Cadmium Zinc Telluride Detector System for Nuclear Material Assay

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CADMIUM ZINC TELLURIDE DETECTOR SYSTEM FOR NUCLEAR MATERIAL ASSAY

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Abstract
This paper will report on the results of over three years of collaborative efforts between Lawrence Livermore National Laboratory (LLNL) and EG&G ORTEC to develop an ambient temperature radiometric instrument. Three tools were developed as a result of this research: The CZT Probe - a cadmium zinc telluride (CZT) -based gamma- and x-ray detector probe, the MicroNOMAD - a low power, portable multichannel analyzer (MCA), and CZTUS - spectral analysis software that provides uranium enrichment analysis. The combination of these three tools with an optional sodium iodide (NaI) detector provides the ability to search for and then analyze uranium as well as other radionuclides in the field. Several national and international organizations including the International Atomic Energy Agency (IAEA), the European Communities Safeguards Directorate (EURATOM), U.S. Customs, and U.S. Department of Energy (DOE), have expressed interest and are currently evaluating these systems. We will report on improvements in peak efficiency presently being made.

INTRODUCTION
BACKGROUND
The primary goal of the project was to develop a portable, low power ambient temperature radiation detection instrument that took advantage of new developments in semiconductor detector technology. The primary design criteria was driven by two significant industrial needs; environmental radiation detection such as 'lead in paint' determination by x-ray fluorescence and gamma-ray measurement to provide uranium enrichment determination in the field. It was recognized that both of these applications would require resolution performance significantly better than sodium iodide (NaI) systems (~25% at 122 keV), but would not require the high resolution performance of high purity germanium (HPGe) systems (<0.5% at 122 keV).

The development of this system began in 1992 with a survey of available semiconductor detector technologies. It became quickly evident that cadmium telluride (CdTe) and cadmium zinc telluride (CZT) were the only ambient temperature radiation detection materials that were truly available and could potentially provide the resolution performance necessary for uranium enrichment analysis. Initial evaluations of current isotopic analysis codes (MGAU, GRPANL[1]) indicated that resolutions of <3% at 122 keV ($^{57}$Co) should be sufficient to obtain reasonable analysis results.

DETECTOR EVALUATIONS
Given the preliminary resolution requirements, sample CZT and CdTe detectors were evaluated with both $^{57}$Co and NIST standard uranium sources. The test configuration consisted of two detectors, one each of CZT (20% zinc) and CdTe, in BNC mountings, an ORTEC 142PC preamplifier, and an ORTEC 572 shaping amplifier (1 μs shaping). The resolution of both detectors exceeded the 3% requirement in addition to being extremely stable with respect to
temperature, and no peak shift was noted during long (> 100 hours) acquisitions. Though the performance was encouraging, several undesirable characteristics were observed.

The first characteristic was low efficiency. Due to the small volume detectors used (2x2x2 mm, planar geometry), counting times for the sources were at times excessive. In uranium samples of very low or very high enrichment, the spectral lines from the low abundance isotope can be lost in the background. The result was that extremely long acquisition times were necessary to be able to differentiate the low abundance peak from the background. These results can be seen in Table 1. The size limitation for spectroscopic grade detector material seems to be primarily due to material resistivity, with CdTe at approximately $10^9 \, \Omega \cdot cm$ [3,4] while CZT is typically about $10^{11} \, \Omega \cdot cm$ (depending on zinc content)[5]. Lower resistivity results in less effective charge collection as well as higher detector leakage currents and corresponding shot noise in the detector signal. CdTe detectors appear to have a thickness limit of approximately 2-3 mm, after which the leakage current and respective shot noise becomes significant. CZT detectors can be fabricated to much larger volumes due to a higher resistivity, which also results in higher efficiency.

The second limiting characteristic was a significant low energy tail, apparent in both CZT and CdTe spectra. Sample $^{57}$Co spectra are shown in Figures 1 and 2, respectively. One problem with this low energy tailing effect was reduced accuracy of the uranium enrichment analysis results because existing code was based on HPGe spectra with characteristic Gaussian peak shapes. Even with the distorted peak shapes of CdTe and CZT, long acquisition times and the use of HPGe-based uranium enrichment analysis code, the initial results as shown in Table 1 were encouraging.

**Table 1**

<table>
<thead>
<tr>
<th>Declared $^{235}$U/$^{238}$U</th>
<th>Measured $^{235}$U/$^{238}$U</th>
<th>RSD* (%)</th>
<th>Measurement Time (hours)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0314</td>
<td>0.0302</td>
<td>16.8%</td>
<td>12</td>
</tr>
<tr>
<td>0.0528</td>
<td>0.0557</td>
<td>12.5%</td>
<td>6</td>
</tr>
<tr>
<td>0.1136</td>
<td>0.1116</td>
<td>4.6%</td>
<td>6</td>
</tr>
<tr>
<td>0.9997</td>
<td>1.0093</td>
<td>13.7%</td>
<td>15</td>
</tr>
<tr>
<td>3.166</td>
<td>4.42</td>
<td>36.3%</td>
<td>15</td>
</tr>
</tbody>
</table>

*R*Relative Standard Deviations (RSD) were determined from the fits to the net counts in the spectra.

A related difficulty arose in performing reliable resolution measurements. There were several methods being employed to report Full Width at Half Maximum (FWHM) resolution specifications by various CZT and CdTe manufacturers that did not always reflect the actual performance of the detectors. To provide consistency in all experiments and allow for a reasonable comparison of detectors, a standard method for determining the resolution (FWHM) of CZT and CdTe detectors was developed. This method was reported earlier [2] and is quite simple. The peak amplitude of the energy line of interest is divided by two with the width of the pulse height distribution at that level reported as the FWHM. This method of determining resolution includes the effects of low energy tailing and removes the arbitrary placement of the low energy set point.
A significant enabling development was the introduction of larger volume (≈125 mm$^3$), moderate resolution CZT detector crystals. While this volume (and resulting efficiency) is relatively small in comparison to typical HPGe and NaI detectors, CZT detectors operate at ambient temperature eliminating the requirement for liquid nitrogen (LN) cooling and can provide spectra with resolution adequate to perform the isotopic analysis of nuclear materials.

**SYSTEM DEVELOPMENT**

**HARDWARE** The system developed is a completely portable instrument and is shown in Figure 3. It consists of a laptop computer that contained both the system operation and uranium enrichment analysis software, a portable multichannel analyzer (MCA), an optional NaI detector used for search mode operation (not shown), and a CZT detector probe used for verification and analysis mode operation. EG&G ORTEC was responsible for the development of the portable MCA including operational software and LLNL developed the CZT detector probe and uranium enrichment analysis software.

**MULTICHANNEL ANALYZER**

The MCA developed by EG&G ORTEC for this application is the MicroNOMAD. It is a 1.5-lb., battery powered, portable gamma-ray spectrometer consisting of an amplifier suitable for NaI operation (bypassed for CZT operation), 2k ADC, spectrum stabilizer, microprocessor and memories, as well as parallel, RS-232, and RS-485 ports. Both the NaI probe and the CZT probe connect to the MicroNOMAD using a single cable. The unit is small, only 7x7x21 cm, and requires < 1.5 W. Eight AA batteries will operate the unit for about 6 hours. Field-mode operation allows the MicroNOMAD to record multiple spectra at the push of a button or the scan of a bar code. Each acquisition is performed in accordance with the preset values of real time, live time, gross ROI counts, or peak ROI counts. Up to 63 spectra can be stored. At the end of each acquisition, the spectral data, start time, real time, live time, and up to 16 characters read by the bar code can be stored. A computer can be connected to the MicroNOMAD using the built-in parallel port. In-field automatic nuclide identification is easy using analytical software specially designed for NaI detectors. Multipoint energy and efficiency calibration is also provided.
CZT ELECTRONICS
The CZT probe was designed specifically to optimize the performance of CZT detectors. Thorough consideration was given to all the CZT characteristics that contribute to less than ideal energy resolution including baseline noise (primarily shot noise), poor hole-mobility and charge trapping. The preamplifier and filter amplifier were designed to minimize these effects and included monolithic devices, which allowed the current implementation of surface-mount technology for further size reduction. This design achieves excellent reduction of spectral noise. A typical example of a spectrum obtained using a $^{57}$Co source and a 5x5x5 mm CZT detector is shown in Figure 4. The energy resolution of the 122 keV peak is 3.32 keV FWHM, with resolutions approaching 2.5 keV FWHM when using the highest quality spectrometry-grade detectors. This monolithic design was able to achieve excellent resolution (10% better than commercial instrumentation) and low power consumption (600 mW). A detailed description of the development of the CZT probe can be found in Reference [6].

![CZT Detector System](Image)

**MicroNOMAD**

**CZT Probe**

**Fig. 3 Portable CZT Radiation Detection System**

URANIUM ENRICHMENT ANALYSIS
The standard analysis codes used for uranium enrichment are based on almost perfectly Gaussian germanium detector peak shapes. The low energy tailing characteristics of CdTe and CZT detector peak shapes required characterization to be performed and the analysis code modified to provide accurate uranium enrichment results. The goal was to provide uranium enrichment determination in the field to an accuracy of within 10% of the known value. Current germanium-based codes with little or no modification provided enrichment results to within 20% of the known value[7].
Several 5×5×5 mm CZT detector peak shapes were analyzed[8]. The results were then used to modify existing analysis codes to provide better peak fit analysis. The performance was moderately improved with results to within 15%, though there were occasional spectra that could not be analyze or resulted in exceedingly large errors. Due to the occasional errors during analysis, a new uranium enrichment analysis code was developed (CZTU). This code was specifically designed to work with CZT-based spectra and provide a more stable analysis. This code was then beta-tested during a radiation detection exercise and the results are shown in Table 2. The CZT detector used was a 5×5×5 mm planar geometry device mounted internal to a prototype CZT probe. The germanium detector was a liquid nitrogen cooled, 1000-mm² LEPS detector. Sample X and Sample Y were fresh fuel; therefore, the $^{238}$U/$^{234}$Th non-equilibrium was corrected for during the analysis. A brief examination of the data shows that the acquisition times for the CZT detector were roughly twice that of the germanium detector. Additionally, with a few marginal exceptions the results tracked the germanium detector and were within the design specifications.

### Table 2
Uranium Enrichment Measurement Exercise Results

<table>
<thead>
<tr>
<th>Sample Number</th>
<th>U-235/U Certified (weight %)</th>
<th>HPGe (weight %)</th>
<th>Acquisition Time (min)</th>
<th>CZT (weight %)</th>
<th>Acquisition Time (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1613</td>
<td>3.1094 ± 0.0044</td>
<td>3.10 ± 0.02</td>
<td>75.5</td>
<td>3.10 ± 0.34</td>
<td>133.4</td>
</tr>
<tr>
<td>1614</td>
<td>1.4972 ± 0.0019</td>
<td>---</td>
<td>---</td>
<td>1.73 ± 0.20</td>
<td>166.6</td>
</tr>
<tr>
<td>1541</td>
<td>1.9952 ± 0.0037</td>
<td>1.99 ± 0.01</td>
<td>61.9</td>
<td>2.01 ± 0.23</td>
<td>161.4</td>
</tr>
<tr>
<td>1542</td>
<td>2.8774 ± 0.0040</td>
<td>2.87 ± 0.02</td>
<td>92.2</td>
<td>2.84 ± 0.32</td>
<td>145.9</td>
</tr>
<tr>
<td>Sample X</td>
<td>3.4317 ± 0.0032</td>
<td>3.3 ± 0.03</td>
<td>75.4</td>
<td>3.03 ± 0.34</td>
<td>158.8</td>
</tr>
<tr>
<td>Sample Y</td>
<td>2.6846 ± 0.0031</td>
<td>2.55 ± 0.02</td>
<td>67.8</td>
<td>2.42 ± 0.28</td>
<td>179.7</td>
</tr>
<tr>
<td>125V</td>
<td>93.1556 ± 0.0227</td>
<td>89.39 ± 4.87</td>
<td>90.9</td>
<td>---</td>
<td>---</td>
</tr>
</tbody>
</table>

**PEAK EFFICIENCY IMPROVEMENTS**

Digirad, a CZT manufacturer located in San Diego, California that specializes in medical imaging, has developed a new detector technology that provides dramatic improvement to the peak-to-valley spectral characteristics of CZT detectors. Typical detectors (5×5×5, 4×4×4, and 3×3×3 mm) operated at
room temperature have a peak-to-valley of up to 7 to 1 at $^{137}\text{Cs}$ (662 keV) with low energy tailing being the limiting factor. The new detectors from Digirad provide a peak-to-valley of up to 60 to 1. This dramatic increase is due to the virtual elimination of low energy tailing as a result of unique and proprietary single charge collection techniques.

CONCLUSIONS
LLNL and EG&G ORTEC have developed a truly portable, ambient-temperature, radiation detection instrument that provides for uranium enrichment determination in the field. There is still considerable room for improvement, specifically in the implementation of larger volume detectors when available and the improvement of the enrichment analysis precision.

References

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