Development of a Portable Ambient Temperature Radiometric Assaying Instrument

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Development of a Portable Ambient Temperature Radiometric Assaying Instrument

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Abstract

There is a strong need for portable radiometric instrumentation that can accurately confirm the presence of nuclear materials and allow isotopic analysis of radionuclides in the field. To fulfill this need we are developing a hand-held, non-cryogenic, low-power gamma- and X-ray measurement and analysis instrument that can both search and then accurately verify the presence of nuclear materials. We will report on the use of cadmium zinc telluride detectors, signal processing electronics, and the new field-portable instrument based on the radionuclides in the field. To fulfill this need we are developing a hand-held, noncryogenic, low-power gamma-ray fluorescence analysis of heavy metals. The MicroNOMAD Multichannel Analyzer from EG&G ORTEC. We will also describe the isotopic analysis that allows uranium enrichment measurements to be made accurately in the field.

I. INTRODUCTION

This paper describes a truly field-portable instrument capable of search and verification of nuclear materials. The benefits of this work are realized in a wide spectrum of applications that include Arms Control, Nuclear Safeguards, Environmental Management, Emergency Response, and Treaty Verification. In addition, we characterize our instrument for use as an environmental screening tool by X-ray fluorescence analysis of heavy metals.

Instrumentation for high-resolution spectrometry that is presently available for the applications described above is physically large, requires high power, and incorporates a liquid-nitrogen cooled detector assembly; these aspects limit its use in field applications and covert operations. Therefore, this work focuses on the use of an ambient temperature detector, low-power electronics, and hand-held operation.

II. DETECTORS

A. Detector Selection

There are currently no ambient temperature detectors known that provide both the high efficiency required to conduct a reasonable search, and the resolution necessary for isotopic analysis. Developing an ambient temperature system to satisfy this need for analyzing radionuclides has been a challenge for a number of years. Most of this type of analysis work has been in conjunction with liquid nitrogen (LN) cooled germanium (Ge) detector systems in laboratories. Obviously, the logistics necessary to support a cooled Ge system has always been a problem outside the laboratory. Since our instrument is intended to provide both a search mode and an isotopic analysis mode of operation, a dual-detector system approach was adopted.

The first detector needed was one capable of high sensitivity searches. Sodium iodide (NaI) detectors have been used in this capacity for some time. These detectors are available in almost any size and configuration and will provide high enough resolution to determine the presence of radionuclides of possible interest. A system of this type is currently available from ORTEC (ScintiPack 296 and a NaI detector) and will be integrated with the MicroNOMAD to fulfill the needs of the search mode detector.

The detector to be selected for the confirmation, or verification technique, of operation, had to meet several requirements. The most critical of the requirements are moderate resolution and stable temperature characteristics. The resolution requirement was determined to be <3% on 57Co (122 keV). Three detector technologies met these requirements though only two were available in quantity; cadmium telluride (CdTe), and cadmium zinc telluride (CZT). The third technology, mercuric iodide, is not yet commercially available.

Due to the use of a modified High Pressure Bridgeman (HPB) crystal growth technique[1], very large CZT boules of excellent homogeneity can be grown. The addition of zinc to CdTe also has the benefits of both increasing the resistivity of the material several orders of magnitude and raising the bandgap from 1.47 to 1.6 eV. The higher resistivity allows for larger detectors and the requisite higher bias voltages necessary for improved charge collection. The higher bandgap reduces the bulk generated leakage current providing better ambient temperature operation. It was for these reasons that CZT was chosen for the isotopic analysis detector.

B. Measurement Techniques

It is important to discuss the method that is used to measure the Full Width at Half Maximum (FWHM)
resolution. At this point in time, there exists several methods for determining FWHM, each resulting in different performance figures for equivalent devices. In an effort to remain consistent, the FWHM data presented in this paper will be calculated using absolute counts from the baseline. The peak amplitude of the energy line of interest will be divided by two, and the width of the pulse height distribution at that level will be used. This method of determining the FWHM takes into account the effects of low energy tailing from charge collection problem associated with the detector. Another paper[3], written by the authors and also presented at this conference, addresses this issue.

C. CZT Detector Characteristics

The first obstacle to overcome was the design of a low power, low noise, front end package that would optimize the performance of CZT detectors without the use of elaborate signal processing techniques. These techniques [e.g., Pulse Shape Discrimination (PSD), Charge Loss Correction, and Time-Variant Shaping] have all been applied successfully in various systems in the past, but they also bring with them additional power requirements, loss of counting efficiency, and other problems which prevent meeting our application criteria.

Many detectors of differing geometries were evaluated for charge transport problems -- such as slow components in the charge collection process.

The risetime data from a 2x2x2 mm CZT detector is shown in Fig. 1 and 2. A single charge collection waveform is shown in Fig. 1. This is a typical CZT charge pulse showing a fast risetime with characteristic slow charge collection at the top of the pulse. Fig. 2 is an average of 1000 charge collection risetime waveforms. These signals were taken from the output of a modified ORTEC 142PC charge sensitive preamp.

A pulse risetime distribution for this material shows a small percentage of slow charge collection components. The high resistivity of CZT (≈10^{11} Ω-cm) allows for a higher bias voltage and the subsequent higher electric field, which results in better charge collection.

This provides only a small overall percentage of slow charge collection components which, in turn, results in the poor response of CZT detectors to PSD techniques. The effect of PSD is to examine the risetime of each pulse and reject any that do not conform to a minimum risetime threshold. As can be seen in Fig. 1, this technique would be of negligible benefit. In practice, the application of PSD actually degrades the system performance. PSD typically resulted in the rejection of 60 to over 90% of the events, depending on the risetime threshold setting. Since the benefit of this technique with respect to resolution and low energy tailing for CZT is marginal, the largest observable result was a significant loss of efficiency.

Recent advances in CZT crystal-growth techniques have led to the fabrication of detectors of relatively large volumes exhibiting the required energy resolution with high yields that result in low cost. This has made it practical to pursue development of this field instrument. It is now reasonable to expect a resolution of < 3% on the 122 keV peak of ^{57}Co with a 5x5x5 mm CZT detector. Significantly, this good resolution was achieved on a relatively large volume detector without pulse shape discrimination or other schemes that drastically reduce the photopeak efficiency, as previously mentioned.

III. ELECTRONICS

A. CZT Electronics

Even with improved CZT material, noise (primarily shot noise), poor hole-mobility, and charge trapping contribute to less than ideal energy resolution. Therefore, the preamplifier and filter amplifier have been designed to minimize these effects. All front-end electronics currently use integrated circuit operational amplifiers (IC opamps) and will use surface-mount technology in the final version.

The prototype system that has been designed and tested is shown in Fig. 3. This first prototype design has limited
correction for poor charge collection, but achieves excellent reduction of spectral noise.

An example of a spectrum obtained using a $^{57}$Co source and a 5x5x5 mm CZT detector is shown in Fig. 4. The energy resolution of the 122 keV peak is 3.32 keV FWHM. The electronic contribution (pulser resolution without the detector) is 1.2 keV FWHM using 0.5 μs shaping (1.0 μs peaking). This monolithic design was able to achieve good resolution (10% better than commercial instrumentation) and low power consumption (600 mW). This low power consumption is compatible with the AA batteries used in the MicroNOMAD.

A low noise field-effect transistor (FET) was selected for optimization of series and parallel noise, front end gain, and rise time. Good stability is achieved by running a 400 MHz opamp (IC1) in an open loop configuration with DC feedback for stability. This IC is a critical component that achieves the necessary combination of high bandwidth and low noise. In previous designs, this portion of the preamplifier was usually a hybrid circuit, which is physically larger and has higher power consumption. Power is further reduced in this design by running IC1 on a single ended supply (+12V). The preamplifier sensitivity is 20V/pC and has a rise time of 24 ns.

The amplifier stage is voltage sensitive and is composed of a low noise, high bandwidth opamp preceded by a differentiator and pole-zero compensation network. Coarse and fine gain controls allow adjustment for processing $^{241}$Am (60 keV), $^{57}$Co (122 keV), or $^{137}$Cs (662 keV) full scale. Further power reduction is accomplished by establishing a voltage reference that allows the remaining circuitry to run on the single-ended supply.

A two-stage Sallen-Key filter was designed using opamps IC3 and IC4. This circuit has two pairs of complex poles and was found to be very stable. This circuit also has a sharp noise-corner response, which is very important in reducing parallel noise and at the same time minimizing ballistic deficit due to the slow charge collection of the detector. The shaping time of this filter is 0.5 μs (1.0 μs peaking time). The optimum shaping time for the electronics is 4-6 μs, based on the capacitance of the detector and other circuit parameters. However, detector charge trapping phenomena has a pronounced effect on resolution, and as a result, the filter time constant must be reduced to 0.5 μs. This reduces the amount of low-energy tailing and is based on the characteristics of many CZT detector measurements. These measurements have shown that trapping can add 2-3 keV (in quadrature) to the resolution if the filter time constant is increased to 1 μs.

The second stage of the filter was designed with a higher Q than the first stage. This design achieves improved performance and results in a nearly Gaussian output pulse shape.

B. Portable Multichannel Analyzer

Both the high-efficiency search detector and the high-resolution analysis detector require a multichannel analyzer. The MicroNOMAD is a 1.5-lb., battery powered, portable gamma-ray spectrometer developed especially for this application. The block diagram is shown in Fig. 5. The unit consists of an amplifier suitable for NaI, 2k ADC, spectrum stabilizer, microprocessor and memories, as well as parallel, RS-232, and RS-485 ports. The internal amplifier can be bypassed when using CZT with its optimized amplifier described above. Both the ScintiPack 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. Its 8 AA batteries will operate the unit for over 8 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 63 512-channel spectra can be saved. 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.

Any computer can be connected to the MicroNOMAD using the built-in parallel port. In-field automatic nuclide identification is easy using analytical software specially.

![Fig. 3. Block Diagram of the Prototype CZT System](image-url)

![Fig. 4. Energy Spectrum of $^{57}$Co Using a 5x5x5 mm CZT Detector](image-url)
designed for NaI detectors. Multipoint energy and efficiency calibration is also provided.

IV. APPLICATIONS

A. Uranium Analysis

Isotopic analysis of uranium has been performed using 2x2x2 mm and 5x5x5 mm CdZnTe detectors with samples of 235U enrichments ranging from 3% to 50%. We use a spectrum analysis technique incorporated in GRPANL[2] that fits 235U, 238U, and U K X-Ray response profiles to data in the 89- to 100-keV region of a gamma-ray spectrum. From the relative magnitudes of the 235U and 238U profiles, we determine the 235U enrichment with an accuracy of about 10% with CdZnTe detectors.

The result of using GRPANL is shown in Fig. 6 and 7. The analyzed output using spectra from a high resolution

![Fig. 6 Uranium Spectrum with High Resolution Detector (484 eV at 100 keV)](image1)

![Fig. 7 Uranium Spectrum with CZT Detector (2.9 keV at 100 keV)](image2)
Table 1
GRPANL Measurement Data

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

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

Low Energy Photon Spectrometer (LEPS) detector is shown in Fig. 6. The enrichment is determined by the ratio of the $^{235}\text{U}$ and $^{238}\text{U}$ peaks located between 92 and 94 keV. In this case, the sample was 10% enriched uranium. The same sample was analyzed by the 5x5x5 mm CZT detector system and the resulting GRPANL data is shown in Fig. 7. The resolution of the CZT system is markedly worse (2.9 keV for CZT and 484 eV for the LEPS detector), but the software was still able to fit the peaks accurately enough to give the same result. The CZT system was then used to analyze several uranium samples of differing enrichments. This data is shown in Table 1. The amount of time necessary for spectral acquisition was extremely long due to the fact that 1 gram samples were used. A commercial instrument using this technology would certainly benefit from larger detectors resulting in better efficiency. The enrichment analysis required a detection system resolution of <3% at 100 keV to be able to adequately discriminate between peaks that were closely spaced in terms of energy.

B. X-ray Fluorescence

The CZT detection system was used to characterize lead by X-ray fluorescence. A lead sample was illuminated by a $^{57}\text{Co}$ source. Fig. 8 shows the resulting spectrum and verifies the ability of this system to be used as an environmental sample screening tool. Energy resolution is 2.8 keV, FWHM -- typically that obtained for $^{241}\text{Am}$ (60 keV). A desirable feature of this technology, with respect to heavy metal X-rays, is that 100% absorption takes place in relatively small volume detectors.

V. CONCLUSIONS

We have successfully shown that ambient temperature radiometric assaying can be performed with current technology. A truly portable instrument has been developed to meet the needs of several important applications. Immediate applications include Safeguards and Environmental Management, illustrated by the successful demonstration of this instrument for actinide and heavy metal characterization.

During the course of this work, it was determined that a standard method was needed for characterization of CdTe and CZT detectors. We have addressed this by developing a proposed standard based on the IEEE Standard Test Procedures for Semiconductor X-ray Energy Spectrometers (ANSI/IEEE Std. 759-1984).

While performing detector characterizations, it was noted that charge trapping appears to be the dominant factor in resolution (FWHM) degradation. This is even true in detectors that have significant shot noise as a result of high leakage currents (≥40 nA). This data supports the need for more research to better understand charge transport mechanisms in this material. In addition, signal processing techniques will be applied in order to address the charge collection problems associated with these ambient temperature detectors.

VI. REFERENCES