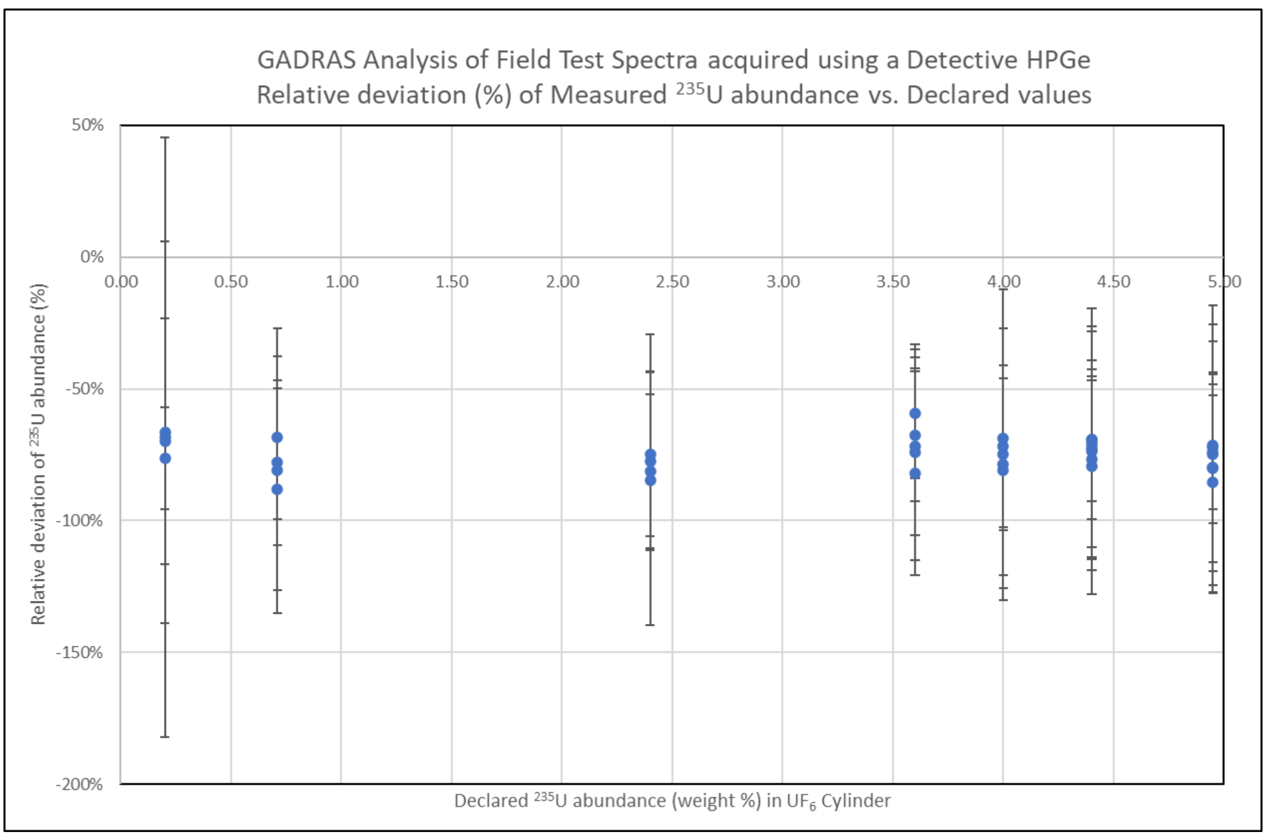

Figure 14. GADRAS results for UF_6 spectra acquired using electrically cooled HPGe

The average deviation of the measured ^{235}U abundances with respect to the declared value is -75% ± 6% for the electrically cooled HPGe detector measurements. The average deviation of the GADRAS results for the HPGe isimilar to the performance of the M400 CZT detectors. This proves that the inaccuracy of the results is due to the inhomogeneous mixture of Th and Pa daughters of ^{238}U with ^{235}U, and not because of the poorer resolution or the non-gaussian peak shapes of the CZT detectors. The standard deviation of the HPGe results is smaller than those of the CZT results probably since the HPGe measurements only consist of a single measurement location, namely L3, whereas the CZT average consists of measurement data taken at L1, L2, and L3.

3.2 ANALYSIS OF FIELD TEST SPECTRA USING THE FRAM CODE

The beta version FRAM 7.0 was used to analyze the CZT and HPGe spectra acquired during the field testing at Framatome Fuel Fabrication Facility. FRAM analysis is based on the peak ratio method. FRAM uses the gamma ray emissions ^{235}U and ^{238}U and their daughter nuclides. Any inhomogeneities in the distribution of ^{238}U daughters at a measurement location inside the cylinder will impact the accuracy of the determination of ^{235}U abundance. Such inhomogeneities are characteristic of UF_6 distributed inside the cylinders. Their impact on FRAM results is evident. A number of measurements had been performed using the ORNL M400 without any collimator just as an interesting aside. The ORNL collimator/shield unit did not arrive from the vendor until the Friday of Week 1 of measurements. Rather than let the detector idle, the measurement team decided to collect uncollimated data using the ORNL M400 CZT. FRAM 7.0 analysis results are presented in figures 15a, 15b, 16, 17, and 18 for collimated ORNL M400, uncollimated ORNL M400, collimated LLNL M400 and collimated PNNL M400 CZT detectors.

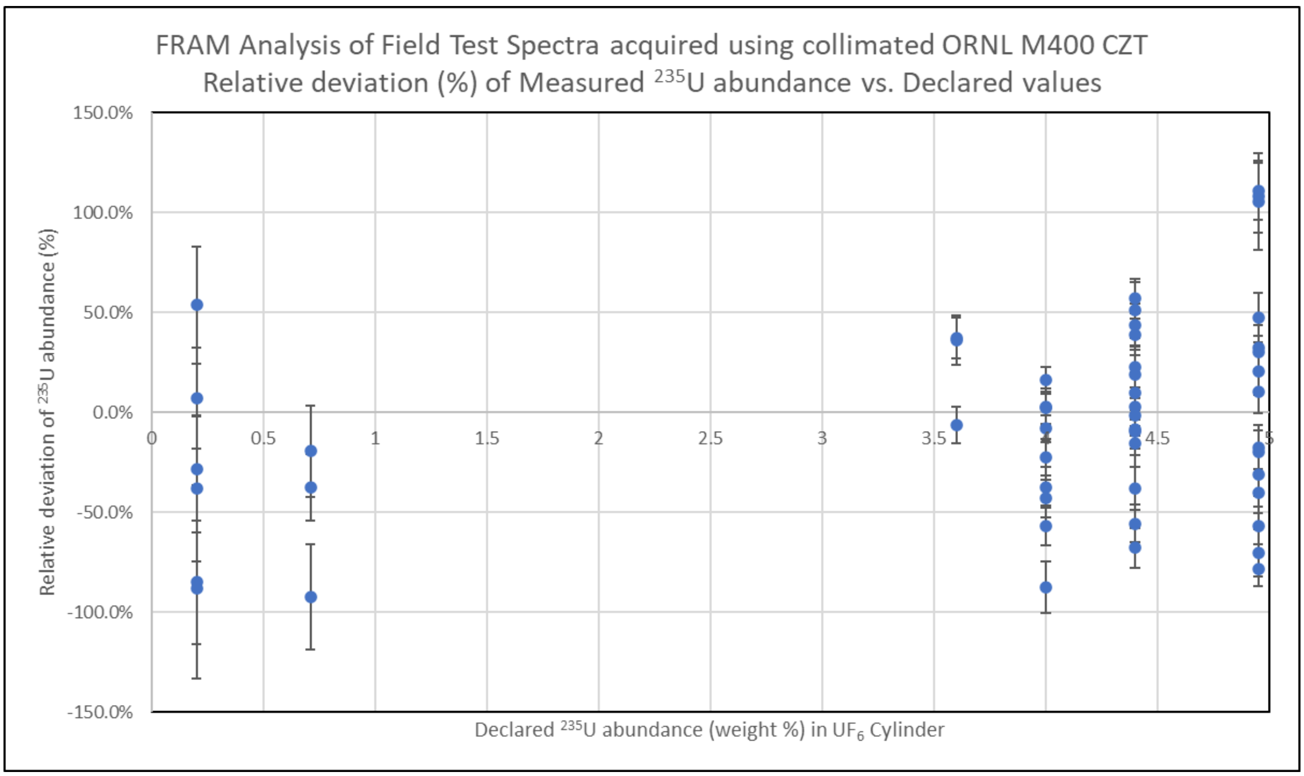

Figure 15a. FRAM 7.0 analysis results for UF_6 spectra acquired using collimated ORNL M400 CZT

For measurements performed using the collimated ORNL M400 CZT, the average relative deviation of ^{235}U abundance estimated from FRAM 7.0 analysis with respect to declared values is -5.8% ± 49.8%. In other words, the positive and negative values of relative deviations are almost equally distributed, and with an overall standard deviation of nearly 50%!

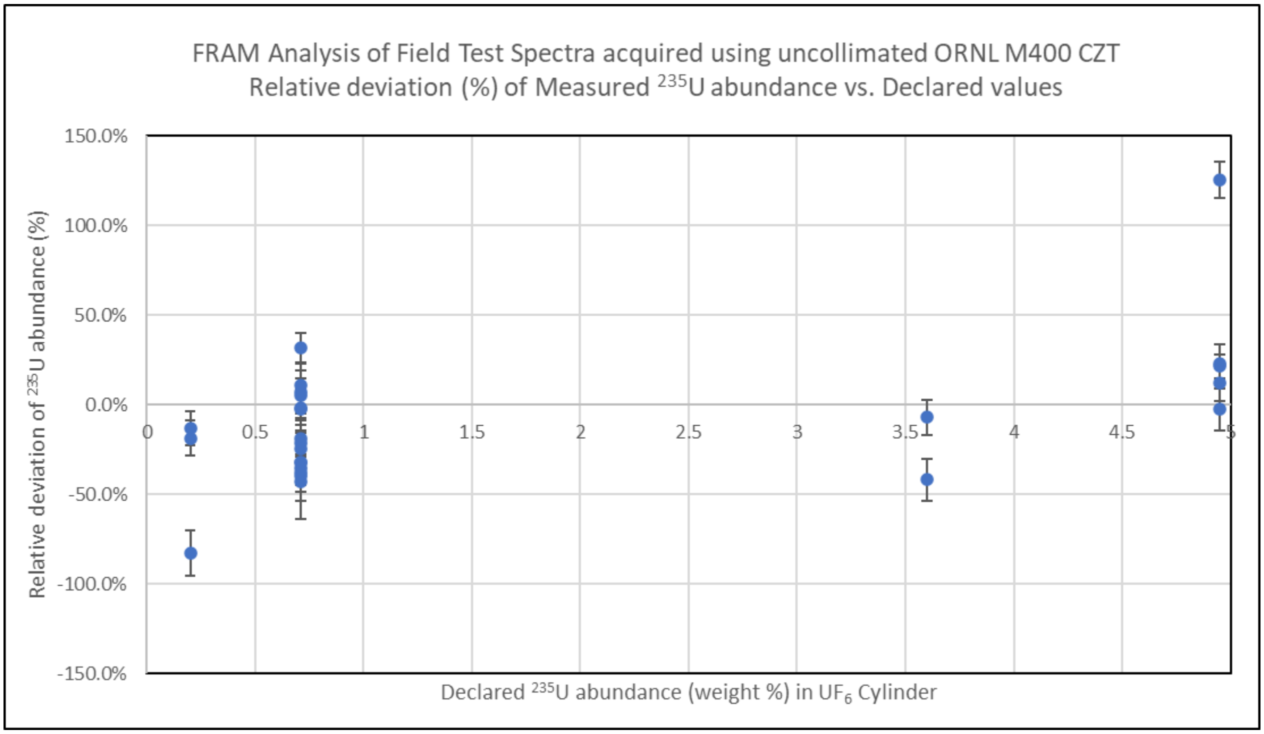

Figure 15b. FRAM 7.0 analysis results for UF_6 spectra acquired using uncollimated ORNL M400 CZT

For measurements performed using uncollimated ORNL M400 CZT, the deviation of ^{235}U abundance estimated from FRAM 7.0 analysis with respect to declared values is -8.6% ± 38.2%. The accuracy of FRAM version 7.0 results for spectra collected using an uncollimated M400 detector is similar to the results for collimated spectra. Both sets of results show a large standard deviation. The presence or the absence of the collimator did not have a significant impact. The large deviations of FRAM results with respect to known ^{235}U abundances is due to the inhomogeneous mixing of Th/Pa and U in the cylinders.

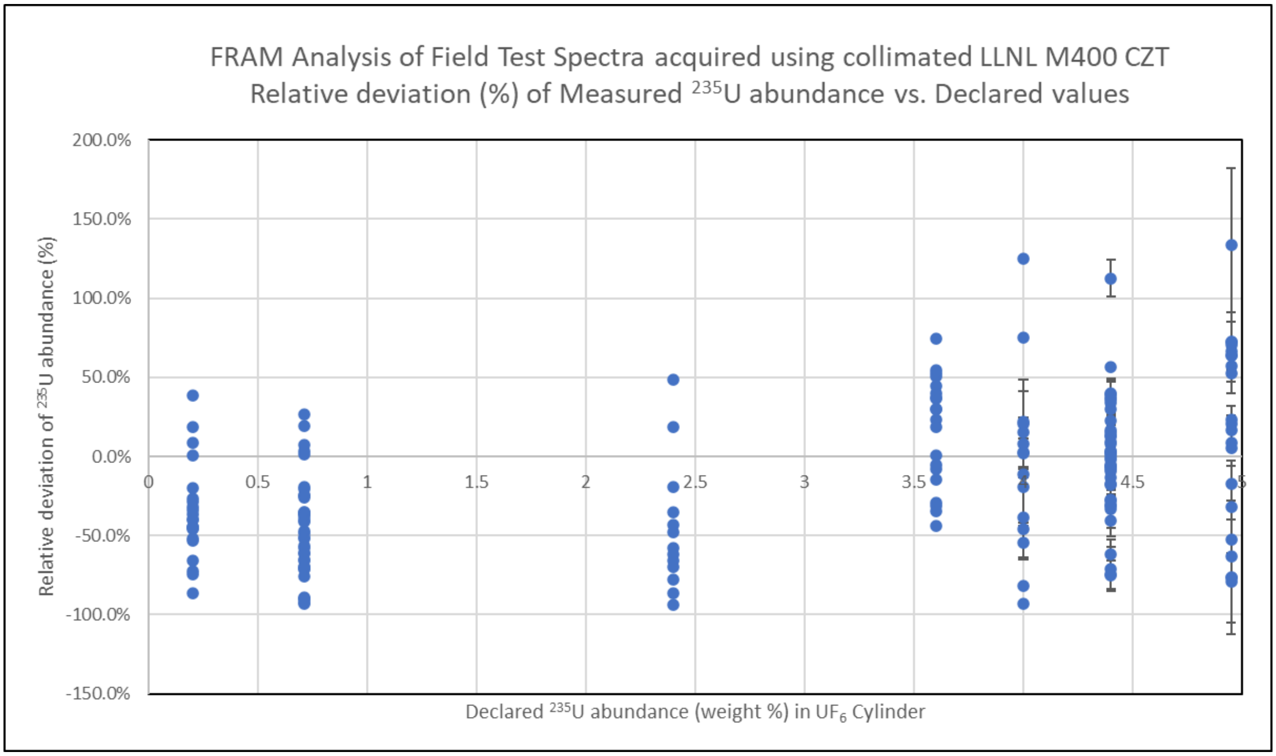

Figure 16. FRAM 7.0 analysis results for UF_6 spectra acquired using collimated LLNL M400 CZT

For measurements performed using uncollimated LLNL M400 CZT, the deviation of ^{235}U abundance estimated from FRAM 7.0 analysis with respect to declared values is -15.4% ± 47.3%.

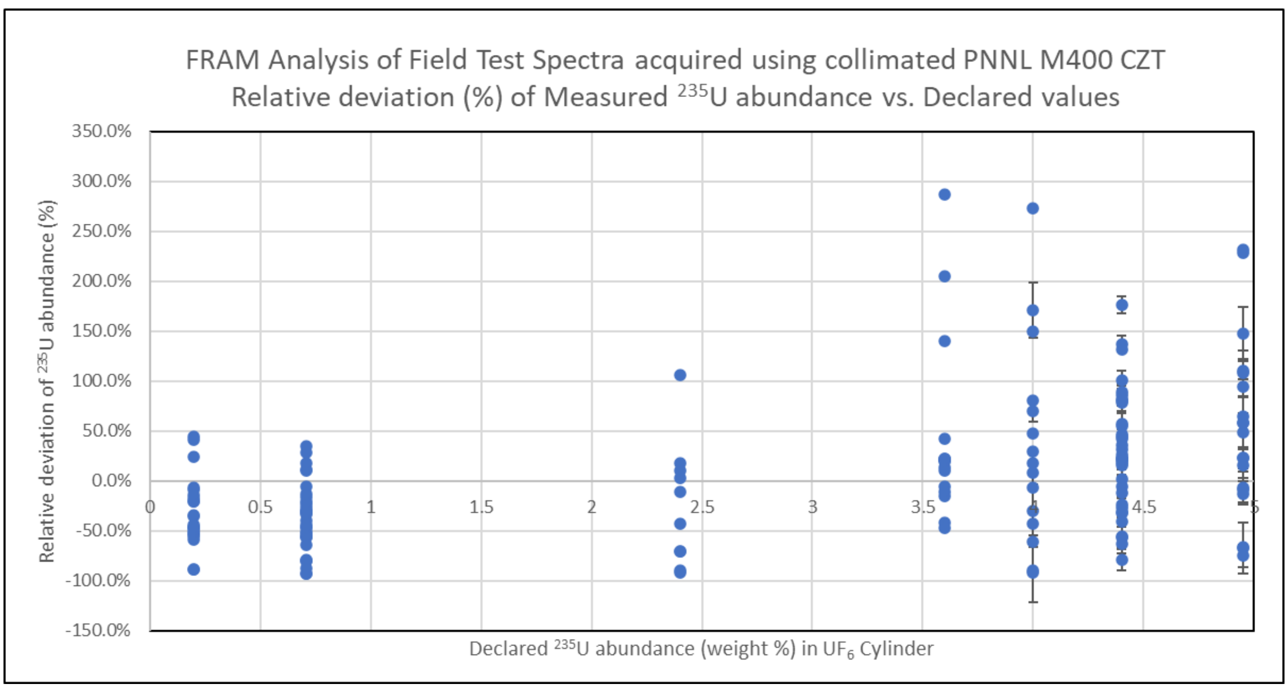

Figure 17. FRAM 7.0 analysis results for UF_6 spectra acquired using collimated PNNL M400 CZT

For measurements performed using uncollimated PNNL M400 CZT, the deviation of ^{235}U abundance estimated from FRAM 7.0 analysis with respect to declared values is +7.4% ± 74.3%.

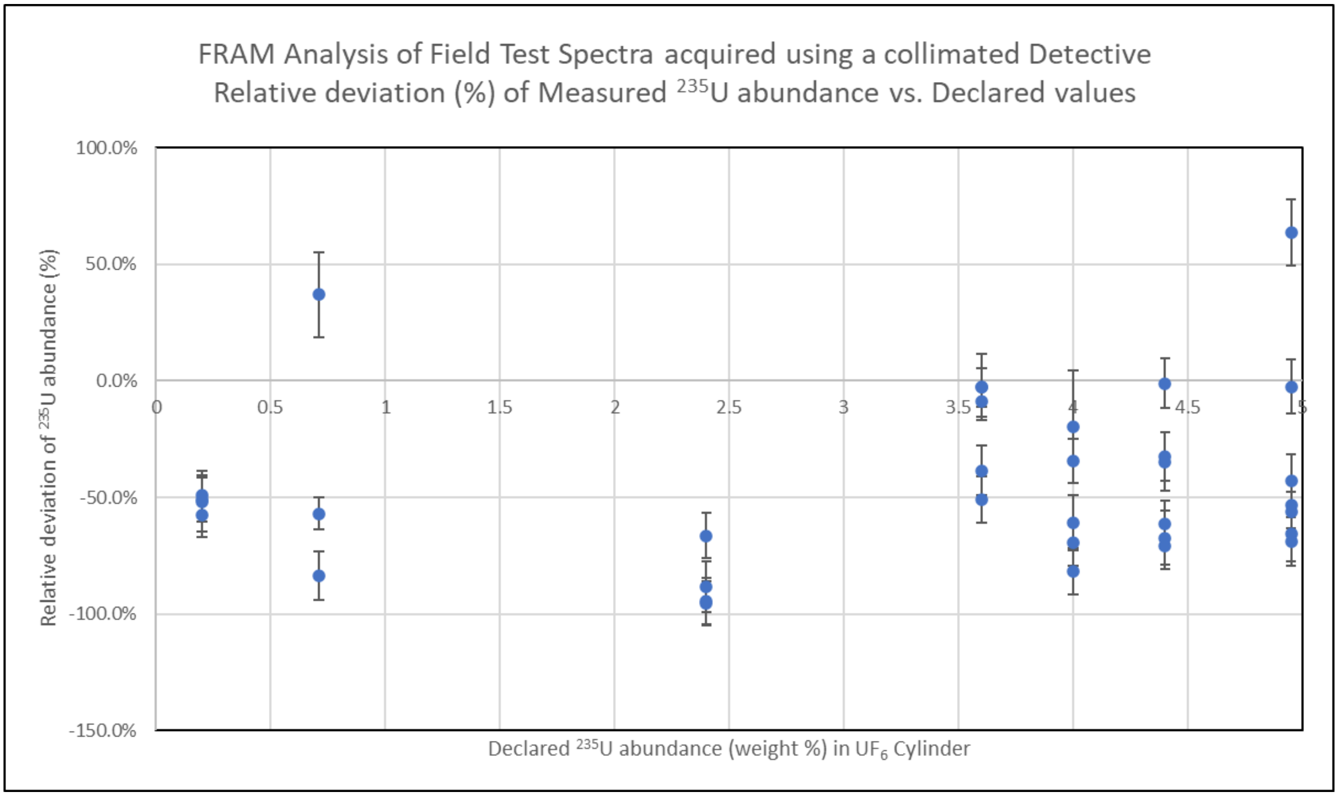

Figure 18. FRAM 7.0 analysis results for UF_6 spectra acquired using collimated electrically cooled HPGe

For measurements performed using a collimated electrically cooled HPGe, the deviation of ^{235}U abundance estimated from FRAM 7.0 analysis with respect to declared values is -44.7% ± 35.8%. The HPGe results from FRAM show the highest average deviation. The FRAM estimated ^{235}U abundances are predominantly low compared to the declared abundances. The HPGe results showcase the difficulties of using a peak ratio based approach to determine isotopic compositions. The large deviations of FRAM results with respect to known ^{235}U abundances is due to the inhomogeneous mixing of Th/Pa and U in the cylinders An enrichment meter based approach such as that used in the GEM code, would work much better.The fact that both the M400 CZT and HPGe measurements show large deviations with respect to known ^{235}U abundances indicates that the large deviations are not due to any shortcomings of M400 CZT.

3.3 ANALYSIS OF FIELD TEST SPECTRA USING THE GEM CODE

The UF_6 spectra were analyzed using the GEM code version 2.2.1 [reference]. The GEM code uses the infinite thickness approach and fits the responses to gamma and x-ray emissions from the uranium sample. The M400 detectors were shielded and collimated and located such that the source completely filled the field of view. Cylinder 59 was used for calibrating GEM measurements for each detector and for each location. This cylinder was chosen because it had a high enrichment (4.95 wt.%), was a full cylinder, and was filled clean (i.e., not over a heel). The results of GEM analysis of spectra collected using the LLNL detector are shown in Figures 19a, 19b, and 19c, corresponding to measurement locations L1, L2, and L3.

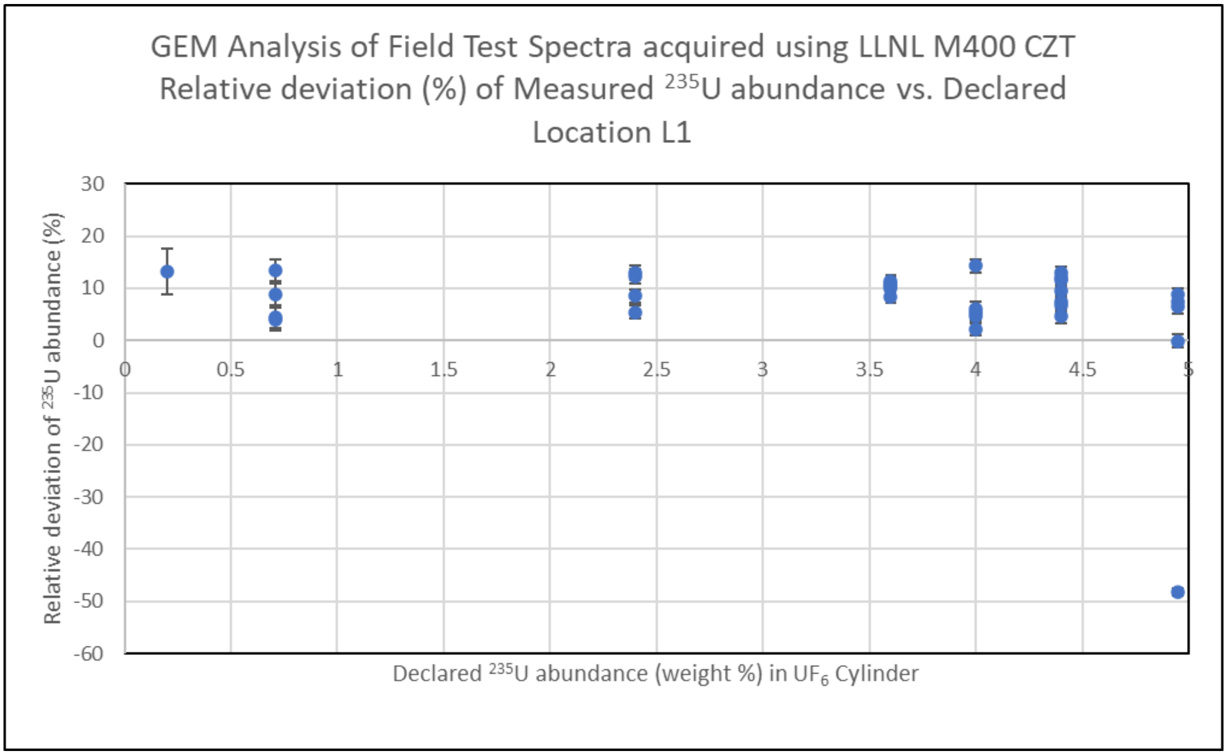

Figure 19a. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated LLNL M400 CZT at Measurement Location L1

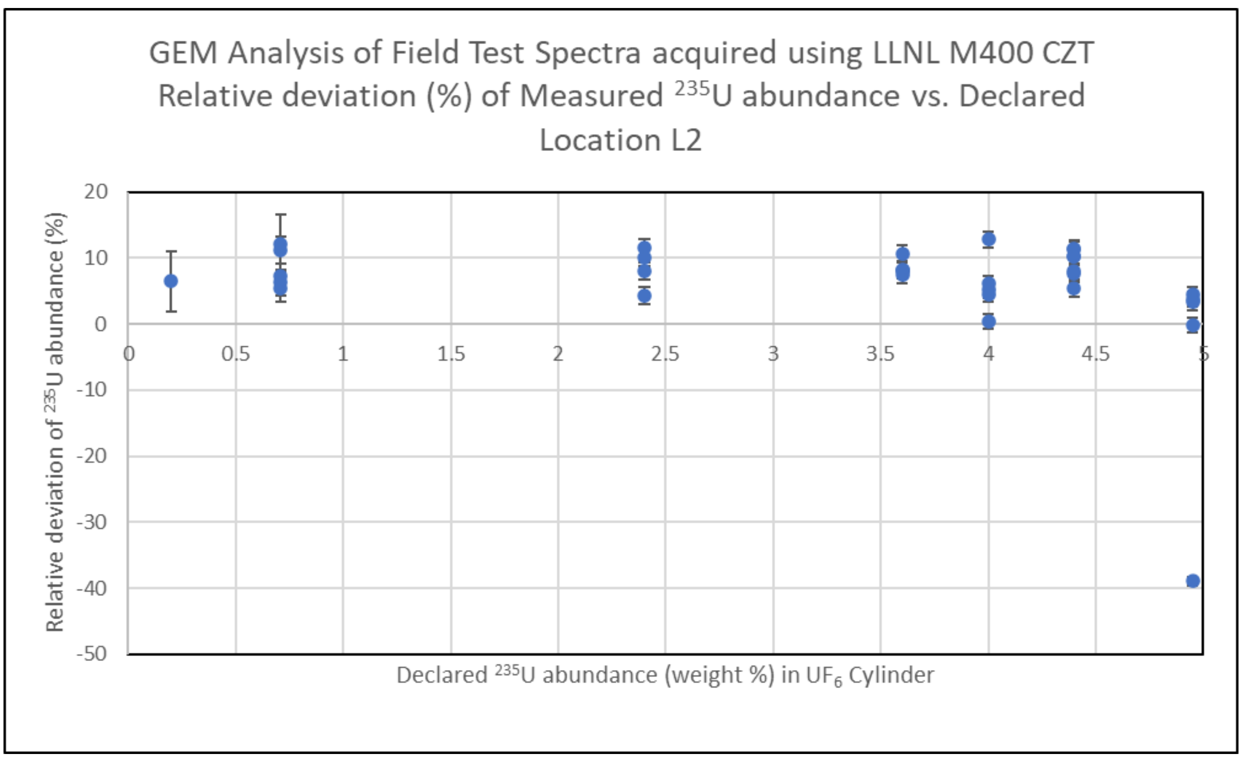

Figure 19b. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated LLNL M400 CZT at Measurement Location L2

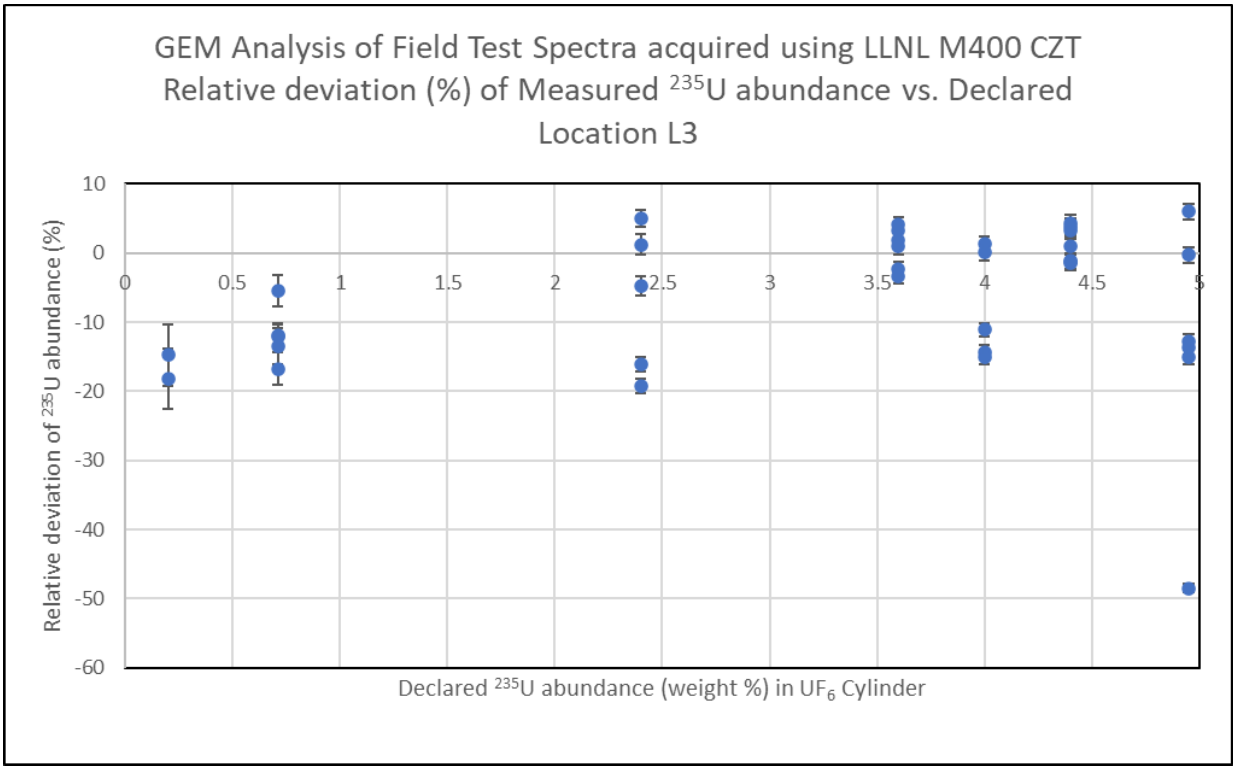

Figure 19c. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated LLNL M400 CZT at Measurement Location L3

It is observed that the GEM analysis result for cylinder ID 587 (4.95%) shows an anomalously high deviation of -48%, -38%, and -48% respectively for measurement data from locations L1, L2, and L3. UF_6 cylinder 587 was only partially filled. Therefore, the results had large negative errors for most detectors (>30%). The reason is, the detector placement was seeing a lot more headspace and less solid UF_6 than the other detectors, so fewer 186 keV photons/s. So, it effectively looked like a ~2.5wt.% cylinder. The results for cylinder 587 were considered to be outliers and were excluded from the statistical analysis.

The results of GEM analysis of spectra collected using the LLNL detector are shown in Figures 20a, 20b, and 20c, corresponding to measurement locations L1, L2, and L3.

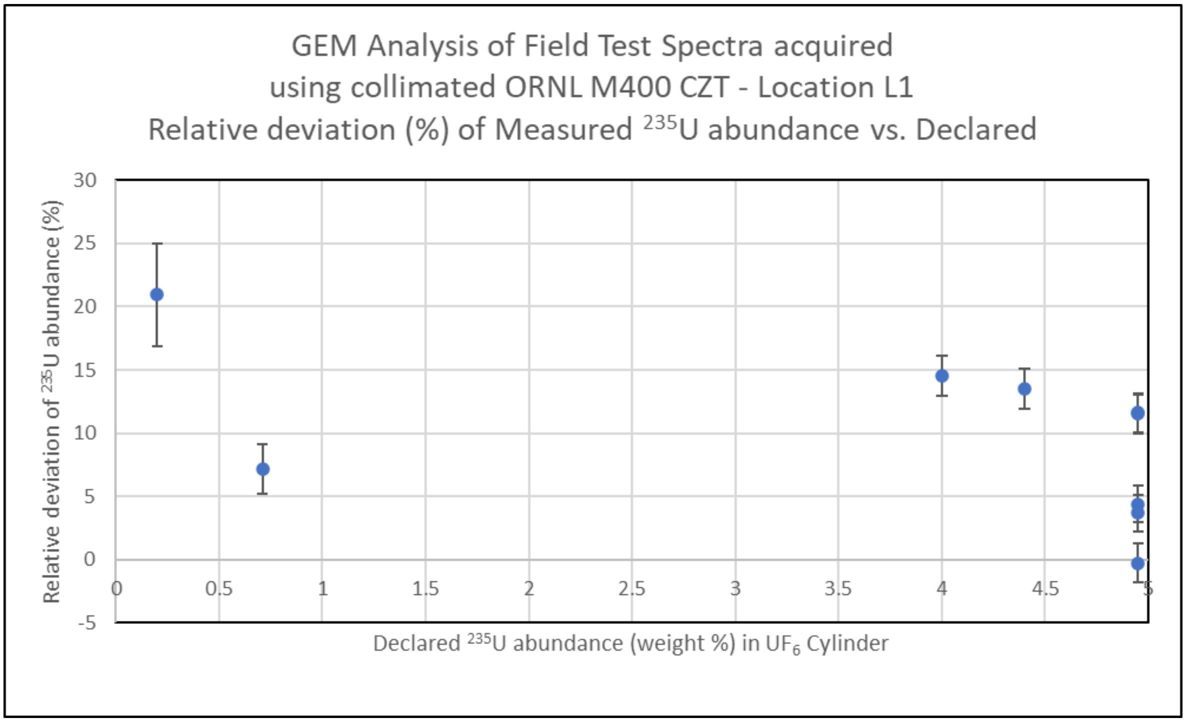

Figure 20a. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated ORNL M400 CZT at Measurement Location L1

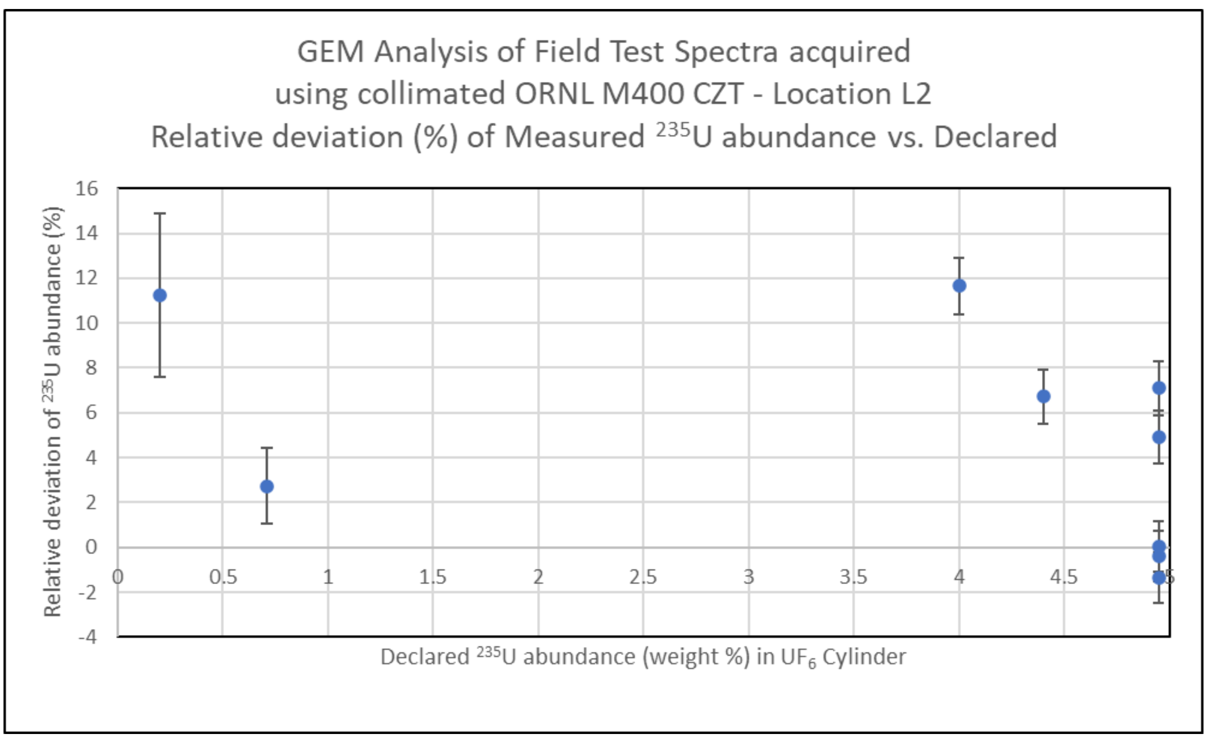

Figure 20b. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated ORNL M400 CZT at Measurement Location L2

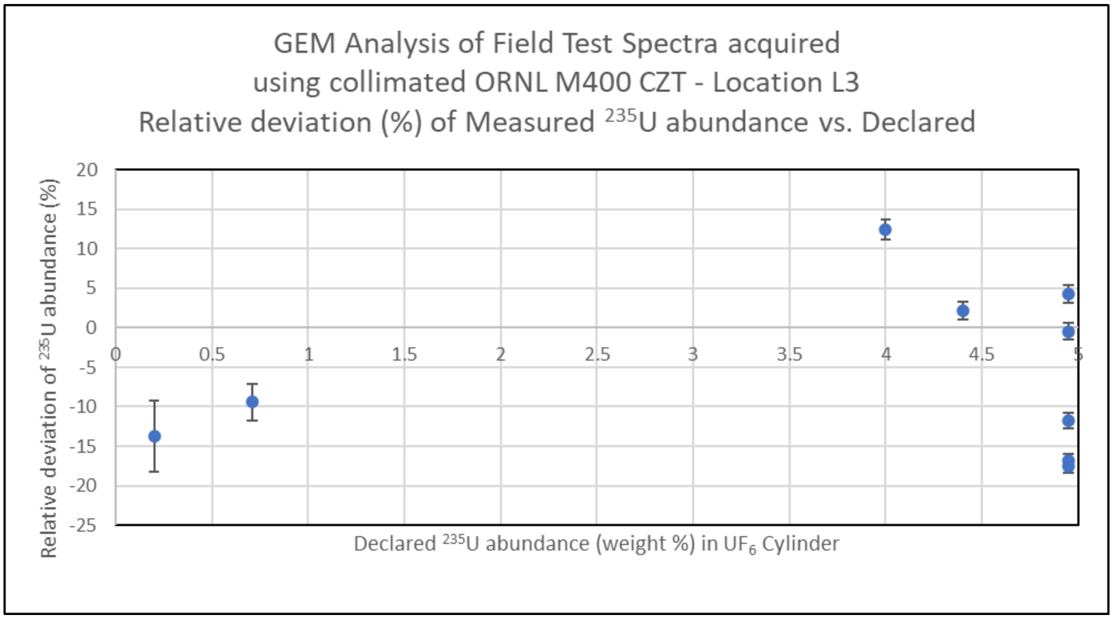

Figure 20c. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated ORNL M400 CZT at Measurement Location L3

The ORNL dataset is sparse since the detector experienced a failure due to water intrusion after the first week of field measurements. The deviation of enrichment results from the GEM code with respect to the declared values is within the -15% to +20% range.

The results of GEM analysis of spectra collected using the PNNL detector are shown in Figures 21a, 21b, and 21c, corresponding to measurement locations L1, L2, and L3.

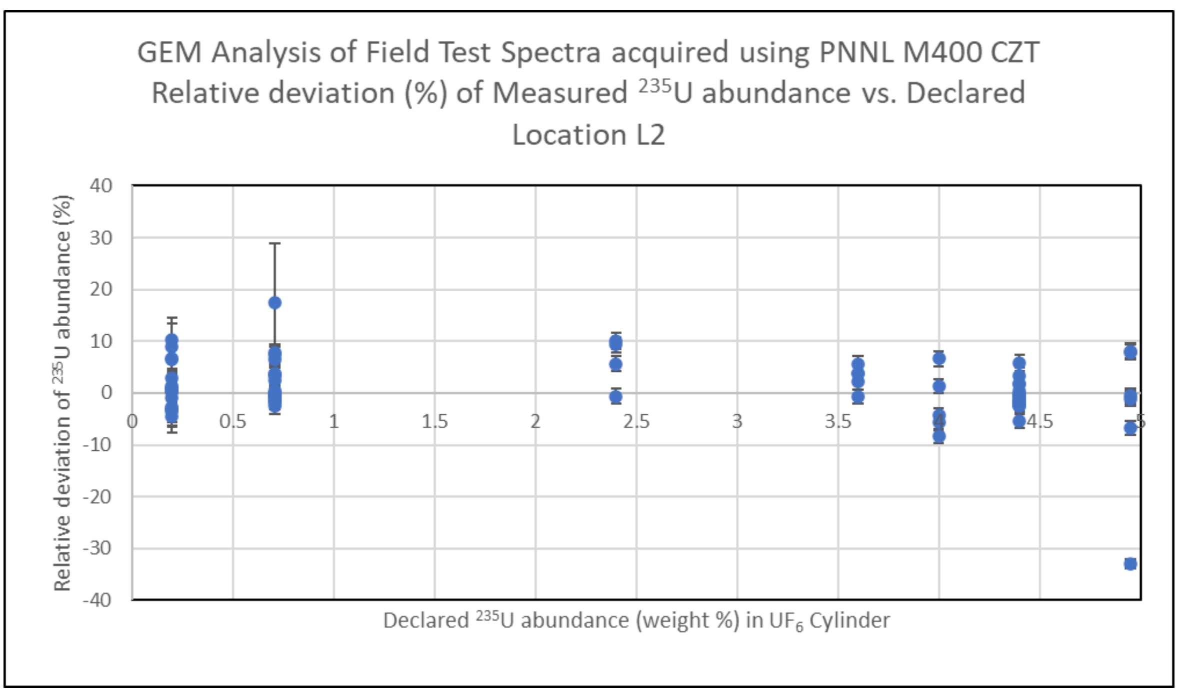

Figure 21a. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated PNNL M400 CZT at Measurement Location L1

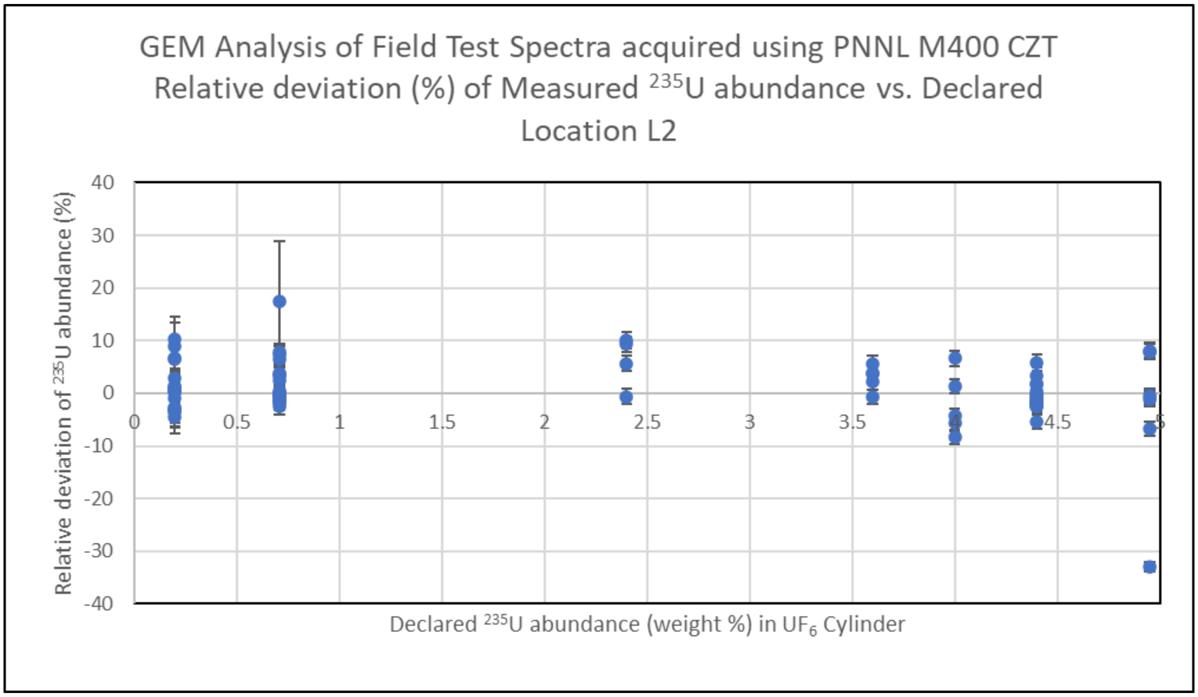

Figure 21b. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated PNNL M400 CZT at Measurement Location L2

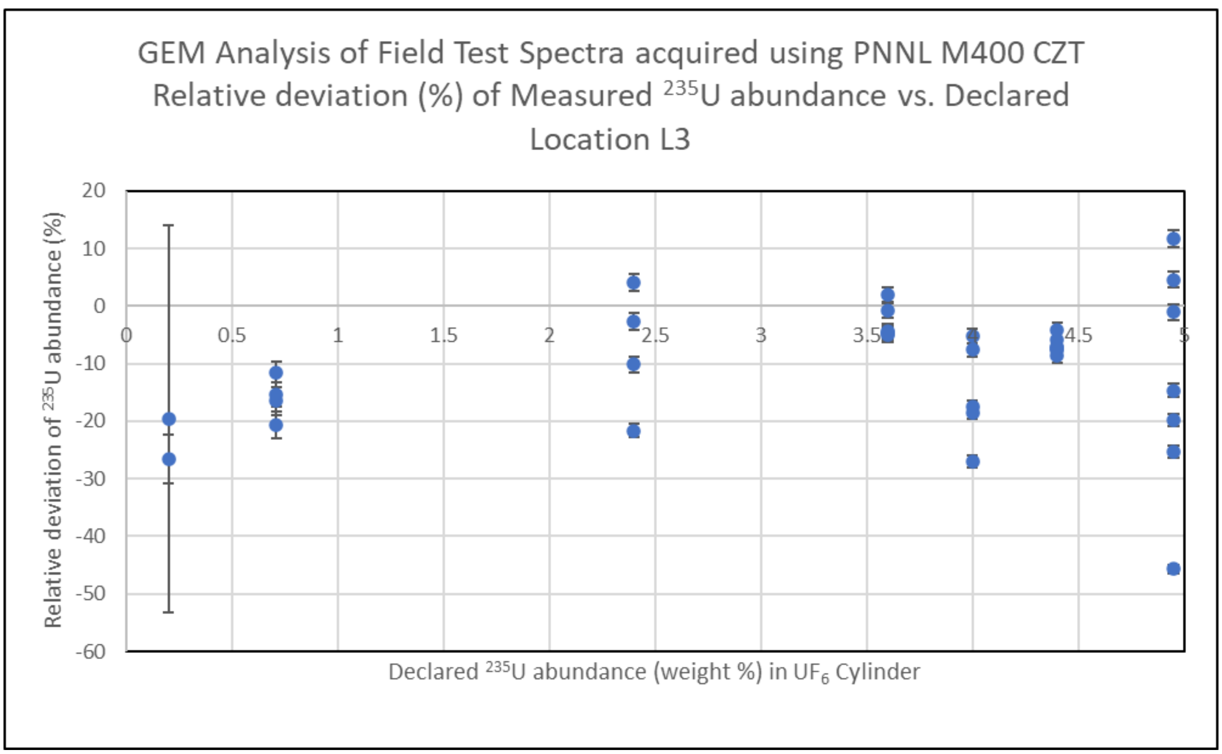

Figure 21c. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated PNNL M400 CZT at Measurement Location L3

The deviations for cylinder ID 587 (4.95%) are anomalously high; -43%, -33%, and -46% for measurements at L1, L2, and L3. It is interesting that the measurement results of the same cylinder using the LLNL M400 also showed similar anomalous deviations.

GEM analysis for HPGe spectra are given in Figure 22.

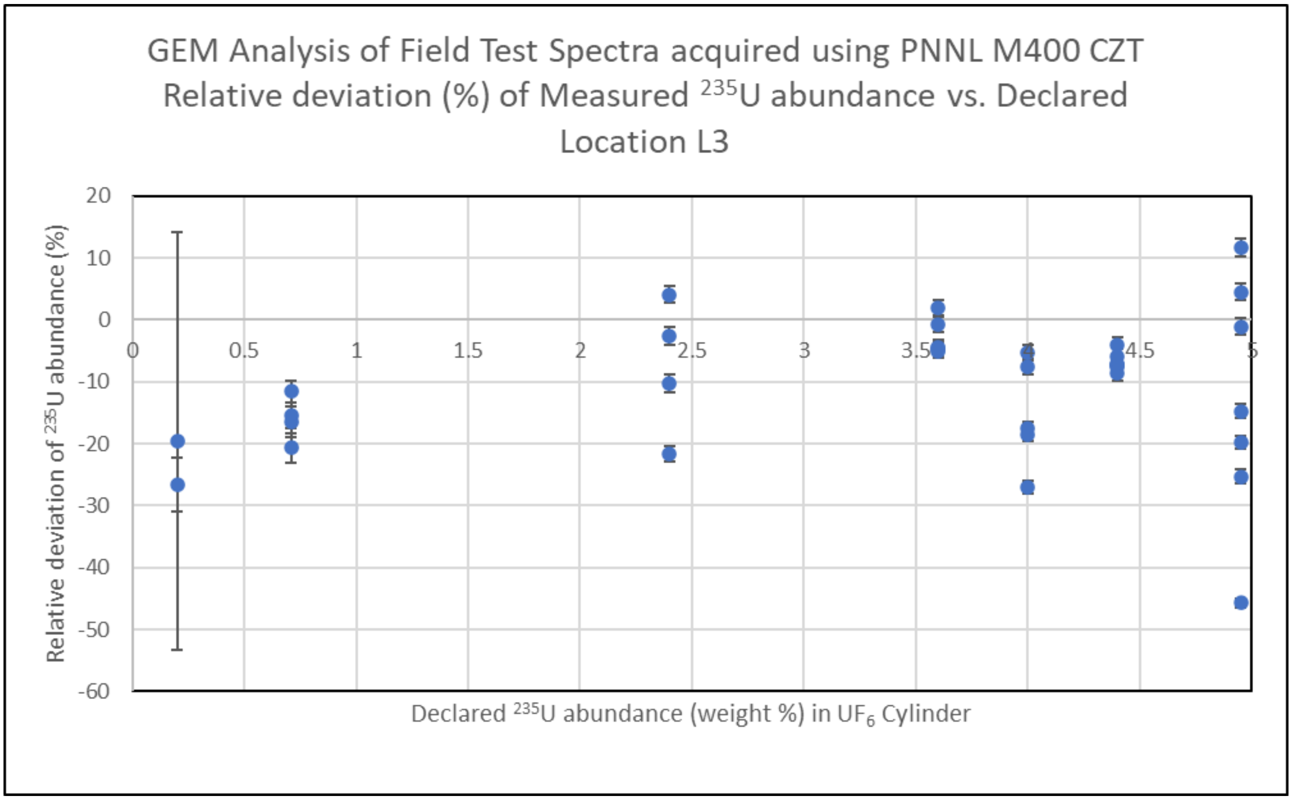

Figure 22. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated Electrically Cooled HPGe (SNL): Measurement Location L3

3.3.1 Discussion of results of GEM analysis on M400 CZT spectra

UF_6 cylinder 59 was used for calibrating GEM measurements for each detector and for each location. This cylinder was chosen because it had a high enrichment (4.95 wt.%), was a full cylinder, was filled clean (i.e., not over a heel), and had reached radioactive equilibrium with ^{234m}Pa. Since the GEM code uses the infinite thickness approach and relies on the net count rate at the 185.7 keV gamma-ray peak only, the secular equilibrium between ^{238}U and its daughters is not a requirement. However, for calibration purposes a cylinder which is "normal", (i.e.) that which does not contain freshly separated uranium, is used.

Relative standard deviations (RSD) were computed only for enriched uranium measurements. This is because, there were only a limited number of DU and NU cylinders. For some datasets the results from cylinder 587 were excluded since this cylinder was only partially filled.. This led to large negative deviations (under estimation of enrichment) for all measurements (>30%). Due to the detector placement with respect to the cylinder, the field of view was dominated by headspace and less solid UF_6 than the other cylinders. So smaller count rates were registered at the 185.7 keV peak, thus effectively underestimating a 4.95wt% enrichment material as ~2.5wt.%.

The LLNL data showed the lowest RSD, while ORNL and PNNL data yielded comparable RSD. The L3 locations always had larger RSDs, and this correlates with the wider variability in the wall thickness measurements for L3 locations among cylinders. As mentioned previously, the results for cylinder 587 were excluded, since the cylinder was only a partially filled cylinder and the measurement results therefore underestimated the ^{235}U abundance by approximately 40%!

Table 1 gives the mean and standard deviation of the relative deviation of GEM analysis results for ^{235}U abundance with respect to the declared for ^{235}U abundance. The mean and standard deviation of the relative deviations are given for low enriched uranium.

Table 1. GEM analysis results for ^{235}U abundance with respect to the declared for ^{235}U abundance

M400 CZT Detector	Mean and Standard Deviation of Relative Deviations		
	Location L1	Location L2	Location L3
ORNL	0.08433 ± 0.05725	0.04102 ± 0.04831	-0.03941 ± 0.11533
LLNL	0.08384 ± 0.03593	0.07218 ± 0.03477	-0.03405 ± 0.07760
PNNL	0.04869 ± 0.05702	0.00402 ± 0.04289	-0.08045 ± 0.09060

The relative standard deviations in Table 1 are compared against the uncertainties in the International Target Values 2020 (ITV2020)[6] tables for ^{235}U abundance in UF_6 cylinders, measured using a CZT detector and enrichment meter based analysis. According to ITV table 5B[6], the overall target uncertainty (ITV) for UF_6 measurements using CZT detector is ±3.6%. From the standard deviation of relative deviation results given in Table 1, the LLNL M400 measurements at L1 and L2 are within the ITV2020 values for UF_6. The ORNL and PNNL results exceed the ITV. All three detector results for location L3 exceed the ITV. The team has identified a few improvements that could be implemented.

1. The measurement geometry could be better optimized. Positioning the detector at the half-way point vertically at the side of the cylinder may not be the best geometry. If a cylinder is only half-filled, which it is typically. In addition, since the detector is offset by 1.5 inches with respect to the outer surface of the UF_6 cylinder, the field of view of the collimated M400 CZT will not be completely covered by sample.
2. Differences in wall thickness at the three locations has not been taken into account in GEM analyses.
3. The spectrum fitting functions in GEM v.2.2.1 are not tailored to the peak shapes M400 CZT.

3.4 ANALYSIS OF FIELD TEST SPECTRA USING THE CZTU CODE

Results from CZTU analyses are available for spectra collected using the LLNL M400 CZT detector. Since the CZTU code relies on the analyzing the low energy region of the spectra, an accurate fit to the Compton continuum was necessary, extending to energies below 60 keV. Since the shielding and collimator material made of tungsten impregnated polymer T-Flex was used, the presence of x-ray peaks from tungsten at 59 keV ($K_{\alpha2}$) and 63 keV ($K_{\alpha1}$) tends to perturb the continuum fit. Therefore a 3.175 mm thick copper filter was used in front of the LLNL M400 CZT to attenuate the tungsten x-rays.

A total of 184 UF_6 spectra were analyzed using the CZTU code. These included multiple spectra acquired at a given location and a given cylinder. The deviation of ^{235}U abundance determined using CZTU with respect to the declared ^{235}U abundance was plotted versus the declared ^{235}U abundance of the UF_6 cylinders. The results corresponding to measurements at locations L1, L2, and L3 are shown, respectively, in Figure 23a, 23b, and 23c.

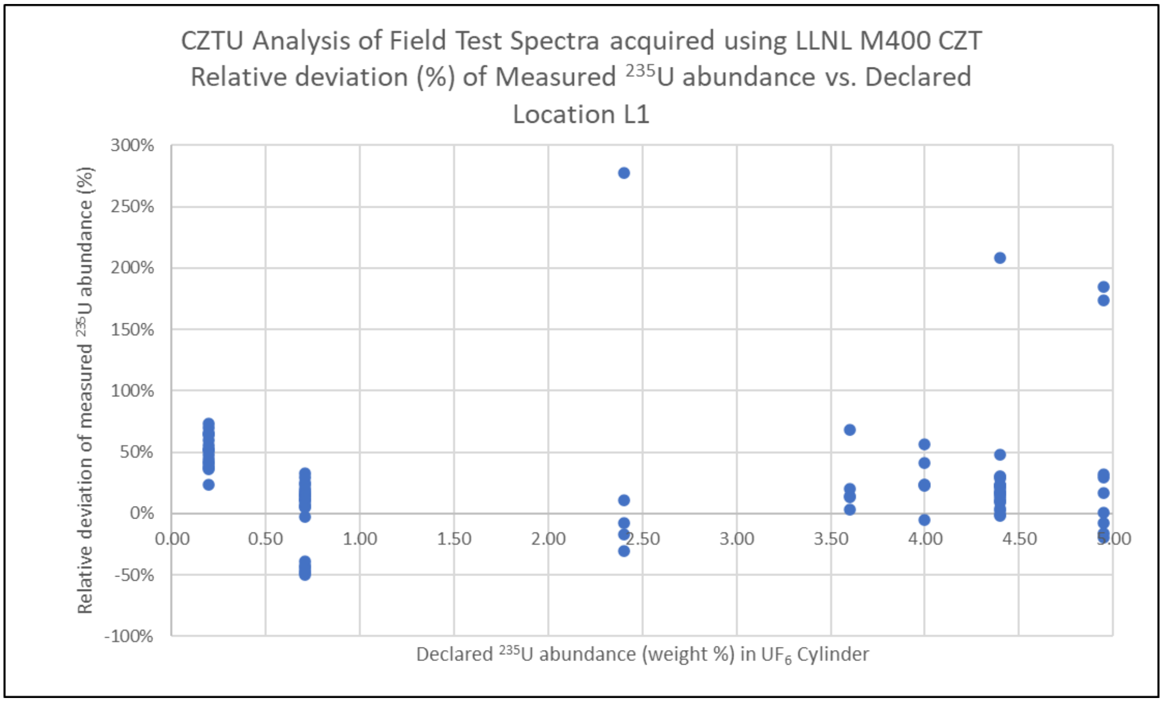

Figure 23a. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated LLNL M400 CZT at Measurement Location L1

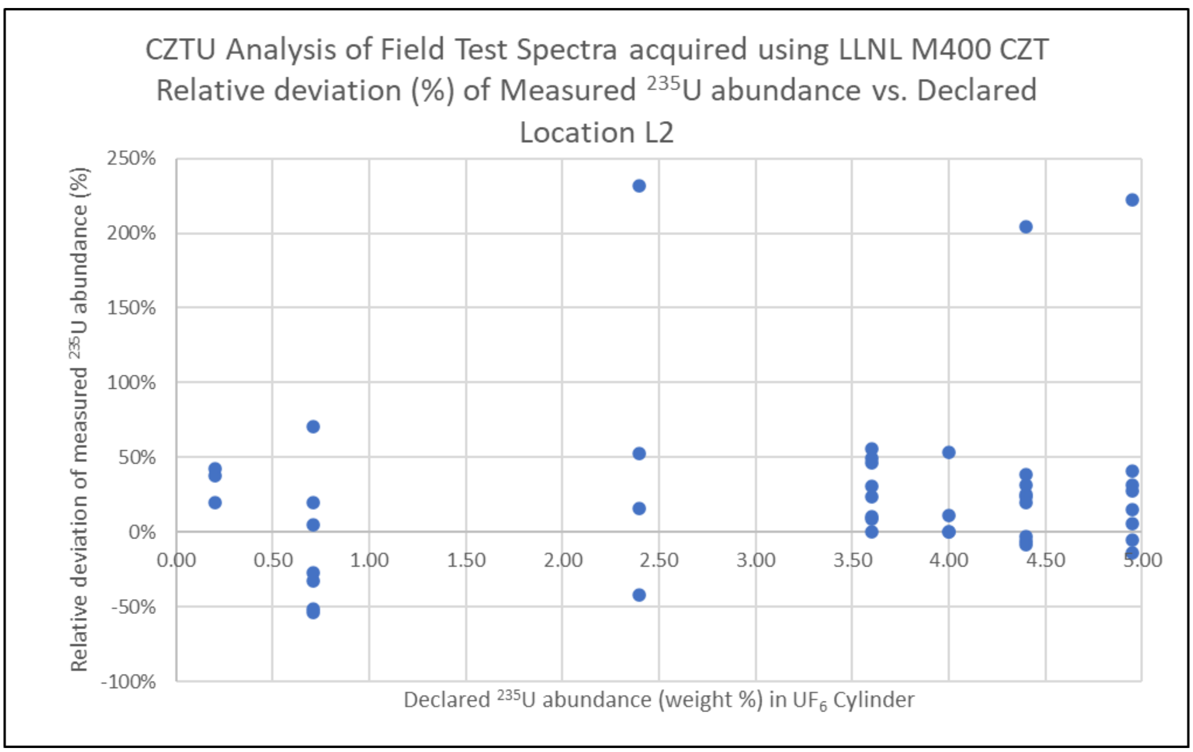

Figure 23b. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated LLNL M400 CZT at Measurement Location L2

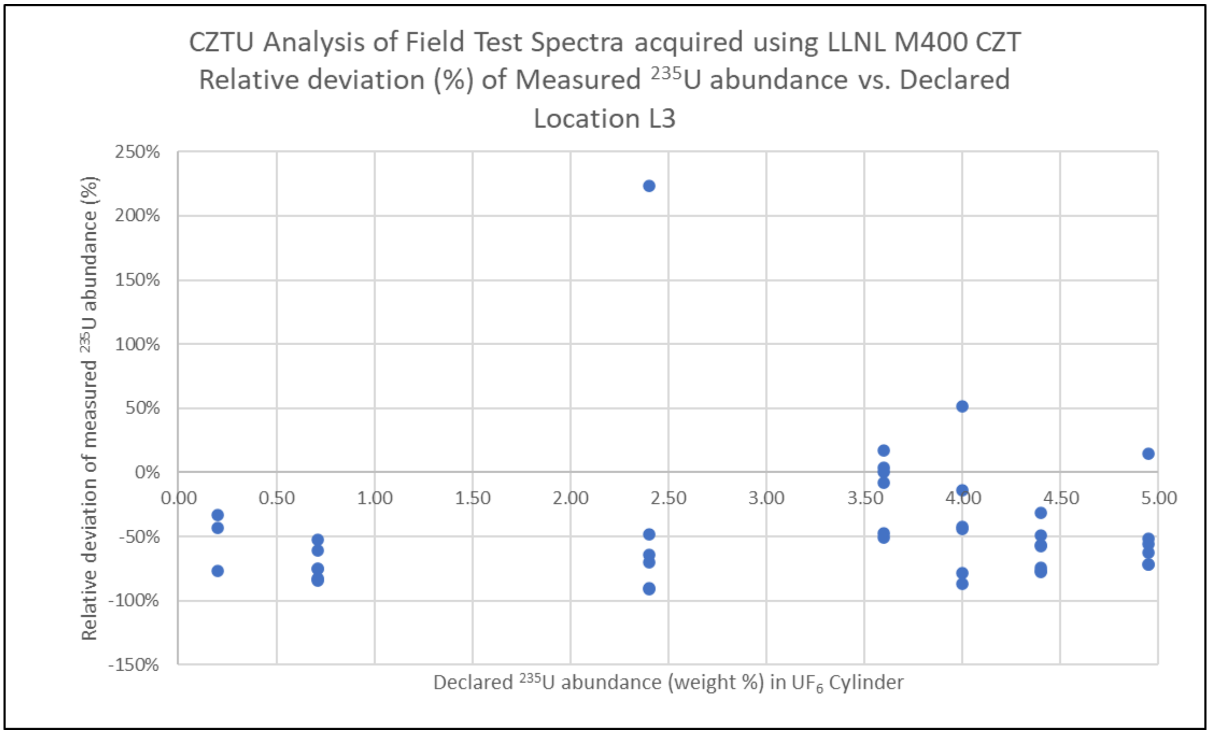

Figure 23c. GEM v2.2.1 analysis results for UF_6 spectra acquired using collimated LLNL M400 CZT at Measurement Location L3

Some of the ^{235}U abundance results from CZTU analysis show anomalously large deviations (> 200%) with respect to the declared ^{235}U abundance. In these cases, the gamma ray peaks from ^{234}Th were not significant enough to be quantified. For the CZTU analysis of spectra acquired at location L1, the mean and standard deviation of the relative deviations was 18.49% ± 27.72%. For measurement location L2 the the mean and standard deviation of the relative deviations was 14.82% ± 29.26%, and for location L3 the result was -51.34% ± 33.26%. The reason for such large relative deviations is, like FRAM, CZTU also works based on the peak ratio method. CZTU uses gamma emissions from ^{235}U, ^{238}U, and their daughters. Any inhomogeneities in the distribution of ^{238}U daughters at a measurement location inside the cylinder will impact the accuracy of the determination of ^{235}U abundance. Such inhomogeneities are characteristic of UF_6 distributed inside the cylinders.

4. TASK 3: SYNTHETIC SPECTRA GENERATION

This task was on hold for the duration of FY22.

5. TASK 4: PLUTONIUM SOURCE MEASUREMENTS USING M400 CZT

Measurements using plutonium sources were carried out at ORNL, LANL and LLNL.

The following plutonium sources are being measured at ORNL:

- PIDIE sources supplied from New Brunswick Laboratory (NBL).
    - A total of 7 sources with ^{239}Pu fraction ranging from 65.13% to 93.81% (Table 1).

- Plutonium tiles (10 sources), each with a mass loading of ~18.4 g of total Pu, and a ^{239}Pu fraction of 97.91%.
- A plutonium source with a 99.9% ^{240}Pu fraction.

Table 1. ORNL PIDIE Plutonium Standards

Source	**^{239}Pu wt%**
ORNL_PIDIE_1	93.8122
ORNL_PIDIE_2	89.4548
ORNL_PIDIE_3	84.8258
ORNL_PIDIE_4	78.2296
ORNL_PIDIE_5	76.4948
ORNL_PIDIE_6	68.8837
ORNL_PIDIE_7	65.1331

ORNL could not complete the measurements before the end of FY2022 because of problems encountered with the M400 detector in August 2022. The communication port on the M400 became faulty. The storage of spectra on the detector memory was also unreliable. Several days' worth of data was lost because of detector memory issues. The measurements had to be repeated. H3D replaced the micro-USB connection with USB-C connections, which are more robust. To circumvent the detector memory issues, H3D has configured data storage directly on to an external hard drive. Plutonium measurements will be completed in Q1 of FY23.

An example plutonium spectrum from ORNL PIDIE_1 is shown in Figure 19. PIDIE 1 is the lowest burn-up PIDIE standards with a ^{239}Pu weight % of 93.8122%. The standard was measured for 1800 seconds real time, at a distance of 5 cm from the uncollimated ORNL M400 detector. A 1 mm thick Cd filter was used to reduce the intensity of the 60 keV gamma ray emitted by ^{241}Am.

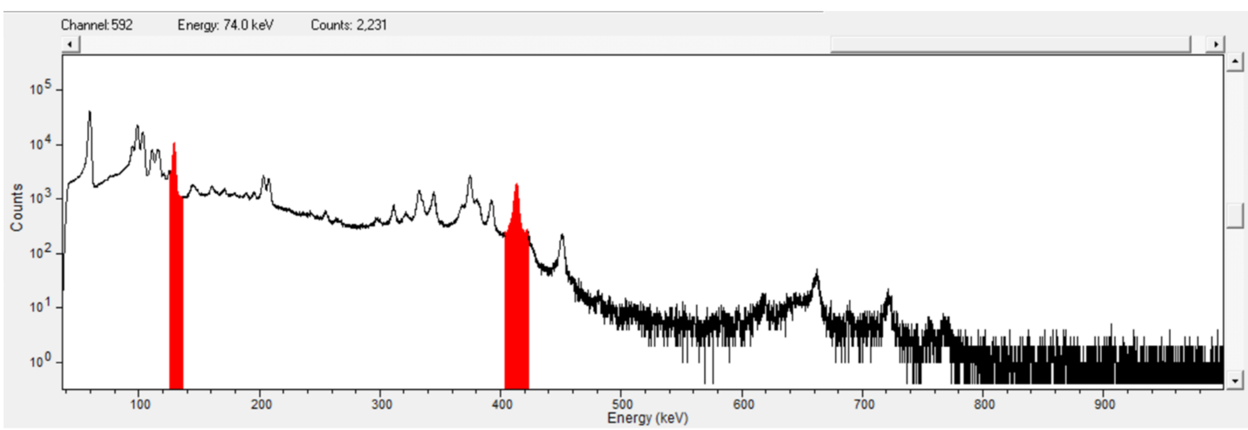

Figure 19. Plutonium spectrum acquired using ORNL PIDIE Source 1, at distance of 5 cm from M400 CZT. The 129 keV and 414 keV peaks from ^{239}Pu are highlighted.

Figure 20 shows an example gamma ray spectrum from PIDIE 7 which has the highest burn up among the ORNL PIDIE standards with a ^{239}Pu weight % of 65.1331%. The source to detector distance was 5 cm, the counting time was 1800 seconds real time, and a 1 mm thick Cd filter was used to reduce the intensity of the 60 keV gamma ray from ^{241}Am.

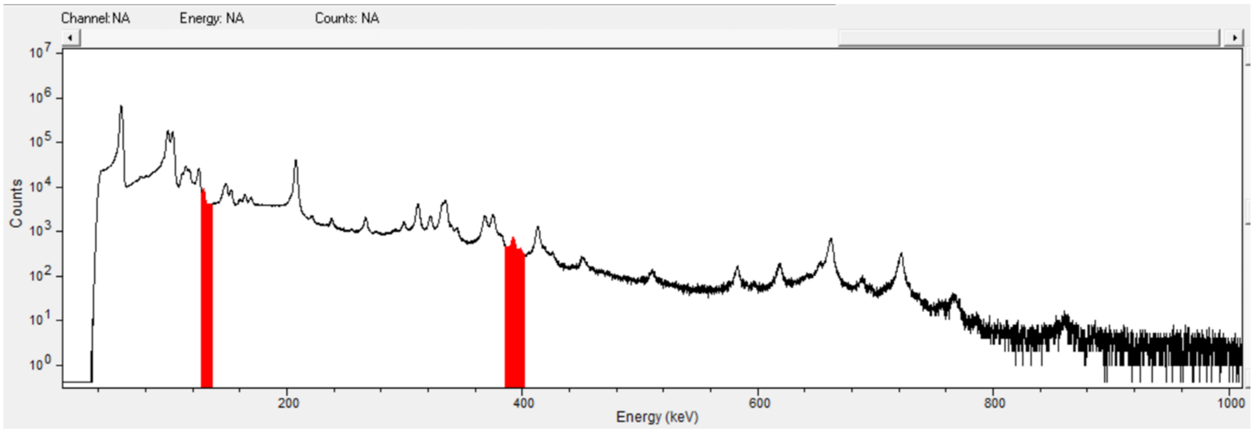

Figure 20. Plutonium spectrum acquired using ORNL PIDIE Source 7, at distance of 5 cm from M400 CZT. The 129 keV and 414 keV peaks from ^{239}Pu are highlighted. Note the relatively low intensities of the ^{239}Pu peaks when compared to PIDIE 1 shown in Figure 19.

The list of plutonium sources being measured at LANL are given in Table 2.

Table 2. Plutonium sources measured at LANL

Source	^{240}Pu %	Pu mass (g)
LANL_PIDIE_A	5.9	0.4
LANL_PIDIE_B	21.2	0.4
LANL_Pu_1	26.9	5.4
LANL_Pu_2	19.1	5.5
LANL_Pu_3	14.3	5.8
LANL_Pu_4	6.3	5.8
LANL_Pu_5	16.3	59
LANL_Pu_6	16.5	170
LANL_Pu_7	16.5	606
LANL_Pu_8	16.4	537
LANL_Pu_9	9.7	149
LANL_Pu_10	3.5	20
LANL_Pu_11	3.5	30
LANL_Pu_12	3.5	50
LANL_Pu_13	3.5	11
LANL_Pu_14	6.1	8
LANL_Pu_15	6.9	12
LANL_Pu_16	11.8	20
LANL_Pu_17	15.4	12

In addition, three MOX sources are also being measured at LANL (Table 3).

Table 3. Isotopics of MOX sources measured at LANL

Source	^{240}Pu %	Pu mass (g)	U/Pu	^{235}U %
LANL_MOX_1	5.9	0.4	8.2	93.2
LANL_MOX_2	5.9	1	3.2	93.2
LANL_MOX_3	11.7	113	6	1.1

6. TASK 5: CODE DEVELOPMENT AND ANALYSIS OF M400 PLUTONIUM SOURCE SPECTRA

LANL configured two FRAM parameter sets to analyze M400 CZT plutonium spectra in several different energy regions. The parameter sets are mid-range 220-420 keV and high energy range 180-1010 keV. LANL modified FRAM code to improve the analysis.

GADRAS programming updates were made to the relative efficiency curve application to provide more flexibility to the user and technical method improvements (SNL). GADRAS programming updates to consolidate/add analysis methods for uranium and plutonium isotopic analysis for better ease of use (SNL).

Plutonium analysis will be completed in Q1 and Q2 of FY23 after all the Pu measurements have been completed and the spectra have been distributed to the analyzing laboratories.

7. CONCLUSIONS

The measurement and analysis tasks using uranium standards of known enrichments (uncollimated and collimated geometries) were completed in FY22. These tasks were on the FY21 PWP but could not be completed in FY21 because of detector problems and repairs. The uranium measurement results were included in an updated version of FY21 annual report. A paper was presented at the INMM 2022 Annual Conference on the uranium measurement using M400 CZT detectors and analysis results from GADRAS, FRAM, GEM, and CZTU.

PNNL led the efforts to conduct the field testing at the Framatome Fuel Fabrication Facility in Richland, WA. PNNL drafted a field test plan, negotiated the contract with Framatome, and executed the logistics of the field testing task. Field testing was conducted at the Framatome facility for two weeks in April 2022 using thirty-four 30B UF_6 cylinders. Three M400 CZT and an electrically cooled High Purity Germanium (HPGe) detector were used in the measurements. Spectra were distributed to the participating laboratories. Analysis results from field test spectra are available from GADRAS,FRAM, CZTU, and GEM codes. The GADRAS analysis results showed a consistent low bias, with the average deviations of measured ^{235}U abundances low by 68% to 75% with respect to declared abundances. The low bias was present in the electrically HPGe results too, indicating that the reason for the low bias was not necessarily the because of the unusual peak shapes generated by the M400 CZT. Rather, the low bias mostly due to the inhomogeneous distribution of thorium and protactinium daughters of ^{238}U, and not being co-located as ^{235}U. The results from FRAM showed smaller average deviations, but the standard deviation of the measured versus declared ^{235}U abundances was much higher, on the order of ±50%. FRAM employs a peak ratio method, and so the results are impacted by the inhomogeneous distribution of thorium and protactinium daughters of ^{238}U, and not being co-located as ^{235}U. The CZTU code, like FRAM, also employs a peak ratio based approach. Therefore the CZTU also uses gamma ray emissions from ^{235}U as well as ^{238}U and its daughters. Hence the inhomogeneities in the UF_6 distribution impact the CZTU analyses and result large deviations, similar to what is observed in the case of FRAM. Not surprisingly, the ITV tables do not consist of uncertainty values for UF_6 spectral analysis performed using peak ratio methods. It is because, the peak ratio methods are not ideal for analyzing gamma-ray spectra acquired from UF_6 cylinders. The ^{235}U abundance results from GEM analyses showed the lowest deviations with respect to the declared ^{235}U abundances. The standard deviations of relative deviations for the LLNL M400 measurements at locations L1 and L2 were within the ITV2020 for UF_6 with low enriched uranium, measured using CZT detectors and analyzed using the GEM code. Recommendations have been made for improving the GEM analyses. These include, better optimization of the measurement geometry, taking into consideration the differences in wall attenuation, and using an updated version of GEM that has the capability to fit the M400 CZT peak shapes.

Plutonium measurements are being carried out at ORNL, LANL, and LLNL. FRAM and GADRAS codes have been updated to be able to analyze plutonium spectra. Analysis results will be available in Q2 of FY23.

8. FY23 WORK

In FY23, the plutonium analysis capability of the codes will be further refined. The spectral data collected in FY22 will be utilized. Analysis results from the refined codes, namely FRAM, GADRAS, and CZTPu will be provided to ORNL by LANL, SNL, and LLNL. ORNL will perform an ANOVA analysis and establish the random and systematic uncertainties for plutonium analysis using M400 spectra.

For the review and presentation of the results from the Gamma Rodeo project, a three and a half day (3.5 days) workshop is proposed at the end of FY23. The workshop will be held at ORNL. The workshop will bring together the DOE NNSA sponsors, subject matter experts from the IAEA and the IRSN (France), the PIs of M400 related projects that are in progress at various DOE laboratories., and experts from the company H3D, Inc., which manufactures the high resolution CZT detectors. The workshop will be an excellent forum to obtain feedback from the IAEA and IRSN, gain further insights from H3D, Inc., into the characteristics of the high resolution CZT, and to exchange lessons learned from the different M400 projects at various DOE laboratories.

9. ACKNOWLEDGEMENT

The work done under this project titled “Gamma Spectrometry Code Rodeo for Uranium Enrichment” (WBS #24.1.3.1) was sponsored by the United States Department of Energy (U.S. DOE) National Nuclear Security Administration (NNSA) Safeguards Technology Development Program NA-241. The project team gratefully acknowledges the support of our sponsors.

ORNL gratefully acknowledges the contributions by collaborators from the participating DOE laboratories.

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APPENDIX A. List of 30B UF_6 Cylinders used in Field Testing at the Framatome Fuel Fabrication Facility, Richland, WA

ALT_ID	STATUS	NOM_ENR	U_WEIGHT	U235_WEIGHT	RECV_DATE	Cyl. RECERT valid thru	GROSS_WHT	TARE_WHT
57	FULL	4.40%	1435.521	63.163	04-Jan-21	1-Jul-24	2752	628.7
58	FULL	4.40%	1426.514	62.767	04-Jan-21	1-Oct-24	2744.6	634.2
60	FULL	4.40%	1428.88	62.871	04-Jan-21	1-Oct-24	2749.5	635.6
61	FULL	4.40%	1447.275	63.68	04-Jan-21	1-Jul-24	2769.1	628.2
62	FULL	4.40%	1429.977	62.919	04-Jan-21	1-Aug-24	2745.7	630.6
96	FULL	4.00%	1494.974	59.814	02-Jan-13	1-May-08	2834.8	623.2
99	FULL	4.00%	1510.678	60.246	14-Oct-21	1-Jul-17	2890.5	656.1
100	FULL	4.95%	1513.179	74.917	15-Feb-22		2880.3	642.2
102	FULL	4.95%	1508.379	74.755	17-Jan-22	1-Jul-23	2876.7	645.7
103	FULL	4.00%	1513.044	60.582	06-Jul-21		2885.4	647.5
104	FULL	4.95%	1510.61	74.851	17-Jan-22	1-Mar-14	2883.1	648.8
105	FULL	4.00%	1512.774	60.587	06-Jul-21	1-Jun-24	2891.1	653.6
59	FULL	4.95%	1526.729	75.573	08-Apr-21	1-Jan-25	2902.6	644.1
339	FULL	4.95%	1512.503	75.05	13-Dec-21		2858.5	621.4
108	FULL	4.00%	1507.297	60.367	15-Jun-21	1-Sep-20	2863.5	634.1
121	FULL	3.60%	1513.991	54.7	14-Oct-21	1-Aug-25	2861.1	621.8
122	FULL	3.60%	1511.895	54.61	14-Oct-21	1-Aug-25	2854.1	617.9
125	FULL	3.60%	1510.137	54.531	14-Oct-21	1-Aug-25	2854.5	620.9
126	FULL	3.60%	1510.543	54.455	14-Oct-21		2858.3	624.1
129	FULL	3.60%	1507.095	54.301	14-Oct-21		2844.4	615.3
285	FULL	0.71%	1518.385	10.796	11-Oct-21	1-May-20	2879.4	633.6
286	FULL	0.71%	1507.906	10.721	19-Jan-21	1-Jan-21	2875.4	645.1
287	FULL	0.71%	1503.984	10.693	11-Oct-21	1-May-20	2856.7	632.2
288	FULL	0.71%	1509.934	10.736	19-Jan-21	1-Apr-22	2889.1	655.8
332	FULL	2.40%	1512.977	36.705	07-Feb-22	1-Apr-24	2862.9	625.1
337	FULL	2.40%	1510.475	36.705	19-Jul-21	1-Dec-22	2884.7	650.6
338	FULL	2.40%	1510.475	36.735	19-Jul-21		2853.1	619
470	FULL	0.20%	1492.351	2.929	22-Dec-14		2870.8	663.8
476	FULL	0.20%	1494.786	2.934	22-Dec-14		2851.6	641
587	FULL	4.95%	465.968	23.065	10-Aug-21		1312.8	623.6
306	FULL	4.40%	1499.117	65.871	07-Mar-22	1-Apr-14	2852.8	635.5
1001								
	EMPTY							
1000								