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Advances in CdZnTe Detectors for Safeguards

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ABSTRACT

In this paper, we will describe recent developments in CdZnTe detector technology, including methods to improve the performance of detectors at high ambient temperature, miniature electronics for spectroscopy, and analytical software for portable detectors. A large-volume, portable detection probe consisting of an array of coplanar grid detectors is demonstrated. The probe is shown to have similar efficiency to NaI(Tl) detectors used for holdup measurements.

I. INTRODUCTION

CdZnTe is a compound semiconductor material that is being developed for portable gamma-ray spectroscopy. The high resistivity of CdZnTe enables it to be used for radiation detection over a wide range of ambient temperatures (-20°C to 40°C). In addition, the pulse height resolution that can be achieved by CdZnTe detectors is typically much better than the resolution of scintillation detectors. Consequently, CdZnTe is being implemented for applications in which the use of intrinsic germanium is cumbersome and for which the resolution of NaI(Tl) is inadequate. These applications include nuclear material holdup measurements, *in situ* assay of nuclear materials with unknown and variable contaminants, spent fuel assay, unattended monitoring, customs inspections, and nuclear material search and identification.

CdZnTe is grown commercially by the Bridgman-Stockbarger method. The principal US supplier (eV Products) primarily uses high-pressure vertical Bridgman (HPVB) furnaces to grow the material. Other variants of the Bridgman method are being explored. However, HPVB is presently the main source of material and is capable of reliably producing high-resistivity material with electron transport properties needed for spectroscopy.

The main limitation of CdZnTe technology is the size of single crystals that can be routinely harvested from ingots grown by HPVB. Grain boundaries are known to trap electrons and, if included within a detector, can reduce the full-energy (photo-peak) efficiency and degrade pulse height resolution. Consequently, single crystal material is desired for gamma-ray spectroscopy. At present, the largest single crystals manufactured for gamma-ray spectrometers from HPVB material are on the order of 15 mm × 15 mm × 7.5 mm. This is adequate for many applications; however, for *in situ* assay and nuclear material holdup, there are many cases in which higher efficiency is needed.

General attributes of HPVB material are contrasted with intrinsic silicon in Table 1. Because the mobility-lifetime products are quite large, practical silicon devices operate in a "sweep-out" mode, in which both types of carriers are fully collected. In this mode of operation, the charge measured

by the preamplifier is insensitive to the position of the gamma-ray interaction, and accurate measurements of the charge produced by radiation interactions are possible. However, for CdZnTe, the product of hole mobility and lifetime is quite small ($<1\times10^{-5}$ cm^2/V). So, under typical bias conditions (100 V/mm), the trapping length for holes is much less than 1 mm. Consequently, devices that operate in sweep out mode must be quite thin. Practical devices are usually thicker than 1 mm and have poor peak shape due to low-energy tailing caused by hole trapping.

Table 1. *Attributes of spectrometer-grade HPVB CdZnTe are contrasted with intrinsic silicon. The table includes parameters given by Knoll [1] and McGregor [2].*

Parameter	**$Cd_{1-x}Zn_xTe$, x=0.1 T=295°K**	**Intrinsic Silicon T=295°K**
Density (g/cm^3)	5.8	2.33
Effective atomic number	49	14
Dielectric constant	10.9	12
Band gap (eV)	1.572	1.115
Energy per electron-hole pair (eV)	5.10	3.62
Fano Factor	<0.1	~0.08
Resistivity (Ω-cm)	$>3\times10^{10}$	2.3×10^5
Electron mobility (cm^2/V-s)	1200	1350
Hole mobility (cm^2/V-s)	100	480
Electron lifetime (μs)	5	>100
Hole lifetime (μs)	<0.1	>100

Electron sensing devices have been developed to reduce or eliminate low-energy tailing caused by the trapping of holes. These devices are designed to reduce the sensitivity of the anode to charge motion within the volume of the detector and to increase sensitivity near the anode. The goal is to mimic the function of a Frisch grid. Only the electrons are able to reach the sensitive region near the anode and contribute to the charge pulse. Because electron trapping is relatively small, the magnitude of the charge pulse is independent of the origin of the electrons (a necessary condition for high-quality spectroscopy).

For example, quasi-hemispheric detectors use the "small pixel effect" to reduce the sensitivity of the anode to the motion of charge within the volume of the detector.[3,4] In addition, the cathode is extended up the sides of the detector to produce an electric field that always points towards the anode. Because electrons always take the shortest path to the anode, variations in the measured charge from electron trapping are minimized.

Hemispheric detectors have significantly better peak shape than planar detectors and can be used for spectroscopy below 1 MeV. An inexpensive variation on the hemispheric detector (called the Capture detector) was developed by eV Products and is being evaluated for safeguards applications.[5] Coplanar grid detectors, which use a different approach for electron sensing that is described in detail elsewhere,[6] provide improved performance for spectroscopy.

The benefits of electron sensing detectors include increased size and detection efficiency along with better resolution and peak shape. The penalty is increased cost, and in the case of coplanar grids, increased complexity for pulse-shaping electronics. Nonetheless, coplanar grid detectors and hemispheric detectors are the instruments of choice for *in situ* assays of nuclear material. Witness, for example, the successful application of coplanar grid detectors for uranium holdup measurements at Y-12.[7]

To illustrate the extremes in detector size, performance, and cost, measurements of ^{57}Co (122 and 136 keV) and ^{137}Cs (662 keV) made using a small detector with planar electrodes are contrasted with measurements made using a coplanar grid detector (Fig. 1). The planar device was 3 mm × 3 mm × 2 mm in size and cost 20 US dollars. The coplanar grid detector was a cylindrical device developed by LANL and manufactured by eV Products.[8] The detector had a diameter of 10 mm and was 5 mm thick. This detector cost 5000 US dollars. The planar detector has poor peak shape at 662 keV and is generally not suitable for quantitative measurements at high energy (e.g., plutonium holdup measurements or spent fuel assay); however, at low energy, the peak shape is adequate for many applications. In applications such as remote monitoring, where many detectors are needed, the low cost, high detection efficiency, and limited capability for spectroscopy provided by planar CdZnTe detectors may warrant their selection over other types of detectors (e.g., silicon diodes).

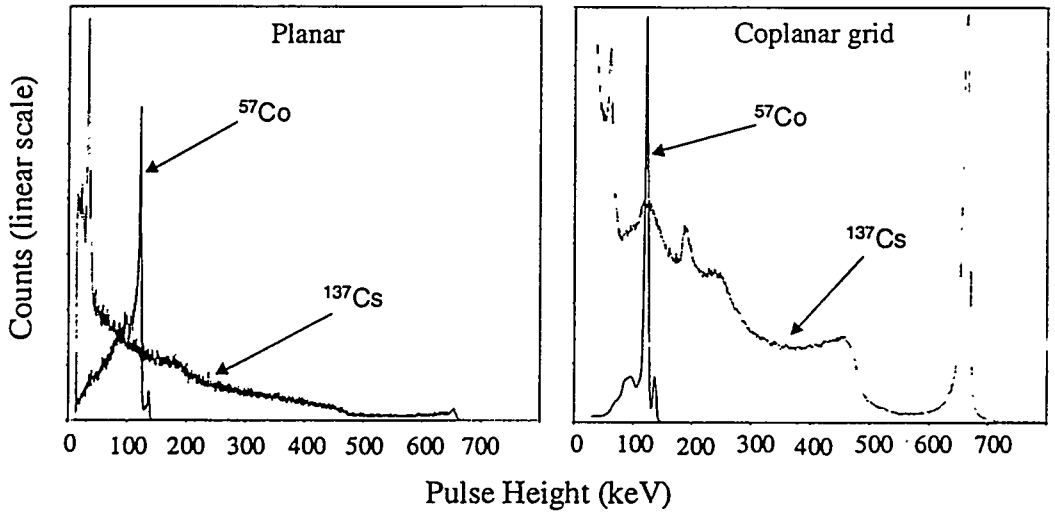

Fig.1. *Performance of a small planar CdZnTe detector is contrasted with a cylindrical coplanar grid detector for the measurement of ^{57}Co and ^{137}Cs.*

II. PERFORMANCE AT ELEVATED TEMPERATURES

CdZnTe detectors are usually selected such that they function effectively at room temperature (e.g., 25°C). What happens when a detector is operated at elevated ambient temperature? There are many situations relevant to safeguards in which the ambient temperature could reach 40°C or higher. As temperature increases, the conductivity of CdZnTe increases. With increased leakage current, it becomes more difficult to measure the minute bursts of charge produced by radiation interactions.

For low-energy measurements (e.g., uranium holdup), the increase in noise with temperature can result in unacceptable pulse height resolution.

With HPVB material, the Fermi level is pinned near the middle of the band gap. This is probably caused by deep donor levels that are present in concentrations greater than the difference between the concentration of shallow donor levels introduced by impurities and the concentration of acceptor levels associated with vacancies and complexes. Consequently, free carrier concentrations approach the minimum theoretical values, despite the fact that the material has a high concentration of impurities.[9,10]

The conductivity of a semiconductor with the Fermi level at the mid-gap is proportional to $\exp(-E_g/2kT)$, where E_g is the bandgap (with units of meV), k is Boltzman's constant (0.086 meV/°K) and T is the temperature (°K). So, a temperature change of 15°C will cause the conductivity to increase by a factor of four. This will cause roughly a factor of two increase in the noise associated with leakage current (parallel noise). Note that the variation predicted by this simple model for conductivity has been confirmed experimentally.[11]

To demonstrate the effect of temperature on detector performance, we acquired low-energy gamma-ray spectra using a cylindrical coplanar grid detector. The source was a combination of ^{109}Cd (88 keV) and ^{57}Co (122 and 136 keV). The experiment was carried out at two ambient temperatures (25°C and 40°C). The results are shown in Fig. 2. The settings used in the experiment were those found to be optimal at room temperature (e.g., the amplifier shaping time was 0.5 μs, the bulk bias was -750V, and the grid bias was -30V). No adjustments were made when the temperature was changed.

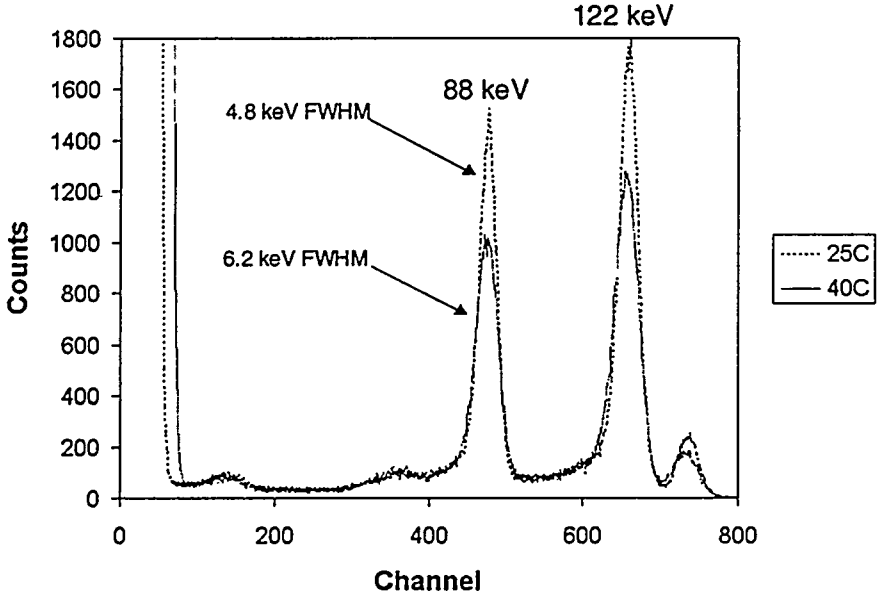

Fig. 2. *Pulse height spectra acquired at two ambient temperatures with a cylindrical coplanar grid detector. The sources were ^{109}Cd (88 keV) and ^{57}Co (122 and 136 keV).*

The change in temperature causes a noticeable loss in resolution and an increase in the noise floor. The change in the full width at half maximum (FWHM) of the 88-keV peak was ~30%, not the factor of two that might have been expected. Note that the performance of this detector at 40°C is still sufficient for uranium holdup measurements. However, at room temperature, most detectors used in the field have a FWHM approaching 8 keV for 122 keV gamma rays. Because their performance is marginal at room temperature, these detectors do not perform adequately at 40°C.

To determine the sources of noise in the experiment, we mapped the noise characteristics of the noncollecting channel of the coplanar grid detector using a precision pulser and an amplifier with a wide range of shaping times. A review of noise sources can be found in Kowalski.[12] The results are shown for two temperatures in Fig. 3, in which the equivalent noise charge was plotted against the amplifier shaping time. Note that the parallel noise, which is associated with leakage current, varies as $\sqrt{\tau}$ where τ is the shaping time. At large shaping times, noise increases by a factor of 1.6 when the temperature is changed from 25°C to 40°C, which is consistent with the predicted change in leakage current. At small shaping times, serial noise, which varies as $1/\sqrt{\tau}$, is dominant. Serial noise does not appear to be affected significantly by temperature. In addition, the flicker noise (1/f noise) appears to be small.

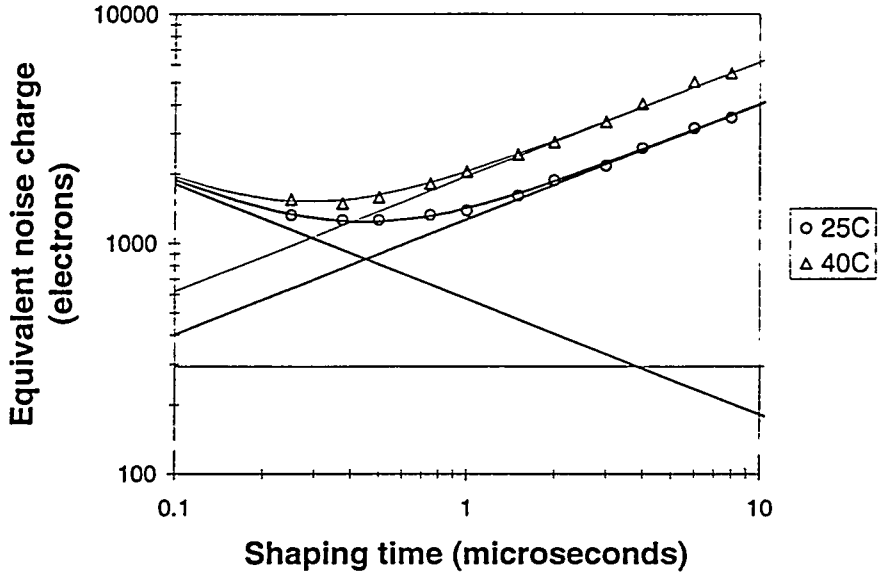

Fig. 3. *Noise characteristics of a cylindrical coplanar grid detector at two temperatures. Dashed lines indicate serial-, parallel-, and flicker-noise.*

The results of this experiment show that short shaping times are desired to minimize the noise at elevated temperature. At 0.5 μs, the increase in noise was only 26%. At 0.25 μs, the increase in noise was only 13%. It seems reasonable to select the shaping time to correspond to the noise corner for the highest expected temperature. The minimum shaping time that can be used is limited by ballistic deficit. However, in electron sensing devices, it should be possible to select the shaping time to be less than the maximum electron drift time, since the electrons spend only a small portion of the drift time in the charge-sensing region near the anode. More research is needed to determine

how user-adjustable parameters such as shaping time affect the performance of coplanar grid detectors at different temperatures.

One obstacle for implementing this approach in the field is that commercially available spectroscopy systems have a limited selection (one or two) of shaping times. In cases where the shaping time cannot be adjusted, there is nothing that can be done to minimize noise short of cooling the detector. We have investigated several options for cooling detectors using thermoelectric coolers and have designed a detector module that can maintain a 15°C temperature difference while consuming less than 200 mW. The module is presently being tested and will be available from the vendor shortly.

III. HIGH EFFICIENCY CdZnTe DETECTORS

As pointed out in the introduction, there is a limitation on the size of single crystals that can currently be manufactured. However, there are some applications, particularly *in situ* assay (e.g., holdup), where detectors larger than the single crystal limit are needed. To achieve high efficiency, we have developed methods to combine multiple single crystals in a low-power, hand-held probe that is compatible with off-the-shelf, portable multichannel analyzers (MCAs). The approach we have taken to develop multi-element detectors is described in detail elsewhere.[13] A summary of our work is presented here.

We have demonstrated a multi-element detector, consisting of an array of eight 10 mm × 10 mm × 5 mm coplanar grid detectors. The array was mounted in a hand-held probe that contained all pulse shaping electronics needed to provide a single spectroscopy output to the analog-to-digital (ADC) converter in the MCA. Bias and low-voltage power was supplied by the MCA. An internal divider circuit was used to provide differential bias to the detectors. Signals from the individual detectors were combined after the amplifier stage where the fine gain adjustments needed to match the output of the detectors could be made. Gated signal combination was used to avoid the introduction of noise from channels that did not receive charge from the radiation interaction. Only channels with signals that exceeded a noise threshold are combined, thus preserving the resolution at low energy.

The performance of the hand-held probe for the measurement of low-burnup plutonium is contrasted in Fig. 4 with two NaI(Tl) detectors commonly used for holdup measurements and a large single crystal, coplanar grid CdZnTe detector. The efficiency of the eight element detector was higher than the 1 in.×0.5 in. NaI(Tl) detector at all energies, and was higher than the 1 in. ×2 in. NaI(Tl) detector at low energy. The pulse height resolution of the eight-element detector was at least a factor of three better than the NaI(Tl) detectors over the entire energy range.

IV. MINIATURIZATION

The hand-held, multi-element detector was based on standard printed circuit board (PCB) technology with surface mount components. To achieve low power, we used rail-to-rail output designs for all stages of the shaping circuit. In addition, we used low-voltage components (±5V). This resulted in an eight-channel detector than consumed less than 1W. Future designs will be developed with ±3V components. We expect to commercialize a four-channel system that

consumes on the order of 300 mW in the near future. Such a system will meet cost, efficiency, and performance requirements for a wide range of safeguards applications.

At the same time, we are working to apply the lessons learned from the multi-element detector to develop miniature single crystal detectors. The use of low-voltage surface mount components with PCB technology allows miniature detectors to be manufactured with minimal cost for design (e.g., compared to application-specific integrated circuits). For example, we have recently designed a CdZnTe spectrometer around a 3.6V lithium-ion battery. The electronics package fits snugly around the battery and the entire detector is roughly 2.5 inches in length by 1 inch in diameter. The detector contains a high-voltage bias supply that can deliver up to 1000V to the detector, a low-voltage power supply, a charge sensitive preamplifier, an amplifier, an integral discriminator, and an infrared transmitter. The detector can operate unattended for ~200 h.

Using this design, we have demonstrated that spectroscopy can be carried out when the power supply is in close proximity to the shaping electronics. Future versions will accommodate coplanar grid detectors and will include an ADC for the analysis of multiple gamma rays. The development of low-cost, miniature spectroscopy systems will reduce the amount of equipment needed in safeguards operations (especially holdup measurements and *in situ* assay) and will enable CdZnTe detectors to be used in locations that are difficult to access.

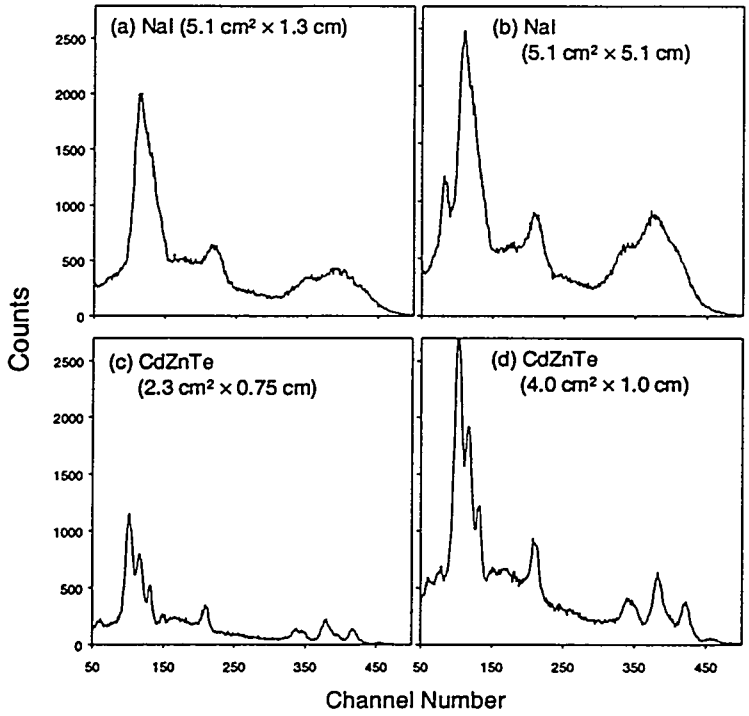

Fig. 4. *Pulse height spectra acquired under identical conditions (same count time, measurement geometry, high-Z filters, etc...) by different detectors for a low-burnup plutonium sample. The detectors were: (a) 1 in. ×0.5 in. NaI(Tl); (b) 1 in. ×2 in. NaI(Tl) detector; (c) 15 mm ×15 mm ×7.5 mm coplanar grid CdZnTe detector; and (d) eight-element CdZnTe detector. Dimensions (area and thickness) of the detectors are given in the figure.*

V. SPECTRUM ANALYSIS SOFTWARE

We are investigating a variety of methods to analyze pulse height spectra measured by CdZnTe detectors. For example, we have developed full-spectrum and regional analysis methods for coplanar grid CdZnTe detectors.[14,15,16] Our goal is to develop analytical tools for peak identification, determination of peak areas (including the analysis of multiplets), and the determination of isotopic abundance. To facilitate the implementation of these methods in the field, we have developed the Spectrum Analysis Shell (SASH).

SASH is a Microsoft Windows™-based software package written in C++ that can acquire, display, and analyze pulse height spectra. SASH runs on notebook personal computers and can acquire data in a variety of formats from different portable MCAs. The graphical user interface for SASH is shown in Fig. 5. SASH is easily configured for different analytical tasks, including isotopic analysis, enrichment measurements, and holdup measurements. A commercial version of SASH is scheduled for release in 2002.

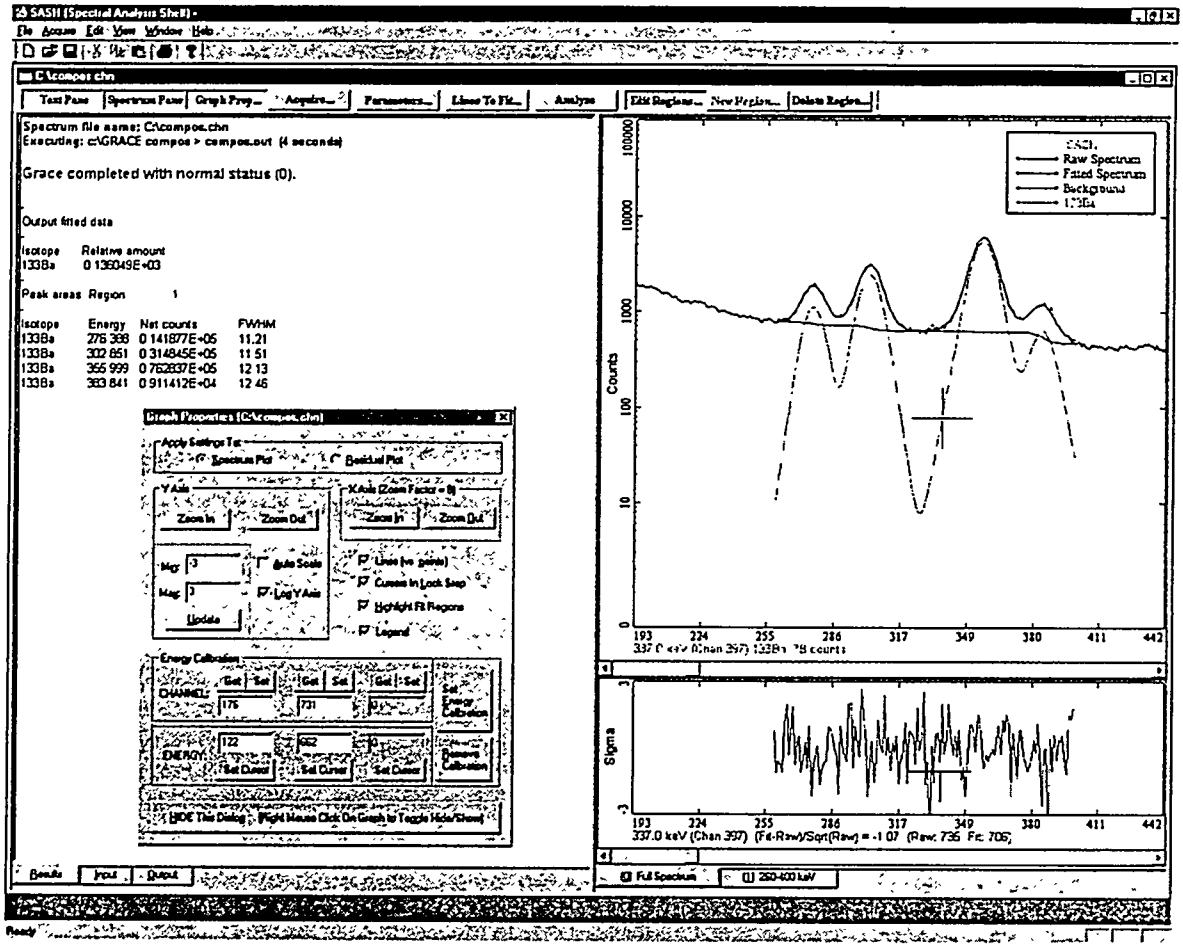

Fig. 5. *The SASH workspace is shown. SASH provides a graphical display of data as well as fitted and residual spectra and a report summarizing the results of the analysis. A menu for adjusting parameters of the graphical display is superimposed on the report.*

VI. SUMMARY

CdZnTe is being implemented by domestic and international safeguards organizations for applications ranging from spent fuel attribute testing to *in situ* nondestructive assay. The main limitation of the technology is detection efficiency, which is constrained by the size of single crystals that can be manufactured routinely. We have solved this problem by developing low-power, hand-held detection probes that can combine multiple single crystals to produce a detector that has similar efficiency to NaI(Tl) detectors used for holdup measurements. We have extended this technology to develop miniature detection probes that can be powered by a small battery. Miniaturization of the spectroscopy electronics will reduce the amount of equipment used in holdup campaigns and will enable the use of CdZnTe in locations that are difficult to access (e.g., monitoring of processing equipment in glove boxes). Software is being developed for the analysis of spectra acquired by CdZnTe detectors that will run on portable computers used in the field. We are also investigating the effect of temperature on CdZnTe performance. Selection of shorter amplifier shaping times is desired to minimize the change in pulse height resolution with temperature. The use of low-power thermoelectric coolers may be required in cases where the low-energy performance is marginal at room temperature.

ACKNOWLEDGMENT

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