Uranium Enrichment Determination with Commercial CZT Detectors and a Non-proprietary Analysis Algorithm

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Abstract

Sandia National Laboratories, in partnership with the Y-12 National Security Complex and Lawrence Livermore National Laboratory, has worked to develop a generalized method for isotopic analysis of enriched uranium using commercial cadmium-zinc-telluride (CZT) detectors and a non-proprietary analysis algorithm. Measurements of a number of different enrichment levels of uranium were made at the Y-12 Nuclear Detection and Sensor Testing Center Site 2, using the site's uranium enrichment standards. These standards were designed with enrichment levels that bracket important enrichment thresholds to assess the accuracy of new measurement systems. These measurements were made using a commercially available 1 cubic centimeter CZT detector and custom collimator with the goal of developing an accurate system which utilizes the "enrichment meter" method. Recent improvements in CZT detector systems, both in crystal size and resolution, make these detectors practical in this application and point the way towards more portable and affordable equipment for potential arms control monitoring applications. The HYPERMET peak detection algorithm, which was first developed for high-purity germanium detectors at the U.S. Naval Research Laboratory, was evaluated for application to the CZT spectra obtained during the measurements. HYPERMET is a well-known algorithm which uses a zero-area filter and a non-linear least-squares fitting routine to provide more accurate peak area calculations, and showed excellent results in determining enrichment levels of the Y-12 standards. Measurements at Y-12 included the addition of attenuator materials to simulate the effects of containers and variations in counting times to determine the minimum measurement time for accurate enrichment determination. Software deployable to desktop or embedded computer systems has been completed and tested. This work was sponsored by the National Nuclear Security Administration Office of Nuclear Verification.

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Introduction

Government and industry research into detector materials has advanced the state of the art in passive detection in recent years. Detector materials such as CZT (Cadmium-Zinc-Telluride) provide good resolution, approaching 1% at room temperature, enabling isotopic analysis based on resolving spectral peaks. However, commercial algorithms are typically based on finding and identifying single peaks. While a single peak detection method works fairly well for the 0.1% resolution High-Purity Germanium systems, it begins to fail when a lower resolution spectrum produces doublets and triplets from actinides. Sandia National Laboratories has used a previously published technique called HYPERMET to build an open source analysis code for determining the presence of highly enriched uranium (HEU) and also determining enrichment levels. The strength of this technique compared to other peak analysis methods is an optimization routine that is effective at fitting peaks, and then finding residuals that may be adjacent energy peaks (doublets or triplets) in the spectrum.

In partnership with the Y-12 National Security Complex and Lawrence Livermore National Laboratory, we worked to develop a generalized method for isotopic analysis of enriched uranium using commercial CZT detectors and an implementation of the HYPERMET analysis algorithm. Measurements of a number of different enrichment levels of uranium were made at the Y-12 Nuclear Detection and Sensor Testing Center Site 2, using the site's uranium enrichment standards. These standards were designed with enrichment levels that bracket important enrichment thresholds to assess the accuracy of new measurement systems. Our measurements were made using a commercially available 1 cubic centimeter CZT detector and custom collimator with the goal of developing an accurate system which utilizes the "enrichment meter" method. Measurements at Y-12 included the addition of attenuator materials to simulate the effects of containers and variations in counting times. Software deployable to desktop or embedded computer systems has been completed and tested. This software automatically analyzes measured spectra and reports both the presence of enriched uranium and the enrichment level.

We believe that the recent improvement in CZT detector size and resolution, combined with our new analysis software, offers an option for more portable and affordable equipment in potential arms control monitoring applications. Our work is sponsored by the National Nuclear Security Administration Office of Nuclear Verification.

Algorithm

The original computer program HYPERMET[i] was developed at the Naval Research Laboratory for "automatic and efficient analysis of multichannel pulse height spectra from high-resolution germanium gamma-ray detectors." HYPERMET was a great improvement over previous codes because of its ability to automatically identify peaks in gamma-ray spectra that—although discernible to the human eye—posed great difficulty for computers. This improvement was achieved through the use of a square-wave-like zero area filter, which when convolved with the measured spectrum effects a transform from the data to the negative smoothed difference. The resulting function is normally distributed about zero, except in the vicinity of a peak or sharply varying feature.

Our modern implementation of HYPERMET was developed in C++. Our initial algorithm can be described by the following three procedures:

1. Zero-area filter convolution identifies location of peaks in the spectrum.
2. Each peak is fit to an analytical model.
3. If a peak shape is found to be irregular (i.e. too broad), an iterative search is conducted for additional peaks in the same area.

Step 3 in the above procedure list is a critical solution to the problem of identifying doublets in the gamma-ray spectrum that may be too close to resolve by eye. One of the great strengths of HYPERMET

is its ability to resolve doublets, gamma-ray peaks too close to be resolved by eye. As stated above, HYPERMET identifies the location of peaks in the gamma-ray spectrum and then attempts to fit a semi-empirical analytic model peak shape. If a good fit to the data is found, the routine moves on to the next identified peak in the spectrum. If a good fit is not found, a search is made for residuals, the resulting difference between the fit and the data at each channel. Locations of peaks in the residuals are then included in the fitting algorithm as possible locations of additional peaks. If the inclusion of such peaks lowers the chi-squared value of the fit, an additional peak is identified and included as a real gamma-ray peak in the reported results.

For this project, data analysis was focused on the identification of HEU through detection of characteristic gamma peaks, and accurate calculation of the ^{235}U 186 keV peak area to determine enrichment. HYPERMET was predicted to achieve more accurate peak area calculations with CZT spectra due to its ability to identify sub-peaks, which in turn would lead to a more accurate determination of enrichment levels in uranium standards. Although HYPERMET has been used previously to analyze CZT spectra[ii, iii], our work has focused on the automated analysis required to identify isotopes and enrichment.

Software Data Processing

In preparation for an accurate enrichment calculation, the software must be provided with one or more calibration datasets. An initial "best guess" linear calibration is provided so that the relationship between channel number and energy is not too disjointed. The user selects a calibration waveform, which typically contains a known isotope with distinct peaks, and the associated calibration peaks (typically the most prominent). Using the estimated calibration, the algorithm will attempt to assign the correct channel number to the prominent peaks by choosing the channel at the maximum amplitude within a sub region around the estimated peak location. The peak channel numbers are validated and/or modified by the user via the software plotting capability, which provides the ability to plot both counts per keV and counts per channel. In many cases, a single calibration waveform is insufficient to span the required spectrum range, so the software allows multiple waveforms to be utilized. One combination that was found to be effective was using the 122 keV and 136 keV peaks from ^{57}Co in conjunction with the 662 keV peak from ^{137}Cs. The calibration algorithm uses a least squares fit to determine the linear mapping between channel number and energy.

The original implementation of HYPERMET was intended for HPGe which has very narrow peaks compared to CZT. The following modifications were made to the peak detection parameters for this new application:

- Increased the size of the FWHM (Full Width Half Maximum) from 3 channels to 15.
- Increased the width of the peak detection filter from 5 channels to 19.
- Utilized a smaller energy region. HPGe detectors can typically measure up to 3 MeV, however, our CZT detector is limited to 1.8 MeV and 4096 channels. The target region was further limited to approximately 80 – 240 keV since this range encompasses the 186 keV peak of interest for ^{235}U.
- Due to the decreased resolution, the number of peaks within the target region is limited to 4 in order to reduce the possibility of spurious peaks being detected and to increase the speed of the algorithm.

Since there are nine parameters used for the peak fitting, special care has to be taken when selecting the upper and lower bounds on each parameter, otherwise undesirable results can be observed. In particular, each peak is composed of the following:

- Main Gaussian curve
    - Lower bound: no less than 10% of the maximum amplitude measured

    - Upper bound: no greater than the maximum amplitude measured
- Approximate peak width (δ): FWHM +/- 25% of FWHM
- Short left tail and step functions: range from 0 to at most 75% of maximum amplitude measured
- Slope of the large and small exponentials:
    - Lower bound: 0
    - Upper bound: 2/[max δ value]
- Long left tail: ranges from 0 to at most 1.5% of maximum amplitude measured
- Y-intercept of linear continuum can vary from -20% to +10% of the estimated intercept using the endpoints of the analysis range.
- Slope of linear continuum can vary by +/- 20% of the estimated intercept using the endpoints of the analysis range.

In order to determine the enrichment of the given sample, the following steps are performed:

- Use the HYPERMET algorithm to extract peaks and positively identify ^{235}U.
- Calculate the area of the 186 keV peak using analytic integration.
    - The analytic model of the peak has the continuum subtracted so that the peak area calculation is not strongly affected by the background spectrum.
- Convert the area to an enrichment percentage by comparing the value to the 186 keV peak area of a known enrichment measurement.
    - A calibrated standard of 93.2% ^{235}U was previously analyzed using the HYPERMET algorithm. The calculated area of the 186 keV peak is used for the enrichment calculation of unknown samples.

The enrichment value (in %) is: $PA_U/PA_K * 93.2$
Where:

- PA_U = 186 keV Peak Area of Unknown ^{235}U spectrum
- PA_K = 186 keV Peak Area of Known 93.2% enrichment ^{235}U spectrum (calibration standard)

Detector Description

We used a commercially available CZT detector, the ***iGEM™,*** from eV Products, Inc., which is an OEM spectrometer "engine" suitable for integration into custom systems. For our measurements, we used one which was packaged in a demonstration configuration and used vendor-supplied software to obtain and record spectra on a laptop computer. The ***iGEM™*** detector utilizes a 10x10x10 mm (1 cm^3) CZT coplanar grid detector and records spectra in 4096 channel files. The detectors purchased for these measurements showed improved resolution over previously purchased CZT detectors. The detector used for this analysis was characterized and the resulting resolution was 1.5% FWHM at 662 keV.

Figure 1: iGEM™ Detector Module

Shielding Enclosure

In order to perform spectrum measurements for the "enrichment meter" method, we constructed a shield and collimator assembly to reduce the field of view of the detector, similar to the shield described for the US-Russian HEU Transparency Program[iv]. The enclosure and collimator blocks are made of an alloy of 80% bismuth and 20% tin with a material thickness of 12.7 mm. With two collimator blocks attached to the top shield plate, the total collimator length is 38.1 mm. Each collimator block, as well as the top shield plate, has a 10mm square opening which matches the dimensions of the CZT crystal below. This effectively positions the detector to "see" only the uranium sample which is placed over the collimator window. The shielded enclosure reduces background counts to an average of less than one count per second, compared to approximately 4 cps for the unshielded, uncollimated detector.

Figure 2: Shield and Collimator Assembly for Enrichment Measurements

Measurement Setup

Measurements of a number of different enrichment levels of uranium were made at the Y-12 Nuclear Detection and Sensor Testing Center Site 2, using the site's uranium enrichment standards. These standards were designed with enrichment levels that bracket important enrichment thresholds to assess the accuracy of new measurement systems. The uranium enrichment standards are formed into 3 cm discs with 3 mm thickness (an "infinite" thickness in terms of the detector). Four of the standards are shown below in Figure 3. The Y-12 NDSTC provided 11 standards for measurement for this test, with the assayed enrichment levels (using Thermal Ionization Mass Spectrometry, or TIMS) shown in Table 1:

Table 1: TIMS Enrichment Standard Assay Results

Enrichment (percent ^{235}U)
0.189
0.712
3.022
3.513
4.735
19.365
20.762
36.198
54.861
70.485
93.160

Figure 3: Uranium Enrichment Standards (photo courtesy of Y-12 National Security Complex)

The uranium discs were placed to completely cover the collimator window, and three 300 second spectra were collected for each sample. The 900 second spectra were created by combining the 300 second spectra. A 600 second background measurement was taken each day, however backgrounds did not vary significantly from one day to another. During the initial measurements on bare discs, the background spectra contained about 580 counts over the 600 seconds. During the subsequent studies with stainless steel attenuators between the disc and detector, each background spectrum had about 400 counts over 600 seconds, due to a change in the measurement location.

Detector Calibration

Detector calibration spectra were acquired each day during the course of the measurements with ^{57}Co and ^{137}Cs calibration sources. Typical calibration spectra collected during our measurements are shown below. These spectra were used during data analysis for energy calibration for the HYPERMET algorithm to identify the presence and energy of the ^{235}U peaks. The detector did not show any significant energy drift during the course of the measurements. The calibration spectra shown in Figure 4 are characteristic of the good quality spectra obtained with the detector system.

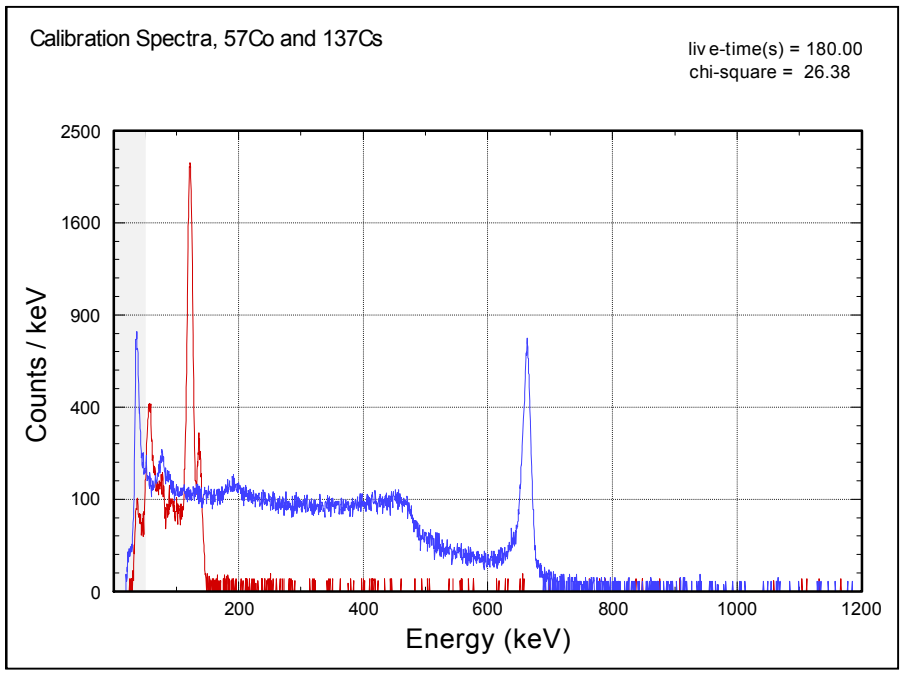

Figure 4: Calibration Spectra Collected During Y-12 Measurements

Uranium Measurements

Measurements on the uranium standards were carried out over several days. A total of three 300 second spectra were collected for each standard, resulting in 900 s of data for each one. Analysis was first performed on the 900s data; the 300s spectra were analyzed separately to understand the effects of shorter

collection times. The collected spectra clearly show the three prominent ^{235}U peaks at 144, 163, 186, and 205 keV. The spectrum for the 93.2% enriched sample is shown in Figure 5.

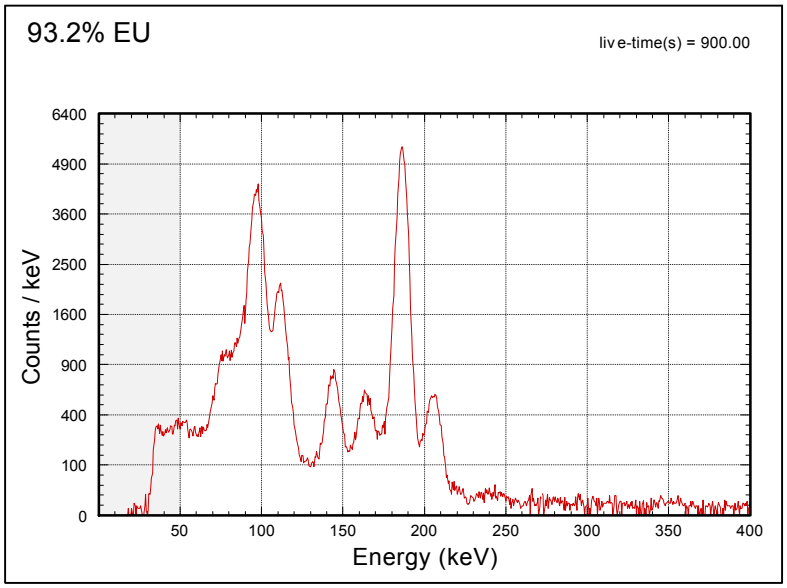

Figure 5: Energy Spectrum of 93.2% Enriched Uranium Sample

The spectra collected for each sample were analyzed using the HYPERMET algorithm to calculate peak area for the 186 keV ^{235}U peak, dividing the observed spectrum into adjoining peaks and background to get the most accurate estimate of peak area. The linear relationship between the 186 keV peak area and enrichment level lends itself well to an automated enrichment calculation based on a collected spectrum, using the enrichment meter principle. Shown on a graph in Figure 6, the peak areas display a linear relationship with actual enrichment values.

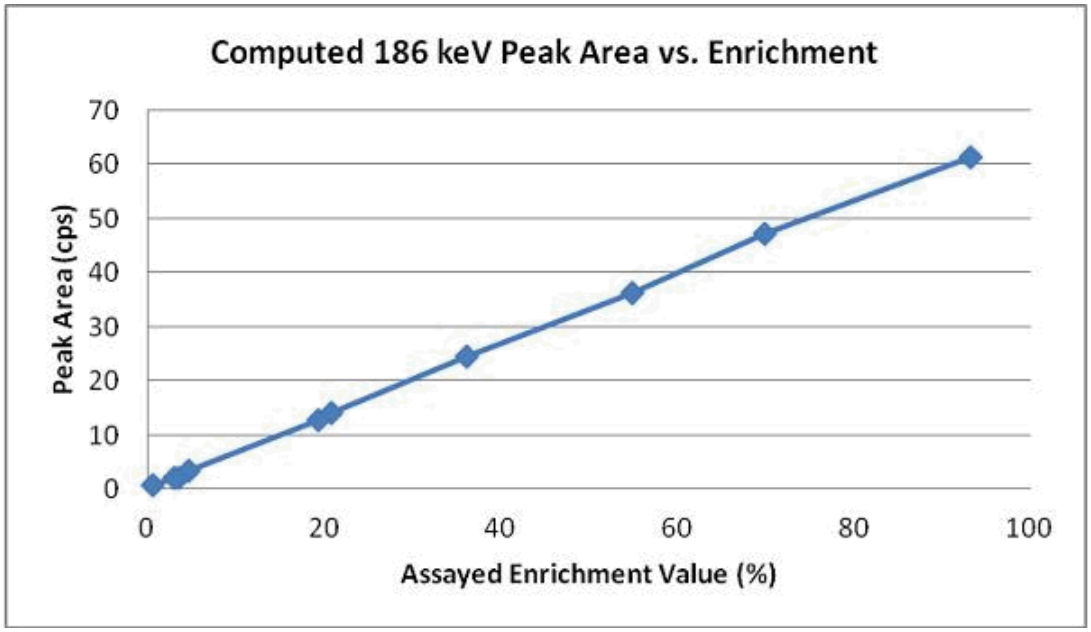

Figure 6: Peak areas (counts per second) vs. actual enrichment.

Figure 7 is a closer look at the lower enrichment samples, from natural to LEU levels of enrichment. The linear relationship of the peak area to enrichment value is continued in this region.

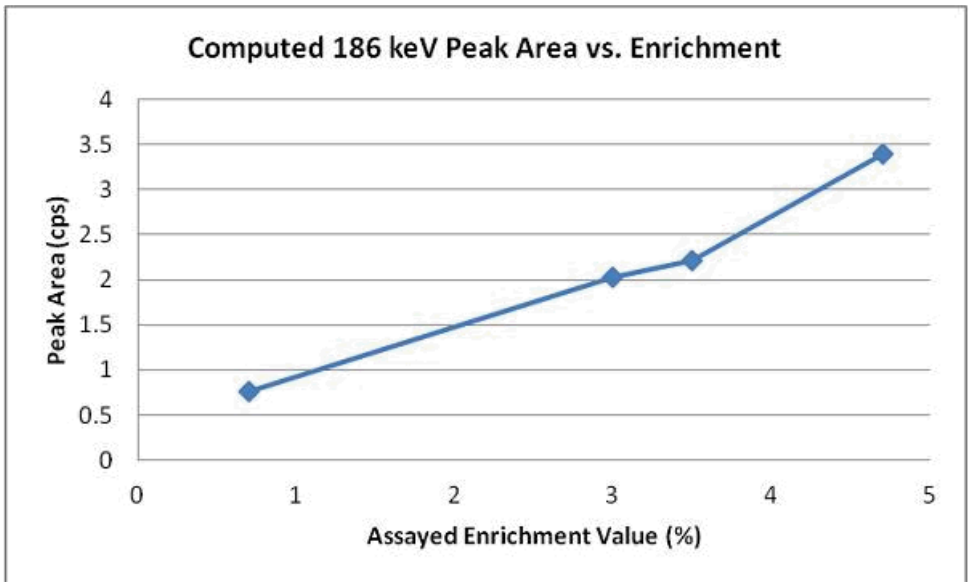

Figure 7: Peak area of low enrichment samples

A Windows™ application was written which calculates enrichment from an acquired spectrum using the HYPERMET algorithm. The application uses calibration spectra to first perform an energy calibration, after which spectral peaks are identified and the areas calculated. The 93% enrichment was used as the reference, and the corresponding lower enrichment values were computed using the ratio of the peak areas. Table 2 displays results from 900 second collection times.

Table 2: Peak ratios and calculated enrichment values

^{235}U 186 keV Peak Ratios [reference = ^{235}U 93.2%]				
Assayed Enrichment (%)	Peak Area (cps)	Ratio	Computed Enrichment (%)	Deviation
^{235}U 93.2	61.362	1	93.2	0.0%
^{235}U 70.5	47.128	0.768	71.6	1.6%
^{235}U 54.9	36.191	0.59	55.0	0.1%
^{235}U 36.2	24.426	0.398	37.1	2.5%
^{235}U 20.8	14.018	0.228	21.2	1.9%
^{235}U 19.4	12.769	0.208	19.4	0.0%
^{235}U 4.7	3.391	0.055	5.1	8.5%
^{235}U 3.5	2.209	0.036	3.4	-2.9%
^{235}U 3.0	2.028	0.033	3.1	3.3%
^{235}U 0.7 (natural)	0.760	0.012	1.2	71%
^{235}U 0.2 (depleted)	No peak found	N/A	N/A	N/A

Relative deviation increases with the lower enrichment values, as fewer counts are found in the 186 keV peak and the peak area calculation becomes more sensitive to counting statistics.

Below in Table 3 are shown results for the 300 second spectra. These results indicate that a shorter count time can still be used to compute enrichment values, although measurement errors are increased compared to the 900 second data. In one case for the 4.7% enrichment, the automated fitting algorithm resulted in a poor fit to the peak; we are still investigating the cause. A larger detector would likely be necessary to decrease count times below 300 seconds and still provide useful results.

Table 3: Enrichment values from 300 sec spectra

Assayed Enrichment (%)	Spectrum 1 Result (%)	Spectrum 2 Result (%)	Spectrum 3 Result (%)	Average Deviation
0.7 (natural)	0.7	1.1	0.8	24%
3.0	2.8	3.6	2.6	13%
3.5	3.7	4	3.2	9.5%
4.7	4.6	bad fit	4.4	4.3%
19.4	19.2	19.1	19.4	0.9%
20.8	23.9	20.7	20.8	5.1%
36.2	37.7	39	38.6	6.2%
54.9	54.7	54.1	56.2	1.4%
70.5	70.9	72.4	71.3	2.1%
93.2	94.0	105.9	91.5	5.4%

Shielding Studies

Stainless steel attenuators with thicknesses of 0.76, 1.85, 2.97, and 4.83 millimeters were tested to understand the effects of shielding on the computed peak areas and resulting enrichment determination. Shields were placed between the uranium sample and the collimator assembly for each test, and data again taken in 300 second intervals for a total of 900 seconds of count time for each enrichment sample and shielding thickness. We had originally hoped that by using the differential attenuation of the nearby uranium peaks (144, 163, and 205 keV), we could infer the shielding thickness and thus automatically compensate for an arbitrary shield. We found, however, that the peak areas of the nearby uranium peaks were too small, and subject to too much statistical variation, to make accurate comparisons of attenuation.

However, the shielding did change the 186 keV peak area in a predictable way, and enrichment measurements with shielding present can be made by calibrating the system for a specific shielding thickness (i.e. for a specific packaging configuration). Following the method for attenuation correction in Decman (1999)[v], the following table displays results from the measurements of the Y-12 standards with shielding. The attenuation correction factors for each of the shielding thicknesses are, respectively: 1.09, 1.24, 1.41, and 1.74, using the equation $ATT_{CORR} = e^{-\mu t}$, where μ is the attenuation coefficient (in this case 0.115 cm^{-1}, as used in Decman, 1999) and t is the material thickness The table shows both the enrichment result, and the corresponding deviation from the actual assayed enrichment. The error, which increases with thicker shielding and higher enrichment values, was also described in Decman (1999) and is likely due to low angle scattering in the stainless steel, however additional measurements are necessary to confirm this.

Table 4: Corrected Enrichment Values for Shielding Measurements for 900 sec collections

Assayed Enrichmt (%)	Calculated Enrichment with Attenuation Correction (%), and Deviation									
	No Attenuator		0.76mm		1.85mm		2.97mm		4.83mm	
	Result	Dev.	Result	Dev.	Result	Dev.	Result	Dev.	Result	Dev.
0.7	1.2	71%	0.87	24%	0.74	5.7%	0.79	12%	1.2	71%
3.0	3.1	3.3%	2.9	-3.3%	3.0	0.0%	3.1	3.3%	3.7	23%
3.5	3.4	-2.9%	3.4	-2.9%	3.3	-5.7%	3.2	-8.6%	3.5	0%
4.7	5.1	8.5%	4.6	-2.1%	4.3	-8.5%	4.4	-6.4%	4.7	0%
19.4	19.4	0.0%	18.9	-2.6%	20.4	5.2%	20.4	5.2%	22.6	16%
20.8	21.2	1.9%	20.6	-1.0%	21	1.0%	20.7	0.5%	22.4	7.8%
36.2	37.1	2.5%	37.5	3.6%	38.1	5.2%	38.4	6.1%	41.2	14%
54.9	55	0.1%	53.6	-2.4%	55.1	0.4%	55.1	0.4%	63.2	15%
70.5	71.6	1.6%	70.4	-0.1%	71.6	1.6%	71.8	1.8%	80.9	15%
93.2	93.2	0.0%	98.9	6.1%	95.7	2.7%	109	17%	108	16%

Conclusion

We have described a method for uranium enrichment determination utilizing a commercial CZT detector system and a non-proprietary analysis algorithm. Using a collimator assembly similar to those developed previously for in-process enrichment monitoring, we made measurements on enriched uranium standards available at the Y-12 National Security Complex. Automated software analysis of the resulting spectra indicates that accurate enrichment measurements can be made in the 300 second to 900 second timeframe. This type of detector system could be made into a portable, battery operated system for in-field measurements useful for potential future arms control agreements. A semi-custom redesign of the CZT detector module would enable the use of a much smaller and lighter collimator and shield assembly. Using commercial detectors and electronics, with non-proprietary software, this type of system has the necessary transparency for joint or multi-lateral monitoring agreements where system certification and authentication is critical.

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[ii] R. Gunnink and R. Arlt, "Methods for evaluating and analyzing CdTe and CdZnTe spectra", *Nuc. Instr. Meth.* A **458,** 196-205 (2001).

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[iv] Decman, D.J., Bandong, B.B., Wong, J., Valentine, J., Luke, S.J., "Portable NDA Equipment for Enrichment Measurements in the HEU Transparency Program", *Proceedings of the 2006 INMM Annual Meeting.*

[v] Decman, D.J., Glaser, J., Hernandez, J.M., and Luke, S.J., "Portable NDA Equipment for Enrichment Measurements for the HEU Transparency Program", *Proceedings of the 1999 INMM Annual Meeting.*