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Uranium Isotopic Analysis with Commercial CZT Detectors and a Generic Algorithm

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Abstract
Sandia National Laboratories, in partnership with the Y-12 National Security Complex, 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 several 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 measurements were made using commercially available CZT detectors with the goal of identifying an appropriate algorithm for measuring enrichment levels. Recent improvements in CZT detector systems, both in crystal size and resolution, may make these detectors practical in this application and point the way towards more portable and affordable equipment for 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 (Phillips and Marlow, 1976), 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 find doublets and triplets which may otherwise be hidden in the data, and was used to find overlapping peaks in the uranium spectrum. Significant areas of interest in the spectrum were analyzed for peaks that could be used to find enrichment levels. Results from this analysis are presented, as well as future potential for applying this algorithm to arms control monitoring applications or other applications such as international safeguards. This work was sponsored by the National Nuclear Security Administration Office of Nuclear Verification.

Introduction
Government and industry research into detector materials has advanced the state of the art in passive detection in the past few years. Detector materials such as CZT (Cadmium-Zinc-Telluride) provide good resolution (1-3%) at room temperature, enabling isotopic analysis based on resolving spectral peaks. However, commercial algorithms are typically based on finding and identifying single peaks. While single peak detection methods work fairly well for the 0.1% resolution High-Purity Germanium systems, it begins to fail when poorer resolution spectra starts producing doublets and triplets from actinides. For Sodium Iodide systems at 7% resolution, single peak detection algorithms provide poor performance and full spectral analysis with template matching becomes necessary. To date, there has been very little research into alternate identification algorithms for the mid-range resolution spectra from materials such as CZT.

Sandia National Laboratories has used a previously published technique called HYPERMET (Philips and Marlow, Naval Research Laboratory, 1976) to build an open source analysis code for determining the presence of HEU and also estimating 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. Past research at Sandia indicates that this approach should be highly effective with spectra produced by 1-3% resolution detectors such as CZT.

Two commercial CZT detectors were used to obtain enriched Uranium test data. The first instrument, the ***iGEM***™ from Endicott Interconnect Technologies, Inc. is an OEM spectrometer "engine" which can be incorporated 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 detector utilizes a 10x10x10 mm (1 cm^3) CZT Coplanar Grid Detector and records spectra in 4096 channel files. The second instrument, the Raider™ from FLIR (formerly iCX Radiation), is packaged as a handheld detection/inspection tool and has a self-contained display screen and controls. The detector is also a 1 cm^3 CZT crystal and outputs spectra in 2048 channel files. Characterization measurements on both detectors show that resolution at 662 keV is approximately 2.1% FWHM. The ***iGEM***™ shows better efficiency at higher energies than the Raider™, although neither instrument displays useful spectral data above 1500 keV. Spectra from both instruments were used to adapt the HYPERMET algorithm to CZT spectra, and were subsequently analyzed to determine the instruments' effectiveness in determining uranium enrichment.

Experiments

Experiments were completed at the Nuclear Detection and Sensor Testing Center (NDSTC), located at the Y-12 National Security Complex in Oak Ridge, Tennessee. The NDSTC provides researchers the ability to test their equipment or analysis techniques using special nuclear material (SNM). The enriched uranium metal standards provided for experimentation are approximately three centimeters in diameter and 3 millimeters thick. The four uranium metal standards have enrichments of 3, 19.4, 54.9 and 93.2 percent (Figure 1).

Figure 1: Uranium Enrichment Standards (photo courtesy of Y-12 Natl Security Complex)

Spectral data was gathered using two detectors, the Raider™ and the ***iGEM***™. Each source was placed at a distance of 10 cm during the measurement process as shown in Figure 2. Radiation spectra were collected for no less than two and no more than five minutes. The collected spectra were then saved for analysis. In addition, several more measurements at long count times were made using uranium standards located at Sandia National Laboratories in order to better characterize the detectors.

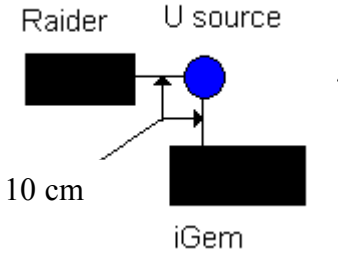

Figure 2: Experimental Setup

Algorithm

The original computer program HYPERMET was developed at the Naval Research Laboratory for "automatic and efficient analysis of multichannel pulse height spectra from high-resolution germanium gamma-ray detectors."[i] HYPERMET was a great improvement over previous codes because of its ability to automatically identify peaks in gamma-ray spectra that—although discernable 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.

A modern implementation of HYPERMET was developed in C++. Our initial algorithm can be described by the following four 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. As we will see below, HYPER-ID has no problem identifying isotopes whose gamma-ray peaks may be so close as to completely overlap. Steps 1-3 in the above list are all described in the original algorithm by Phillips and Marlow.

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 is found to the data (i.e. the fit has a width of approximately 1-3 keV and low chi-squared value) 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.

Data

Sample spectra from the two commercial detectors are shown below in Figure 3. Both show excellent resolution of the characteristic 186 keV peak from ^{235}U, as well as several other uranium peaks at 144 keV, 163 keV, and 205 keV. Numerous ^{238}U, ^{235}U, and X-ray peaks in the 90-110 keV region are visible but unresolvable into separate peaks. While HPGe spectra can be analyzed in this latter region to obtain Uranium enrichment levels, the resolution of CZT detectors is not yet sufficient to perform a meaningful analysis of this region, even with the HYPERMET algorithm. However, the well-defined 186 keV peak from the two CZT detectors should present an improvement over lower resolution sodium-iodide based detectors in the calculation of enrichment ratios.

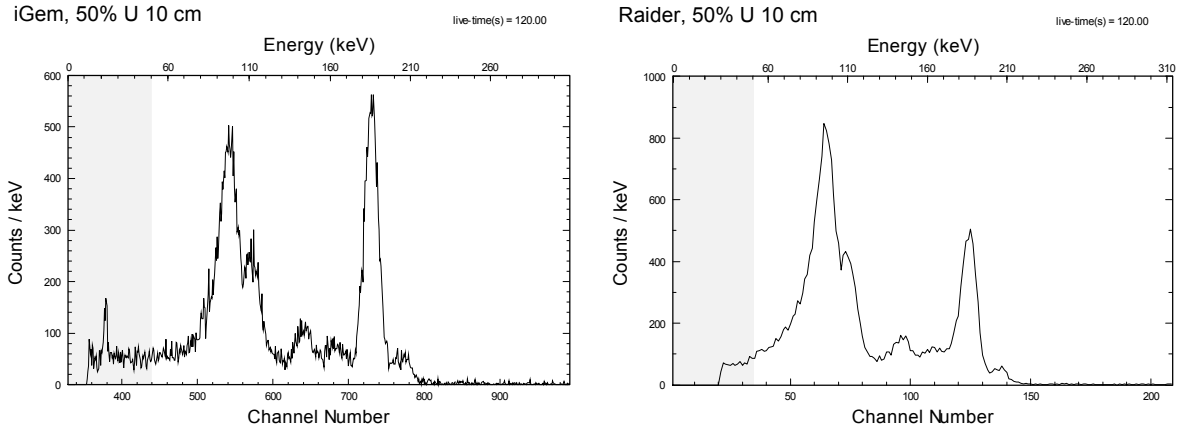

Figure 3: 50% enriched uranium spectrum from ***iGEM***™ detector (left) and Raider™ (right).

Data Analysis

In each test case, the HYPERMET algorithm identified the peaks associated with ^{235}U and therefore signaled the presence of enriched uranium with high confidence. The higher resolution of the CZT detectors made positive identification of ^{235}U relatively straightforward due to the multiple identified peaks.

For determination of enrichment, peak area is plotted against actual enrichment values. As a reference, when peak areas are plotted against enrichment for High-Purity Germanium detectors, the result is linear; therefore, enrichment can be extrapolated directly from the area of the 186 keV peak. In the case of the two commercial CZT detectors under test, Figure 4 shows that both return results which show increasing peak area with enrichment, as would be expected. However, the Raider shows better linearity over the range of enrichments in our test. More extensive testing with a wider range of enrichment standards should provide a confirmation of these results and will aid in the refinement of the algorithm for computing enrichment values. These results

show that commercial CZT detectors have potential as deployable tools for enrichment determination.

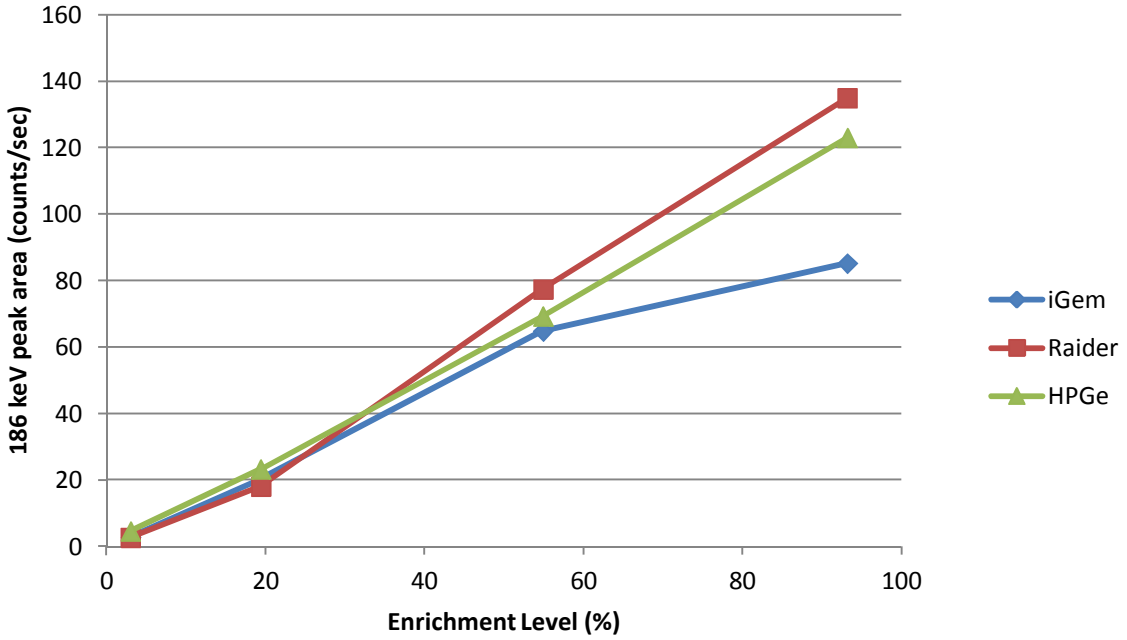

Figure 4: 186 keV peak area plotted against actual enrichment level for the two CZT detectors. HPGe data is plotted for reference and has been normalized.

Conclusion

A method of detecting Uranium enrichment levels was investigated utilizing two new commercial CZT-based radiation spectrometers in conjunction with a previously developed peak analysis algorithm called HYPERMET. Using HYPERMET to analyze data taken with uranium enrichment standards provided by the Y-12 site, good correlation was found between the computed and actual enrichment levels based on the area of the 186 keV ^{235}U peak. The affordability of new CZT-based detection systems, combined with the accuracy of the openly published HYPERMET algorithm, could potentially result in the development of new, more economical systems for the measurement of uranium enrichment in arms control applications. Additional work to fully develop a software package and measurement procedure, including a method of calibration, must be completed to create a usable system; however these initial results show promise for further development.

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[i] Gary Phillips and Keith Marlow, "Automatic Analysis of Gamma-ray Spectra from Germanium Detectors", *Nuc. Instr. Meth.* **137,** 525-536 (1976).

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

[iii] P. Mortreau and R. Berndt, "Determination of ^{235}U enrichment with a large volume CZT detector", *Nuc. Instr. Meth.* A **556**, 219-227 (2006).