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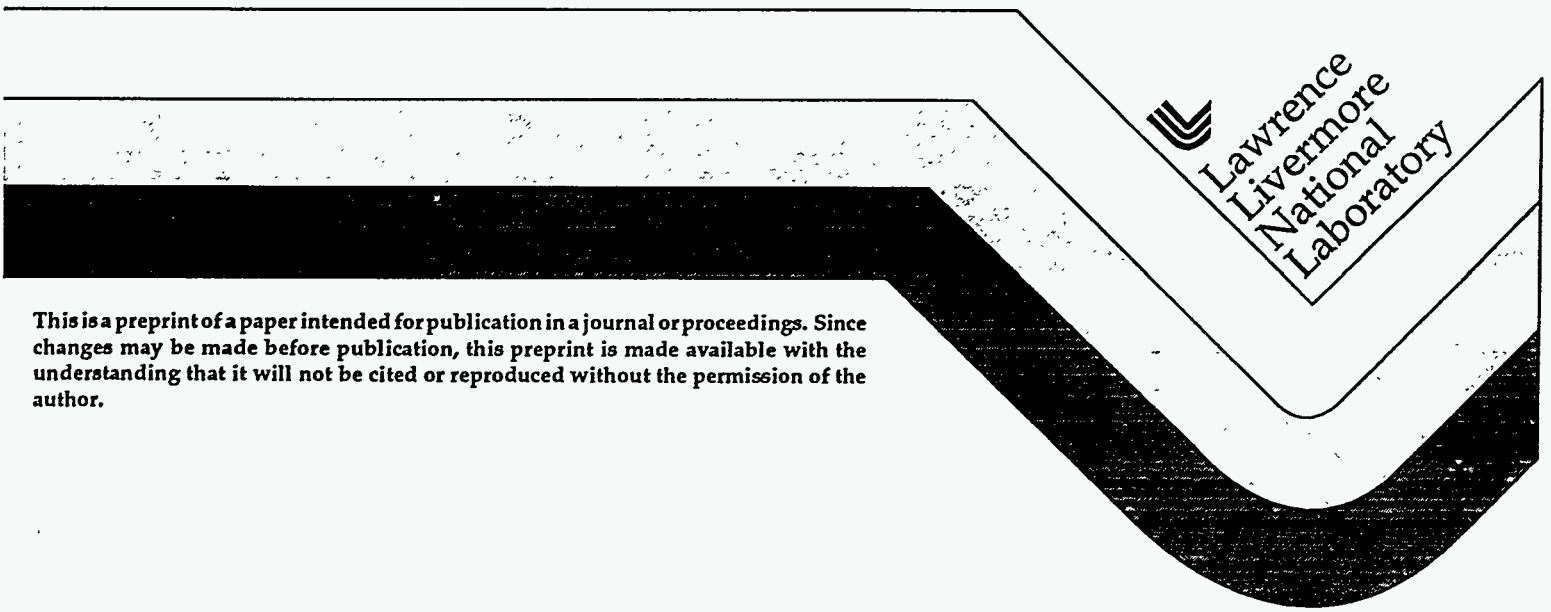

A PORTABLE MEDIUM-RESOLUTION GAMMA-RAY SPECTROMETER AND ANALYSIS SOFTWARE

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Abstract

There is a strong need for portable radiometric instrumentation that can both accurately confirm the presence of nuclear materials and allow isotopic analysis of radionuclides in the field. To fulfill this need the Safeguards Technology Program at the Lawrence Livermore National Laboratory, in collaboration with EG&G ORTEC, has developed a hand-held, non-cryogenic, low-power gamma- and x-ray measurement and analysis instrument that can both search for and then accurately verify the presence of nuclear materials. We will report on the use of cadmium zinc telluride (CZT) detectors, detector electronics, and the new field-portable instrument being developed. We will also describe the isotopic analysis that allows enrichment measurements to be made accurately in the field. These systems provide capability for safeguards inspection and verification applications and could find application in counter-smuggling operations.

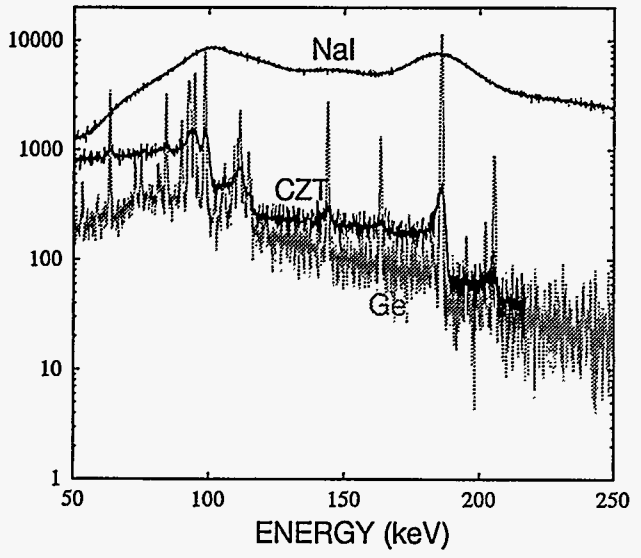

Figure 1. Comparison of Low, Medium and High Resolution Gamma Spectra

INTRODUCTION

BACKGROUND INFORMATION

The development of this system began in 1992 prompted by the recognized need for an analytical field instrument with stable performance between that of High Purity Germanium (HPGe) and Sodium Iodide (NaI) based systems. A significant enabling development was the introduction of large volume (≈125 mm^3), moderate resolution (<3% at 122 keV) Cadmium Zinc Telluride (CZT) detector crystals. While these volumes are relatively small in comparison to typical HPGe and NaI detectors, CZT detectors operate at ambient temperature eliminating the requirement for liquid nitrogen (LN) cooling. In addition, these detectors can provide spectra with resolution adequate to perform isotopic analysis of nuclear materials. Earlier CZT and Cadmium Telluride (CdTe) detectors also provided adequate resolution though were of such small volumes (≈8 mm^3) that the relative detection efficiency was extremely low. Long acquisition times resulting from the low efficiency of these detectors made them unsuitable for portable instrumentation in most applications.

EARLY DETECTOR RESEARCH

The first efforts to develop this instrument were focused on the determination of whether CZT and CdTe detectors could perform well enough to allow for isotopic analysis of the spectra using conventional means GRPANL[1]. GRPANL analyzed the 100 keV energy region to determine uranium enrichment, therefore ^{57}Co was chosen as the characterization energy source. Peak shape was the primary characteristic studied. The test configuration consisted of two detectors, one each of CZT, 20% zinc (molecular), and CdTe, in BNC mountings, an ORTEC 142PC preamplifier, and an ORTEC 572 shaping amplifier (1 μs shaping). The spectral resolutions were characterized by measuring the full width at half maximum (FWHM) of each peak after background subtraction [2]. This method of determining the FWHM takes into account the effects of low energy tailing resulting from charge collection problem associated with the detector. Cobalt spectra with resolutions of less than 3% were easily obtained using both materials with small detector volume geometries (2x2x2 mm, planar). Initial analysis results of 10% enriched uranium samples indicated that spectra with resolutions of up to 3% could be analyzed with a reasonable degree of confidence.

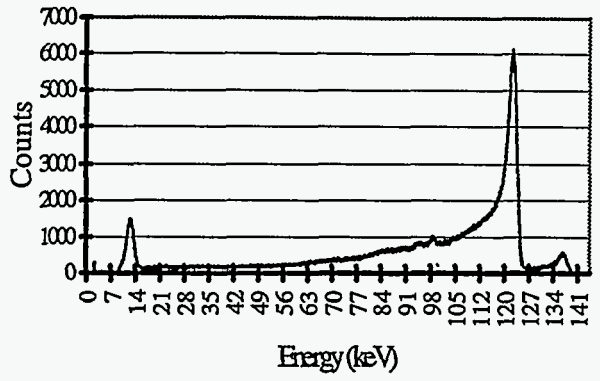

Figure 2 ^{57}Co Spectrum Using a 5x5x5 mm CZT Detector

During these tests, several operational differences were noted between the two detector types. The most significant difference was the bias voltage applied to the detector. The CdTe detector required +120 VDC while the CZT detector operated with +500 VDC. The reason for this difference is the resistivity of the materials. CdTe has a resistivity of up to 10^9 Ω • cm[3,4] while CZT is typically about 10^{11} Ω • cm, depending on the zinc content[5]. The benefit of a higher bias voltage is a stronger electric field aiding charge collection resulting in shorter charge collection times. In addition, the higher resistivity of CZT allowed for the development of larger volume detectors addressing efficiency issues and higher energy applications for this family of detectors.

Tests to compare resolution of several detectors indicated that larger volume CZT detectors performed comparable to, and many times better than, smaller volume CdTe detectors (FWHM results at 122 keV). More importantly, when the spectra are compared it appears that the CdTe spectrum has an elevated and constant background while the CZT

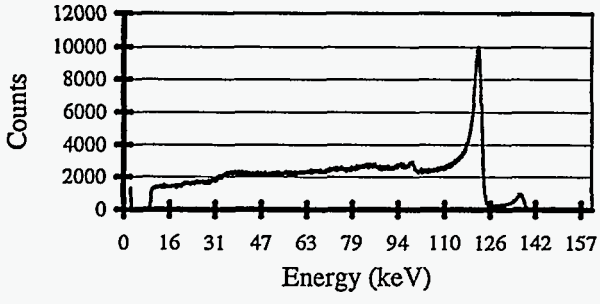

Figure 3 ^{57}Co Spectrum Using a 2x2x2 mm CdTe Detector

spectrum has an increasing background with energy. This could be the result of better charge collection related to the higher bias voltages in CZT. Sample spectra from a 5x5x5 mm CZT detector and a 2x2x2 mm CdTe detector are shown in Figures 2 and 3, respectively. The difference in background, as well as the characteristic low energy tail, can easily be seen.

While the early success in the computerized analysis of spectra was encouraging, several disadvantages that affected the analysis procedures were discovered when using these materials. The analysis software compares peak heights from each uranium isotope. In uranium samples of very low or very high enrichment, the spectral lines from the low abundance isotope can be lost in the background. Solving this problem requires extremely long acquisition times to obtain sufficient counting statistics to differentiate the low abundance peak from the background. To realize a portable instrument that would be able to acquire useful spectra in a reasonable amount of time was recognized as a potential limitation.

Another immediate concern resulting from the early experiences was a significant low energy tail, characteristic of high Z, ambient temperature detector materials resulting from low carrier mobility and charge trapping[6]. The analysis software includes a peak fitting routine that was optimized for relatively Gaussian peak shapes such as those from germanium detectors. The tailing phenomenon associated with CZT or CdTe detectors reduces the accuracy of the analysis due to difficulty fitting the peaks.

A number of tests were conducted to determine the reason for the low energy tail. Though not conclusive, it appears that the main cause of the tail is due to charge trapping rather than shot noise. By varying the shaping time, the contribution of shot noise to the signal can be varied. Examining spectra from various shaping times from 0.25 to 4 μs indicates a corresponding change in resolution accompanied by a relatively constant low energy tail.

Since the low energy tail appeared to be a material artifact, many techniques currently being evaluated in other applications[7,8,9,10] were attempted to correct for the effects on resolution. Time variant shaping was evaluated in an effort to tailor the integration time to the varying charge collection time. The result was reduced resolution due to an increase in baseline noise being integrated into the signal. Pulse shape and rise time discrimination were also used with the undesirable result of greatly diminished efficiency. Such a large percentage of the pulses were discarded (up to 90%) that the already marginal efficiency of the detectors became completely unsatisfactory. Finally, charge loss correction was evaluated and showed no appreciable benefit. This technique is useful when applied to a cooled system with improved charge collection compared to ambient temperature operation.

SYSTEM DEVELOPMENT

HARDWARE

The prototype system was developed as a completely portable instrument. It consisted of a laptop computer containing both the system operating software and an uranium enrichment analysis program, a portable multichannel analyzer (MCA), a NaI detector used for search mode operation, and a CZT detector probe used for verification and analysis mode operation. The initial research performed on the characteristics of CdTe and CZT detectors precipitated the definition of the system that could be realized:

- CZT was chosen due to larger available detector volumes.
- Initial uranium enrichment computer code analysis indicated that a detector resolution of <3% at 122keV was necessary for reasonable results.
- During the resolution and low energy tail experiments an optimum shaping time of 0.5 μs was determined.
- The system was to be portable, therefore no cooling would be implemented due to power consumption requirements.
- Detection efficiency was to be maximized, therefore signal processing techniques that discarded pulses would not be implemented.

EG&G ORTEC would be responsible for the development of the portable MCA including operational software and LLNL would develop the CZT detector probe and uranium enrichment analysis software.

MULTICHANNEL ANALYZER

The MCA developed especially for this application by EG&G ORTEC is named the MicroNOMAD and is a 1.5-lb., battery powered, portable gamma-ray spectrometer. The block diagram is shown in Figure 4. The unit consists of an amplifier suitable for NaI (bypassed for CZT operation), 2k ADC, spectrum stabilizer, microprocessor and memories, as well as parallel, RS-232, and RS-485 ports. 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 alkaline 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

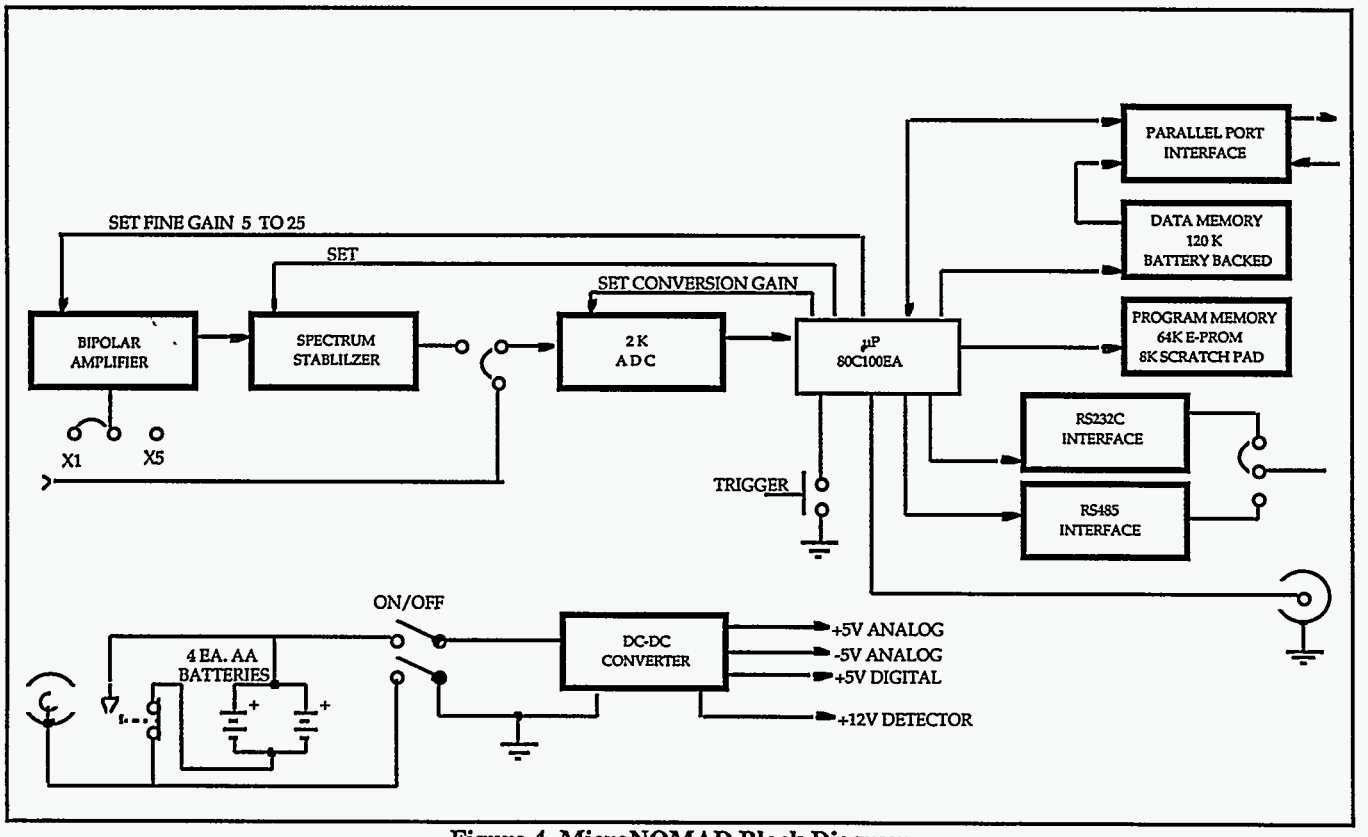

Figure 4 MicroNOMAD Block Diagram

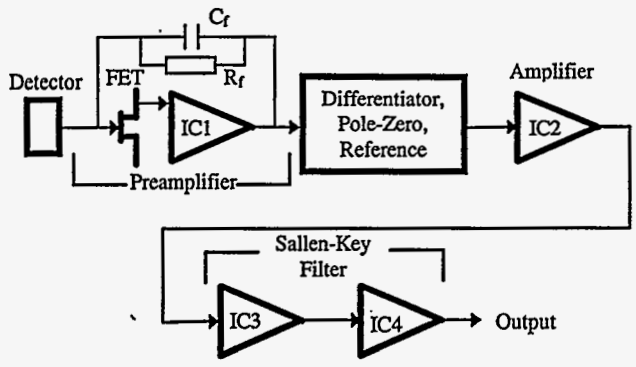

Figure 5 Block Diagram of the Prototype CZT Detector Probe

with the preset values of real time, live time, gross region-of-interest (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 PC-compatible 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

Even with improved CZT material, noise (primarily shot noise), poor hole-mobility, and charge trapping contribute to less than ideal energy resolution. The preamplifier and filter amplifier were 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 Figure 5. 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 Figure 2. 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 Figure 5) 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 power 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 power 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 characterization 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.

ANALYSIS SOFTWARE

The main driving factors in implementing isotopic analysis software for CZT detectors are resolution, efficiency, peak shape, and peak-to-valley ratio. The state of the art for CdTe and CZT detectors and electronics is changing rapidly. The envelope of what analyses are possible is expanding with the detector development. We have had a prototype 'MGA-style' uranium analysis code for almost two years. Resolution and efficiency of detectors at that time were not sufficient to consider plutonium analysis. Based on capabilities and anticipated capabilities of the newest CZT detectors, we are developing

plutonium analysis software. Figure 6 highlights significantly improved peak-to-valley in new detectors being developed by Digi-Rad. When the resolution of these new detectors is comparable to the current state-of-the-art, improved analysis with shorter collection times will be possible.

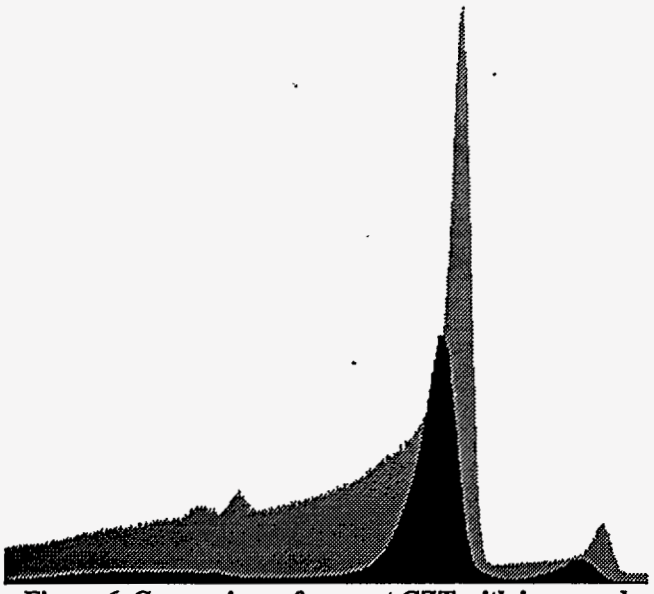

Figure 6 Comparison of current CZT with improved peak-to-valley CZT for 57Co

URANIUM ENRICHMENT ANALYSIS

We developed a prototype uranium enrichment code: CZTU. It is an adaptation of methods used for analysis of HPGe spectra, performing an MGA-style analysis in the 100 keV region. Typical count times run from 5 to 30 minutes. CZTU is now being modified to make use of experience gained in the development of our U235 code.

Work on the CZTU code includes adapting a different "more accurate" background determination in the U235 code to CZT spectra. CZT peaks are enough different than the Ge peaks that new background algorithms and new criteria are being refined in the CZTU code.

Other changes include using a more complete set of peaks in the $^{235}U/^{238}U$ determination; currently 13 peaks instead of 10.

The current prototype achieves 10% uncertainties for short counts over a wide range of enrichments. At high enrichments (>75%) the current code can have higher uncertainties due to a very small ^{238}U signal. This is being addressed currently in software development and it is anticipated that detectors with improved peak-to-valley will also alleviate this problem.

PLUTONIUM ISOTOPIC ANALYSIS

The 100 KeV region of plutonium is too complex to attempt an MGA style analysis with medium-resolution CZT detectors. We are developing a code that performs analysis primarily in the 120-210 keV region. The purpose of our plutonium code is to confirm the presence and isotopics of plutonium in a short time. The code will analyze for $^{238,239,240,241}Pu$ and ^{241}Am. It will also analyze for $^{235,238}U$ in the presence of Pu. We are looking at modifying and adapting the IAEA Blue-Box codeWe have not characterized the performance of the code over a range of samples, but the design goal is 10% uncertainties in major isotopics.

HAND-HELD SCREENING INSTRUMENT FOR COUNTER-SMUGGLING APPLICATIONS

There is a strong need for portable radiometric instrumentation that can accurately confirm the presence or absence of special nuclear materials and verify the presence industrial or medical radionuclides. The instrumentation must be usable by customs inspectors or others not familiar with radiation measurement equipment. Figure 7 shows a prototype of an instrument under development at LLNL using a CZT detector system, MicroNOMAD MCA, and a hand-held computer.

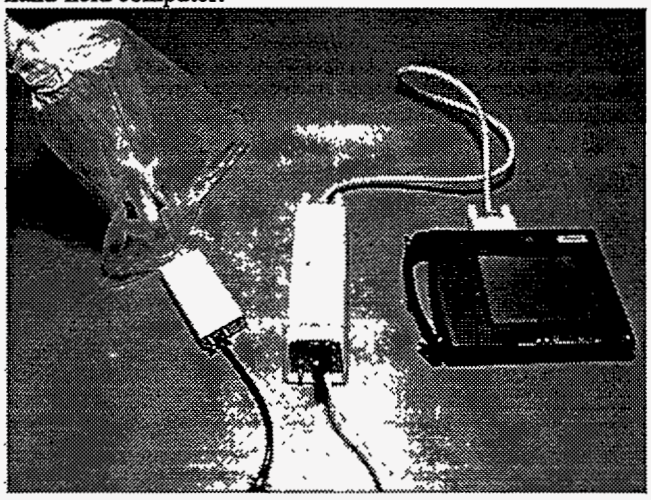

Figure 7 Prototype CZT-based Radiation Screening Instrument

A very simple graphical user interface has been implemented on a touch screen with data collection and analysis software that can run with little more than single button push. Uranium and plutonium analysis are performed on data collected from 0-210 keV and screening for medical and industrial isotopes is performed on data collected from 0-1000 keV. Data will typically be collected for 5 minutes.

A report indicating the presence or absence of various isotopes is displayed on the screen. Since the MCA/detector used for this prototype does not have software programmable gain and zero, actinide and medical/industrial screening require different setups.

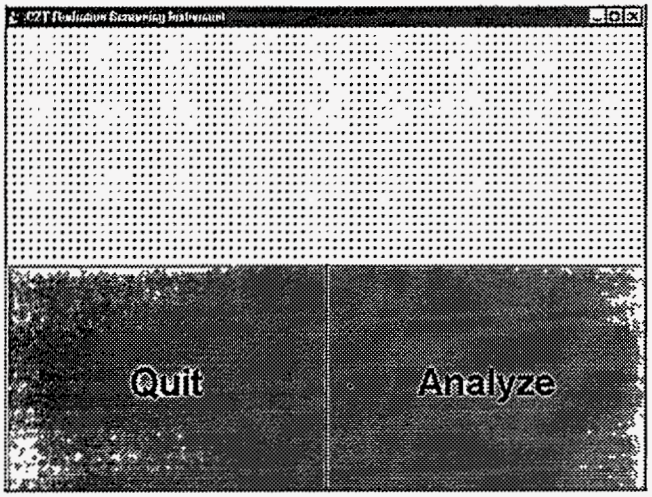

Figure 8 Example Screen from CZT Radiation Screening Instrument

We anticipate that fielded units will consist of a single hand-held box with a small LCD display.

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AUSPICES

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